Surface Science Division
Fachverband Oberflächenphysik (O)

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Overview of Invited Talks and Sessions

(Lecture halls H2, H3, H4, H6, S051, S052, S053, and S054; Poster P3 and P4)

Invited Talks

O 1.1	Mon	9:30–10:15	S054	**Laser-excited electrons: how hot are they?** — •Baerbel Rethfeld
O 24.1	Tue	9:30–10:15	S054	**Oxygen Evolution on Rutile Ruthenium and Iridium Dioxides** — •Yang Shao-Horn
O 36.1	Wed	9:30–10:15	S054	**Heterogeneous chemistry of liquid-vapor interfaces investigated with X-ray photoelectron spectroscopy** — •Hendrik Bluhm
O 60.1	Thu	9:30–10:15	S054	**Exciting states in atomically thin layers** — •Thorsten Deilmann
O 77.1	Fri	9:30–10:15	S054	**Sub-molecular fluorescence microscopy with STM** — •Guillaume Schull
O 84.1	Fri	13:15–14:00	S054	**Exploring the Mysteries of Topology in Quantum Materials** — •Claus M. Schneider

Invited Talks of the joint Symposium Frontiers of Orbital Physics: Statics, Dynamics, and Transport of Orbital Angular Momentum (SYOP)

See SYOP for the full program of the symposium.

SYOP 1.1	Mon	9:30–10:00	H1	**Orbital degeneracy in transition metal compounds: Jahn-Teller effect, spin-orbit coupling and quantum effects** — •Daniel Khomskii
SYOP 1.2	Mon	10:00–10:30	H1	**Orbital magnetism out of equilibrium: driving orbital motion with fluctuations, fields and currents** — •Yuriy Mokrousov
SYOP 1.3	Mon	10:30–11:00	H1	**Orbitronics: new torques and magnetoresistance effects** — •Mathias Kläui
SYOP 1.4	Mon	11:15–11:45	H1	**Orbital and total angular momenta dichroism of the THz vortex beams at the antiferromagnetic resonances** — •Andrei Sirenko
SYOP 1.5	Mon	11:45–12:15	H1	**Observation of the orbital Hall effect in a light metal Ti** — •Gyung-Min Choi

Invited Talks of the joint Symposium SKM Dissertation Prize 2022 (SYSD)

See SYSD for the full program of the symposium.

SYSD 1.1	Mon	10:15–10:45	H2	**Charge localisation in halide perovskites from bulk to nano for efficient optoelectronic applications** — •Sascha Feldmann
SYSD 1.2	Mon	10:45–11:15	H2	**Nonequilibrium Transport and Dynamics in Conventional and Topological Superconducting Junctions** — •Raffael L. Klees
SYSD 1.3	Mon	11:15–11:45	H2	**Probing magnetostatic and magnetotransport properties of the antiferromagnetic iron oxide hematite** — •Andrew Ross
SYSD 1.4	Mon	11:45–12:15	H2	**Quantum dot optomechanics with surface acoustic waves** — •Matthias Weiss

Invited Talks of the joint Symposium United Kingdom as Guest of Honor (SYUK)

See SYUK for the full program of the symposium.

SYUK 1.1	Wed	9:30–10:00	H2	**Structure and Dynamics of Interfacial Water** — •Angelos Michaelides
SYUK 1.2	Wed	10:00–10:30	H2	**A molecular view of the water interface** — •Mischa Bonn
SYUK 1.3	Wed	10:30–11:00	H2	**Motile cilia waves: creating and responding to flow** — •Pietro Cicuta
SYUK 1.4	Wed	11:00–11:30	H2	**Cilia and flagella: Building blocks of life and a physicist's playground** — •Oliver Bäumchen
SYUK 1.5	Wed	11:45–12:15	H2	**Computational modelling of the physics of rare earth - transition metal permanent magnets from $SmCo_5$ to $Nd_2Fe_{14}B$** — •Julie Staunton
SYUK 2.1	Wed	15:00–15:30	H2	**Hysteresis Design of Magnetic Materials for Efficient Energy Conversion** — •Oliver Gutfleisch
SYUK 2.2	Wed	15:30–16:00	H2	**Non-equilibrium dynamics of many-body quantum systems versus quantum technologies** — •Irene D'Amico
SYUK 2.3	Wed	16:00–16:30	H2	**Quantum computing with trapped ions** — •Ferdinand Schmidt-Kaler
SYUK 2.4	Wed	16:45–17:15	H2	**Breaking the millikelvin barrier in cooling nanoelectronic devices** — •Richard Haley
SYUK 2.5	Wed	17:15–17:45	H2	**Superconducting Quantum Interference Devices for applications at mK temperatures** — •Sebastian Kempf

Invited Talks of the joint Symposium Frontiers of Electronic-Structure Theory: Focus on Artificial Intelligence Applied to Real Materials (SYES)

See SYES for the full program of the symposium.

SYES 1.1	Thu	15:00–15:30	H1	**Machine-learning-driven advances in modelling inorganic materials** — •Volker L. Deringer
SYES 1.2	Thu	15:30–16:00	H1	**Machine-Learning Discovery of Descriptors for Square-Net Topological Semimetals** — •Eun-Ah Kim
SYES 1.3	Thu	16:00–16:30	H1	**Four Generations of Neural Network Potentials** — •Jörg Behler
SYES 1.4	Thu	16:30–17:00	H1	**Using machine learning to find density functionals** — •Kieron Burke
SYES 1.5	Thu	17:00–17:30	H1	**Coarse graining for classical and quantum systems** — •Cecilia Clementi

Sessions

O 1.1–1.1	Mon	9:30–10:15	S054	**Overview Talk Bärbel Rethfeld**
O 2.1–2.10	Mon	10:30–13:00	H3	**Ultrafast Electron Dynamics at Surfaces and Interfaces 1**
O 3.1–3.8	Mon	10:30–12:45	H4	**Focus Session: Single Atom Catalysis 1**
O 4.1–4.4	Mon	10:30–11:30	H6	**Topology and Symmetry-Protected Materials**
O 5.1–5.9	Mon	10:30–13:00	S051	**Organic Molecules at Surfaces 1: Substrate Effects**
O 6.1–6.10	Mon	10:30–13:00	S052	**Nanostructures at Surfaces 1**
O 7.1–7.9	Mon	10:30–13:00	S053	**New Methods and Developments 1: Scanning Probe Techniques 1 (joint session O/KFM)**
O 8.1–8.8	Mon	10:30–12:45	S054	**Solid-Liquid Interfaces 1: Reactions and Electrochemistry**
O 9.1–9.13	Mon	15:00–18:15	H3	**Ultrafast Electron Dynamics at Surfaces and Interfaces 2**
O 10.1–10.7	Mon	15:00–17:30	H4	**Focus Session: Single Atom Catalysis 2**
O 11.1–11.6	Mon	15:00–16:30	H6	**Electronic Structure Theory**
O 12.1–12.12	Mon	15:00–18:00	S051	**Organic Molecules at Surfaces 2: Characterization of Organic Monolayers**
O 13.1–13.10	Mon	15:00–17:30	S052	**Nanostructures at Surfaces 2**
O 14.1–14.5	Mon	15:00–16:15	S053	**New Methods and Developments 2: Scanning Probe Techniques 2 (joint session O/KFM)**
O 15.1–15.11	Mon	15:00–18:00	S054	**Solid-Liquid Interfaces 2: Structure and Spectroscopy**
O 16.1–16.12	Mon	18:00–20:00	P4	**Poster Monday: Ultrafast Processes 1**
O 17.1–17.12	Mon	18:00–20:00	P4	**Poster Monday: Organic Molecules at Surfaces 1**
O 18.1–18.28	Mon	18:00–20:00	P4	**Poster Monday: 2D Materials 1**
O 19.1–19.10	Mon	18:00–20:00	P4	**Poster Monday: Scanning Probe Techniques 1**
O 20.1–20.11	Mon	18:00–20:00	P4	**Poster Monday: Solid-Liquid Interfaces**
O 21.1–21.5	Mon	18:00–20:00	P4	**Poster Monday: Topology and Symmetry-Protected Materials**
O 22.1–22.8	Mon	18:00–20:00	P4	**Poster Monday: Surface Structure, Epitaxy, Growth and Tribology**

O 23.1–23.9	Mon	18:00–20:00	P4	**Poster Monday: Nanostructures 1**
O 24.1–24.1	Tue	9:30–10:15	S054	**Overview Talk Yang Shao-Horn**
O 25.1–25.7	Tue	10:30–13:00	H2	**Focus Session: Atomic-Scale Characterization of Correlated Ground States in Epitaxial 2D Materials**
O 26.1–26.9	Tue	10:30–12:45	H4	**Surface Magnetism**
O 27.1–27.5	Tue	10:30–11:45	H6	**Electron-Driven Processes**
O 28.1–28.9	Tue	10:30–12:45	S051	**Organic Molecules at Surfaces 3: Theory**
O 29.1–29.7	Tue	10:30–12:15	S052	**Metal substrates 1**
O 30.1–30.8	Tue	10:30–12:45	S053	**Semiconductor Surfaces**
O 31.1–31.7	Tue	10:30–12:30	S054	**Solid-Liquid Interfaces 3: Reactions and Electrochemistry**
O 32.1–32.9	Tue	11:00–13:00	P3	**Poster Tuesday: Adsorption and Catalysis 1**
O 33.1–33.11	Tue	11:00–13:00	P3	**Poster Tuesday: Ultrafast Processes 2**
O 34.1–34.10	Tue	11:00–13:00	P3	**Poster Tuesday: Scanning Probe Techniques 2**
O 35.1–35.8	Tue	11:00–13:00	P3	**Poster Tuesday: Plasmonics and Nanooptics 1**
O 36.1–36.1	Wed	9:30–10:15	S054	**Overview Talk Hendrik Bluhm**
O 37.1–37.8	Wed	10:30–12:45	H3	**Plasmonics and Nanooptics 1**
O 38.1–38.7	Wed	10:30–12:30	H4	**Solid-Liquid Interfaces 4: Reactions and Electrochemistry**
O 39.1–39.7	Wed	10:30–12:15	H6	**Tribology**
O 40.1–40.9	Wed	10:30–12:45	S051	**Organic Molecules at Surfaces 4: Chemistry on Surfaces**
O 41.1–41.8	Wed	10:30–12:30	S052	**Graphene: Growth, Substrate Interaction, Intercalation, and Doping**
O 42.1–42.5	Wed	10:30–11:45	S053	**Metal substrates 2**
O 43.1–43.10	Wed	10:30–13:00	S054	**Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 1**
O 44.1–44.10	Wed	15:00–18:30	H3	**Focus Session: Surfaces and Interfaces of (Incipient) Ferroelectrics (joint session O/KFM)**
O 45.1–45.9	Wed	15:00–18:00	H4	**Focus Session: Catalysis at Liquid Interfaces**
O 46.1–46.8	Wed	15:00–17:00	H6	**New Methods and Developments 3: Theory**
O 47.1–47.10	Wed	15:00–18:00	S051	**Focus Session: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy 1**
O 48.1–48.10	Wed	15:00–17:30	S052	**2D Materials 1: Electronic Structure of Transition Metal Dichalcogenides**
O 49.1–49.10	Wed	15:00–17:30	S053	**Oxide Surfaces 1**
O 50.1–50.12	Wed	15:00–18:00	S054	**Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 2**
O 51.1–51.10	Wed	18:00–20:00	P4	**Poster Wednesday: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy**
O 52.1–52.12	Wed	18:00–20:00	P4	**Poster Wednesday: Adsorption and Catalysis 2**
O 53.1–53.5	Wed	18:00–20:00	P4	**Poster Wednesday: Spins and Magnetism**
O 54.1–54.14	Wed	18:00–20:00	P4	**Poster Wednesday: 2D Materials 2**
O 55.1–55.16	Wed	18:00–20:00	P4	**Poster Wednesday: Organic Molecules at Surfaces 2**
O 56.1–56.6	Wed	18:00–20:00	P4	**Poster Wednesday: Nanostructures 2**
O 57.1–57.7	Wed	18:00–20:00	P4	**Poster Wednesday: Electronic Structure**
O 58.1–58.15	Wed	18:00–20:00	P4	**Poster Wednesday: New Methods and Developments, Frontiers of Electronic Structure Theory**
O 59.1–59.8	Wed	18:00–20:00	P4	**Poster Wednesday: Plasmonics and Nanooptics 2**
O 60.1–60.1	Thu	9:30–10:15	S054	**Overview Talk Thorsten Deilmann**
O 61.1–61.8	Thu	10:30–12:45	H2	**Plasmonics and Nanooptics 2**
O 62.1–62.9	Thu	10:30–12:45	H4	**Surface Reactions and Heterogeneous Catalysis 1**
O 63.1–63.7	Thu	10:30–12:30	H6	**New Methods and Developments 4: Spectroscopies, Diffraction and Others (joint session O/KFM)**
O 64.1–64.5	Thu	10:30–13:00	S051	**Gerhard Ertl Young Investigator Award**
O 65.1–65.8	Thu	10:30–12:30	S052	**2D Materials 2: Growth, Structure and Substrate Interaction**
O 66.1–66.9	Thu	10:30–13:00	S053	**Oxide Surfaces 2**
O 67.1–67.9	Thu	10:30–12:45	S054	**Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 3**
O 68.1–68.10	Thu	15:00–18:30	H3	**Focus Session: Time-Resolved Momentum Microscopy**
O 69.1–69.10	Thu	15:00–17:45	H4	**Surface Reactions and Heterogeneous Catalysis 2**
O 70.1–70.10	Thu	15:00–17:45	H6	**Supported nanoclusters: Structure, Reactions, Catalysis**
O 71.1–71.10	Thu	15:00–18:00	S051	**Focus Session: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy 2**

O 72.1–72.8	Thu	15:00–17:00	S052	**2D Materials 3: hBN and Electronic Structure**
O 73.1–73.11	Thu	15:00–17:45	S053	**Electronic Structure of Surfaces 1**
O 74.1–74.12	Thu	15:00–18:00	S054	**Organic Molecules at Surfaces 5: Molecular Switches**
O 75	Thu	19:00–19:30	H1	**Members' Assembly**
O 76	Thu	19:30–20:30	H1	**Post-Deadline Session**
O 77.1–77.1	Fri	9:30–10:15	S054	**Overview Talk Guillaume Schull**
O 78.1–78.8	Fri	10:30–12:30	H3	**Plasmonics and Nanooptics 3**
O 79.1–79.8	Fri	10:30–12:45	H4	**Surface Reactions and Heterogeneous Catalysis 3**
O 80.1–80.7	Fri	10:30–12:15	S051	**Focus Session: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy 3**
O 81.1–81.6	Fri	10:30–12:00	S052	**2D Materials 4: Heterostructures**
O 82.1–82.7	Fri	10:30–12:15	S053	**Electronic Structure of Surfaces 2**
O 83.1–83.10	Fri	10:30–13:00	S054	**Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 4**
O 84.1–84.1	Fri	13:15–14:00	S054	**Overview Talk Claus M. Schneider (joint session O/CPP)**

Members' Assembly of the Surface Science Division

Thursday 19:00–19:30 H1

- Report of the Chairman
- Presentation of the Gerhard Ertl Young Investigator Award
- Miscellaneous

O 1: Overview Talk Bärbel Rethfeld

Time: Monday 9:30–10:15 Location: S054

Invited Talk O 1.1 Mon 9:30 S054

Laser-excited electrons: how hot are they? — •Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany

Hot electrons play an important role in surface science. They are responsible for the functionalization of solar cells and the emission of intense THz beams, they drive chemical reactions at surfaces and are utilized in biosensing methods. Excitation and decay as well as transport of laser-excited electrons are fundamentally studied in pump-probe experiments using photoemission methods.

The term “hot electrons” is, however, not well-defined. It is used for single laser-excited electrons, for non-equilibrium electron distributions, and for Fermi distributions at elevated temperature. In all cases, the relaxation of such electrons towards an equilibrated situation is of interest. Its dynamics may be complex, since several scattering mechanisms act on different timescales. Therefore, athermal electron distributions as well as highly excited electrons can exist much longer than the single-electron lifetime predicts.

In this presentation, I introduce the calculation method of Boltzmann collision integrals, allowing to study the effect and mutual influence of different scattering mechanisms. I further show how such intertwined relaxation processes affect measurable macroscopic quantities on ultrafast timescales, particularly in multiband materials. Examples include the electrical conductivity in highly excited noble metals, spin and charge transport in magnetically ordered systems and the nonthermal electron-phonon coupling strength in metals and dielectrics.

O 2: Ultrafast Electron Dynamics at Surfaces and Interfaces 1

Time: Monday 10:30–13:00 Location: H3

O 2.1 Mon 10:30 H3

Floquet Dressing and Multiphoton Photoemission on Metal Surfaces — •Yun Yen[1,2] and Michael Schüler[2] — [1]Condensed Matter Theory Group, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland — [2]Ecole Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland

Floquet engineering allows us to realize the control of quantum materials and explore exotic properties from light-matter interaction. Along with the development of various ultrafast experimental techniques, non-equilibrium quantum systems have gained more interests.

Metal surfaces such as Cu(111) or Ag(111) are considered as benchmarks for photoemission spectroscopy with nonlinear optical responses. The interplay between Shockley surface states (SS), image potential states (IP), and the related dressed bands can be studied with ultrafast pump-probe pulses. Starting from model potential, we utilize real space time-dependent surface flux method and non-equilibrium Green*s function to simulate the non-equilibrium dynamics on the metal surfaces. We calculate the interferometric-time-resolved multiphoton photoemission (ITR-mPP) spectrum. The Fourier analysis of the simulated ITR-mpp spectrum shows the optical dressing of the surface electronic structures. Our approach successfully reproduce the previous experimental data, which allows us to investigate the origins of these signatures.

O 2.2 Mon 10:45 H3

Femtosecond orbital tomography of exciton dynamics in C_{60} thin films — •G. S. Matthijs Jansen[1], Wiebke Bennecke[1], Ralf Hemm[2], Andreas Windischbacher[3], David Schmitt[1], Jan Phillip Bange[1], Christian Kern[3], Daniel Steil[1], Sabine Steil[1], Marcel Reutzel[1], Martin Aeschlimann[2], Peter Puschnig[3], Benjamin Stadtmüller[2,4], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen — [2]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern — [3]Institute of Physics, University of Graz — [4]Institute of Physics, Johannes Gutenberg-University Mainz

Time-resolved photoemission orbital tomography promises to be a unique probe of out-of-equilibrium electronic wavefunctions at the Ångström level and with femtosecond time resolution. Particularly interesting is the application to excitons, quasiparticles consisting of a bound electron and electron hole that govern the opto-electronic response of organic semiconductors. Here, we consider the prototypical organic semiconductor C_{60}, for which it has been proposed that the exciton cascade involves the two-step decay of a delocalized charge-transfer exciton into a localized Frenkel excitonic state. We will discuss how the combination of multi-dimensional photoemission spectroscopy with calculations within the many-body perturbation theory framework of the Bethe-Salpeter equation can shed light on the real-space femtosecond electron dynamics, and in particular show how the delocalization of excitons in C_{60} evolves on the femtosecond timescale.

O 2.3 Mon 11:00 H3

Anisotropic carrier dynamics in a laser-excited Fe/(MgO)(001) heterostructure from real-time TDDFT — •Elaheh Shomali, Markus Ernst Gruner, and Rossitza Pentcheva — Department of Physics and Center for Nanointegration, CENIDE, University of Duisburg-Essen, Germany

The interaction of a femtosecond optical pulse with a $Fe_n/(MgO)_m(001)$ (n=1,3,5 and m=3,5,7) metal/oxide heterostructure is addressed using time-dependent density functional theory (TDDFT) calculations in the real-time domain. We systematically study electronic excitations as a function of laser frequency (around and higher than the bulk MgO band gap). We find a marked anisotropy in the response to in- and out-of-plane polarized light, which changes its character qualitatively depending on the excitation energy: the Fe-layer is efficiently addressed at low frequencies by in-plane polarized light, whereas for frequencies higher than the MgO band gap, we find a particularly large sensitivity of MgO-layers to cross-plane polarized light. Moreover, the interface plays an important role, as it mediates transitions from the valence band of MgO into the $3d$ states of Fe closely above the Fermi-level and transitions from the Fe-states below the Fermi level into the conduction band of MgO. As these transitions can occur simultaneously without altering the charge balance of the layers, they could potentially lead to an efficient transfer of excited carriers into the MgO part [1]. Funding by SFB 1242, project C02, is gratefully acknowledged. [1] E. Shomali, M. E. Gruner, R. Pentcheva, Phys. Rev. B, in press, arXiv:2205.03178.

O 2.4 Mon 11:15 H3

Ultrafast electron transport across interfaces in Au/Fe/MgO(001) heterostructures — Yasin Beyazit[1], Florian Kühne[1], Markus Heckschen[1], Björn Sothmann[1], Elaheh Shomali[1], Markus Gruner[1], Rossitza Pentcheva[1], Ping Zhou[1], Detlef Diesing[2], and •Uwe Bovensiepen[1] — [1]Universität Duisburg-Essen, Fakultät für Physik, 47048 Duisburg — [2]Universität Duisburg-Essen, Fakultät für Chemie, 45141 Essen

The dynamics of electronic excitations at buried interfaces is decisive in electron transfer and carrier multiplication in heterostructures. Following recent developments in time-resolved two-photon photoemission spectroscopy the relaxation dynamics of optically excited, hot electrons can be distinguished in the individual layers of Au/Fe/MgO(001) by analysis of the Au thickness dependent relaxation times [Beyazit et al., PRL **125**, 076803 (2020)]. Here we report on recent experimental results and the separation of the primary, optically excited electrons and the secondary, relaxed and propagating electron distribution. This study is complemented by calculations using the Boltzmann transport equation for the propagation through the Au layer and density functional theory calculations resolving the interface contribution.

Funding by the Deutsche Forschungsgemeinschaft through SFB 1242 is gratefully acknowledged.

O 2.5 Mon 11:30 H3

Non-equilibrium carrier dynamics of different surface reconstructions of Sn on SiC(0001) — •Maximilian Stecher[1], Maria-Elisabeth Federl[1], Niklas Hofmann[1], Leonard Weigl[1], Johannes Gradl[1], Neeraj Mishra[2,3], Stiven Forti[2], Camilla Coletti[2,3], and Isabella Gierz[1] — [1]Department of Physics, University of Regensburg, 93040 Regensburg, Germany — [2]Center for Nanotechnology Innovation @NEST, Istituto Italiano di Tecnologia, 56127 Pisa, Italy — [3]Graphene Labs, Istituto Italiano di Tecnologia, 16163 Genova, Italy

Sn on SiC(0001) forms different surface reconstructions in the monolayer coverage regime, among them a Mott insulating phase with a $(\sqrt{(3)}\mathrm{x}\sqrt{(3)})\mathrm{R}30^\circ$ structure. We grow these structures and characterize them with low-energy electron diffraction (LEED), X-ray photoemission spectroscopy (XPS), and angle-resolved photoemission spectroscopy (ARPES). We then investigate their light-induced non-equilibrium carrier dynamics with time- and angle-resolved photoemission spectroscopy, looking for possible photo-induced phase transitions and metastable transient states. These preliminary studies then pave the way towards dynamical control of the electronic properties of Sn/SiC(0001) via resonant excitation of the IR-active phonon modes of the substrate that will modulate the atomic positions of the covalently bound Sn layer coherently.

O 2.6 Mon 11:45 H3

ultrafast carrier dynamics of graphene - 2D Sn van der Waals interface — •Maria-Elisabeth Federl[1], Niklas Hofmann[1], Leonard Weigl[1], Johannes Gradl[1], Tim Wehling[2,3], Niklas Witt[3], Neeraj Mishra[4], Camilla Coletti[4,5], Stiven Forti[4], and Isabella Gierz[1] — [1]Department of Physics, University of Regensburg, 93040 Regensburg, Germany — [2]Institute for Theoretical Physics, University of Bremen, 28359 Bremen, Germany — [3]Institute for Theoretical Physics, University of Hamburg, 22607 Hamburg, Germany — [4]Center for Nanotechnology Innovation @NEST, Instituto Italiano di Tecnologia, Piazza San Silvestro 12, 56127 Pisa, Italy — [5]Graphene Labs, Instituto Italiano di Tecnologia, 16163 Genova, Italy

The interface between epitaxial graphene and SiC(0001) is a confined space that allows for the growth of novel two-dimensional materials (2DMs) and thus graphene/2DM heterostructures. The electronic properties of these heterostructures are usually more complex than those expected from the sum of the individual layers as hybridization between the layers opens up band gaps in the electronic structure and produces electronic wavefunctions that are localized on both rather than individual layers of the heterostructure. We use time- and angle-resolved photoemission spectroscopy to reveal important differences between the non-equilibrium carrier dynamics of quasi-freestanding graphene and graphene proximity-coupled to a 2D metallic Sn layer that we interpret in terms of doping, scattering phase space, and ultrafast charge transfer with the help of ab initio band structure calculations.

O 2.7 Mon 12:00 H3

tr-ARPES experiment with mid-infrared excitation for dynamical band structure engineering — •Leonard Weigl[1], Niklas Hofmann[1], Johannes Gradl[1], Neeraj Mishra[2,3], Stiven Forti[2], Camilla Coletti[2,3], and Isabella Gierz[1] — [1]Department of Physics, University of Regensburg, 93040 Regensburg, Germany — [2]Center for Nanotechnology Innovation @NEST, Istituto Italiano di Tecnologia, 56127 Pisa, Italy — [3]Graphene Labs, Istituto Italiano di Tecnologia, 16163 Genova, Italy

The band structure of a solid is mainly determined by the orbital overlap between neighboring atoms. Therefore, electronic properties are commonly controlled via the chemical composition that determines structural parameters such as bond angles and lengths. Recently, control of the effective orbital overlap has been achieved by periodic modulation of solids with strong mid-infrared (MIR) light fields. The ideal probe for investigating driving-induced band structure changes is time- and angle-resolved photoemission spectroscopy (tr-ARPES). We have built a tr-ARPES setup that combines a strong-field wavelength-tunable MIR pump source with extreme ultraviolet probe pulses providing access to the band structure of driven solids across the complete first Brillouin zone with sub 100meV energy and sub 250fs temporal resolution. We present first results on driven graphene, analyze the MIR pump-induced carrier dynamics, and discuss the possible formation of photon-dressed states. The excellent performance of the setup now paves the way for the future investigation of light-induced superconductivity as well as light-induced topological phase transitions.

O 2.8 Mon 12:15 H3

Anisotropic response to optical excitations in naturally layered delafossite $\mathrm{PdCoO_2}$ from time-dependent DFT — •Mike J. Bruckhoff, Markus E. Gruner, and Rossitza Pentcheva — Faculty of Physics and Center of Nanointegration, CENIDE, University of Duisburg-Essen, 47048 Duisburg, Germany

In the framework of time-dependent density functional theory (TDDFT), we investigate the layer-resolved dynamics of the electronic structure of the metallic delafossite $\mathrm{PdCoO_2}$ after optical excitations. $\mathrm{PdCoO_2}$ can be conceived as a natural multilayer system consisting of highly conductive Pd layers separated by insulating $\mathrm{CoO_6}$ octahedra. We calculate the responses to optical pulses with two polarization directions, multiple frequencies, different pulse durations and laser fluences within the real-time approach (RT-TDDFT) and compare the results to optical absorption spectra obtained within the linear-response regime (LR-TDDFT). We observe a strong anisotropy of the electronic response to different polarization directions and particular frequencies of the incident electric field, which corresponds to the strong anisotropy visible in the absorption spectra. In particular, we see a significant charge redistribution and time-dependent changes of occupation numbers, which depend on the orbital character of the involved *d*-orbitals. Analogies to previous studies for a $\mathrm{Fe_1/(MgO)_3(001)}$ heterostructure [1] are discussed. Funding by DFG within SFB1242 is gratefully acknowledged.
[1]: M. E. Gruner and R. Pentcheva, Phys. Rev. B 99, 195104 (2019)

O 2.9 Mon 12:30 H3

Far-from-equilibrium electron-phonon interactions in optically-excited graphene — •Marco Merboldt[1], Marten Düvel[1], Jan Philipp Bange[1], Michael Stellbrink[1], Hannah Strauch[1], Klaus Pierz[2], Hans Werner Schumacher[2], Davood Momeni[2], Daniel Steil[1], G. S. Matthijs Jansen[1], Sabine Steil[1], Dino Novko[3], Stefan Mathias[1], and Marcel Reutzel[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Göttingen, Germany — [2]Physikalisch-Technische Bundesanstalt, Braunschweig, Germany — [3]Institute of Physics, Zagreb, Croatia

Comprehending far-from-equilibrium many-body interactions is one major goal of current ultrafast condensed matter physics research. A particularly interesting but barely understood situation occurs during strong optical excitation, where electron and phonon systems are significantly perturbed from their equilibrium and cannot be described by Fermi-Dirac or Bose-Einstein distributions, respectively.
Here, we use time- and angle-resolved photoelectron spectroscopy (trARPES) to study such situation for the prototypical material graphene. We show that upon optical excitation, it exhibits a complex non-equilibrium many-body response by evaluating the Dirac state linewidth and thus the imaginary part of the quasiparticle self-energy $\mathrm{Im}\Sigma$ from spectrally deconvoluted trARPES data. By employing first-principles theoretical modeling, we find that the observed experimental features are caused by ultrafast NEQ scatterings between optical phonons and photoexcited charge carriers, active on timescales well below 100 fs. Düvel, Merboldt *et al.*, Nano Letters, accepted.

O 2.10 Mon 12:45 H3

Coherent Time-Resolved Above-Threshold Plasmoemission from the Au(111) Shockley Surface State — •Pascal Dreher, David Janoschka, Alexander Neuhaus, Michael Horn-von Hoegen, and Frank Meyer zu Heringdorf — Faculty of Physics, University of Duisburg-Essen, 47048 Duisburg, Germany

Strong nonperturbative interactions of an intense driving light field with the electronic band structure in a solid can result in exotic material properties that do not exist under equilibrium conditions. For suitable driving conditions, Floquet theory predicts that the originally unperturbed electronic structure is modified by the formation of light-dressed electron states in strong fields. Observing such dressing requires electronic state resolution as well as precise control over the intense periodic driving field to overcome intrinsic dissipation and decoherence in the solid.

Here, we explore nano-focusing of femtosecond surface plasmon polariton (SPP) pulses on flat surfaces as a possible route towards strong-field control over electronic states within a solid using time- and angle-resolved photoemission spectromicroscopy. We observe above-threshold electron emission from the Au(111) Shockely surface state by the absorption of up to seven SPP quanta. Two-dimensional time-resolved photoelectron spectroscopy using a birefringent delay line provides us with direct access to the coherent and incoherent dynamics of the electron emission process with attosecond precision. The presented results clearly indicate the coherent nature of the interaction of the intense SPP nano-focus with the band structure of the material.

O 3: Focus Session: Single Atom Catalysis 1

The field of single atom catalysis has emerged rapidly in recent years out of the desire to utilize less and less precious metals, and remarkable successes have been reported in thermo-, electro-, and photocatalysis. While much of the early focus was on establishing synthesis routes to achieve atomic dispersion, it has become clear that "single atom" systems cannot be considered as the smallest possible nanoparticle. Because isolated adatoms are stabilized by chemical bonds to the support, they have much more in common with mononuclear complexes used in homogenous catalysis. Thus, the focus has now changed towards understanding the role of coordination of the active site, and developing ways to tailor it toward specific processes. This has led to much interest in fundamental work, which opens up a tremendous opportunity for surface science to contribute to a hot topic in catalysis.

Organizer: Gareth Parkinson (Technical University Vienna), Matthias Meier (University of Vienna)

Time: Monday 10:30–12:45 Location: H4

Topical Talk O 3.1 Mon 10:30 H4

Rational design of single atom electrocatalysts: handle with care — •Gianfranco Pacchioni — Dipartimento di Scienza dei Materiali, Università Milano-Bicocca, Milano, Italy

One of the objectives of electronic structure theory is to predict chemical and catalytic activities. This is a challenging target due to the large number of variables that determine the performance of a heterogeneous catalyst. The complexity of the problem has reduced considerably with the advent of single atom catalysts (SAC) and, in particular, of graphene-based SACs for electrocatalytic reactions such as the oxygen reduction (ORR), the oxygen evolution (OER) and the hydrogen evolution (HER) reactions. In this context we assist to a rapidly growing number of theoretical studies based on density functional theory (DFT) and of proposals of universal descriptors that should provide a guide to the experimentalist for the synthesis of new catalysts. In this talk we critically analyze some of the current problems connected with the prediction of the activity of SACs based on DFT: accuracy of the calculations, neglect of important contributions in the models used, physical meaning of the proposed descriptors, inaccurate data sets used to train machine learning algorithms, not to mention some severe problems of reproducibility. It follows that the 'rational design' of a catalyst based on some of the proposed universal descriptors or of the DFT screening of large number of structures should be considered with great caution.

O 3.2 Mon 11:00 H4

Single atom co-catalyst dispersion on KTaO3 (001) by surface polarity compensation. — •Aji Alexander[1], Jesús Redondo[1], Dominik Wrana[1], Lukáš Fusek[1], Viktor Johánek[1], Josef Mysliveček[1], and Martin Setvin[1,2] — [1]Department of Surface and Plasma Science, Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic — [2]Institute of Applied Physics, TU Wien, Vienna, Austria

Redox chemistry on perovskite surfaces attracts attention due to these materials* promising catalytic properties, good ability to separate electron-hole pairs in light-harvesting, and the presence of ferroelectricity in many perovskites. This work focuses on enhancing catalytic activity achieved by activating the perovskite surface with extrinsic metals. Combined STM/AFM measurement together with XPS data shows the tendency of the cobalt atoms to disperse in the form of single adatoms on the polarity uncompensated KTaO3 (001) surface [1]. The interaction of cobalt with KTO surfaces was studied under various reducing and oxidizing conditions, as well as a function of temperature. This, in turn, will allow characterization of the metallic, oxide, and hydroxide phases of cobalt in dependence on the environment. The work was supported by projects GACR 20-21727X and GAUK Primus/20/SCI/009. [1] M. Setvin, M. Reticcioli, F. Poelzleitner et al., Science 359, 572 (2018)

O 3.3 Mon 11:15 H4

Comparison study of different transition metals on two TiO2 model supports: anatase TiO2(101) and rutile TiO2(110) — •Lena Puntscher[1], Kevin Daninger[1], Panukorn Sombut[1,2], Matthias Meier[2], Michael Schmid[1], Cesare Franchini[2], Ulrike Diebold[1], and Gareth S. Parkinson[1] — [1]Institute of Applied Physics, TU Wien, Austria — [2]Faculty of Physics, University of Vienna, Vienna, Austria

Single-atom catalysis (SAC) offers an opportunity to minimize the amount of precious catalyst material required for traditional heterogeneous catalysis and to heterogenize reactions presently requiring homogeneous catalysis. Unravelling how metal atoms bind to oxide supports is crucial for a better understanding of the SACs catalytic properties. Using STM and XPS, we compare the adsorption geometry and stability of several transition metals (Pt, Rh, Ir and Ni) on TiO2 model supports: anatase TiO2(101) and rutile TiO2(110), and the influence of water on the dispersion of these systems. While most of the metals rapidly sinter on both surfaces, there are a few exceptions: Ir forms stable adatoms on the anatase support. Ni is much more dispersed when water vapor is added to the deposition and a very low coverage of Pt single atoms can be stabilized in oxygen vacancies formed on rutile (110), which are only stable in UHV conditions. This study points out the importance of metal-support interaction and the surprisingly different behaviour of the transition metals on TiO2 model supports.

O 3.4 Mon 11:30 H4

Methane Activation by Free Tantalum Cluster Cations: When the Atomic System is Different — •Martin Tschurl, Jan Eckhard, and Ueli Heiz — Physical Chemistry, Department of Chemistry and Catalysis Research Center, Technical University of Munich

The efficient conversion of methane into valuable chemicals represents a challenge in chemistry in general. It also carries the potential of becoming a key technological process, given the availability of the substance from fossil, as well as agricultural, sources. Despite decades of research, only steam reforming is used today on commercially relevant scales. Nevertheless, the interest in the topic remains steadily strong, in particular in heterogeneous catalysis. Studies of ion-molecule reactions have already provided substantial contributions to the field and enabled the identification of fundamental reaction pathways and key properties governing them. We use of this approach in order to reveal pathways and reaction kinetics in the C-H bond activation in methane. This way, we comprehensively elucidate the size-dependent reactivity of different cluster cations of tantalum - a metal, which has already been noted as a prospective candidate in the activation of C-H bonds. One of our main finding, in the combined experimental and theoretical study, reveals a significant difference in the reaction of single atoms compared to that of larger clusters. The single atom is the only size that facilitates an often-desired C-C coupling, which further suggests a possible intrinsic benefit of single atoms over clusters in the catalytic conversion of methane.

O 3.5 Mon 11:45 H4

Assessing the environmental benefit of palladium-based single-atom heterogeneous catalysts for Sonogashira coupling — •Dario Faust Akl[1], Dario Poier[1,2], Sharon Mitchell[1], Roger Marti[2], Gonzalo Guillén-Gosálbez[1], and Javier Pérez-Ramírez[1] — [1]Institute of Chemical and Bioenginnering, ETH Zurich, Switzerland — [2]ChemTech, HEIA Fribourg, Switzerland

The Pd-Cu catalyzed Sonogashira coupling of terminal alkynes and aryl halides is a cornerstone organic transformation. A cradle-to-gate life cycle analysis (LCA) reveals a two orders of magnitude potential improvement in process footprint when replacing an organometallic Pd catalyst with a heterogeneous analogue. The latter could be easily separated permitting full Pd recovery and catalyst reuse. Heterogeneous catalysts based on isolated metal atoms (single-atom catalysts, SACs) demonstrate promising potential to synergize the benefits of solid and molecular catalysts for efficient Pd utilization. By anchoring Pd atoms on nitrogen-doped carbon we achieve full recovery of the metal, allowing catalyst reuse over multiple cycles. A hybrid process using the Pd-SAC with a homogeneous CuI cocatalyst is more productive than a fully heterogeneous bimetallic Pd-Cu SAC, which deactivates severely due to copper leaching. In some scenarios, the LCA-based metrics demonstrate the process footprint of the hybrid SAC system is leaner than the purely homogeneous counterpart already upon single reuse. Combining LCA with experimental evaluation could guide the design of reusable catalysts for more sustainable organic transformations.

O 3.6 Mon 12:00 H4

Nature of the active species in low-temperature CO oxidation over Pt-CeO2 model catalysts — •Alexander Simanenko[1], Maximilian Kastenmeier[1], Lesia Piliai[2], Tomáš Skála[2], Yuliia Kosto[2], Nataliya Tsud[2], Sascha Mehl[3], Mykhailo Vorokhta[2], Iva Matolínová[2], Yaroslava Lykhach[1], and Jörg Libuda[1] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany — [2]Charles University, Prague, Czech Republic — [3]Elettra-Sincrotrone Trieste SCpA, Basovizza-Trieste, Italy

The low-temperature CO oxidation has wide applications in energy production and exhaust gas cleaning. Pt-CeO2 catalysts with high Pt content allow achieving 100% CO conversion below room temperature. We investigated the nature of active sites on Pt-CeO2 catalysts under the conditions of low-temperature CO oxidation by means of synchrotron radiation photoelectron spectroscopy, resonant photoemission spectroscopy, and near ambient pressure XPS. Model Pt-CeO2 systems were prepared by reactive physical vapor co-deposition and annealed in UHV or O2 in order to obtain different types of platinum species on the surface. These included mainly Pt2+ species accompanied by a small number of Pt4+ species that were reduced to Pt2+ or metallic Pt during annealing. Exclusively, stable Pt4+ species were formed on a thick Pt-CeO2 catalyst with high Pt content after annealing in oxygen at NAP conditions. The results of our study showed that there is no reduction or oxidation of the Pt-CeO2 catalysts by CO below 450 K. These observations suggest that low-temperature CO oxidation on Pt-CeO2 catalyst does not follow a Mars-Van Krevelen mechanism.

O 3.7 Mon 12:15 H4

Adsorption sites and thermal stability of Pt adatoms on $\mathbf{Fe_2O_3(1\bar{1}02)}$ — •Ali Rafsanjani Abbasi, Florian Kraushofer, Lena Haager, Moritz Eder, Jiri Pavelec, Giada Franceschi, Michele Riva, Michael Schmid, Ulrike Diebold, and Gareth S. Parkinson — Institute of Applied Physics, TU Wien, Austria

Oxide-supported Pt catalysts offer superior performance because of their high activity and/or selectivity for many important chemical reactions. However, the high cost of Pt and its susceptibility to CO poisoning are two drawbacks to its role as a catalyst. Downsizing catalyst clusters to single atoms is an effective way to reach maximum efficiency, and so-called “single-atom catalysis” is now an important field of research [1]. Nevertheless, stabilizing single Pt atoms on oxide supports without compromising catalytic activity is still a key challenge. Here, we present a surface science study to investigate the local binding environment of Pt adatoms deposited on $\mathrm{Fe_2O_3}(1\bar{1}02)-(1\times1)$ under UHV conditions. Extensive STM and XPS studies at different Pt surface coverages on $\mathrm{Fe_2O_3}(1\bar{1}02)$ revealed that Pt single atoms are highly stable at room temperature. STM images showed that Pt single atoms are adsorbing at two distinct sites with different apparent heights. Through an atom-by-atom analysis, the relative contributions of two types of Pt adsorption sites were determined for different surface coverages. Moreover, thermally induced sintering of the Pt single atoms is traced by means of XPS and STM.

[1] G. S. Parkinson, Catal. Lett. 149, 1137 (2019).

O 3.8 Mon 12:30 H4

Comparison of Single Metal Atoms on a Fe2O3 Model Support — •Gareth Parkinson[1], Ali Rafsanjani-Abbasi[1], lena Puntscher[1], Florian Kraushofer[1], Panukorn Sombut[1], Chunlei Wang[1], Matthias Meier[1,2], Moritz Eder[1], Jiri Pavelec[1], Giada Franceschi[1], Michele Riva[1], Michael Schmid[1], Cesare Franchini[2,3], and Ulrike Diebold[1] — [1]TU Wien, Vienna, Austria — [2]University of Vienna, Vienna, Austria — [3]Università di Bologna, Bologna, Italy

Understanding how the local environment of a ”single-atom” catalyst affects stability and reactivity remains a significant challenge. Fe2O3 is the most common iron-oxide support material utilized for SAC, but little is known about how metal adatoms bind at its surfaces. In this talk, I will compare and contrast the behavior of Pt, Rh, and Ir atoms on the flat, well-ordered (1x1) termination of Fe2O3(1-102). Using a combination of scanning probe microscopy and spectroscopic data, as well as theoretical calculations, I will demonstrate significant differences between the adsorption site and thermal stability of the metals, as well as differences in their interaction with water and CO.

O 4: Topology and Symmetry-Protected Materials

Time: Monday 10:30–11:30 Location: H6

O 4.1 Mon 10:30 H6

Tracing a Weyl Nodal Line in 3D-k-space via ARPES — •Tim Figgemeier[1], Maximilian Ünzelmann[1], Philipp Eck[2], Jakub Schusser[1], Jennifer Neu[3,4], Theo Siegrist[3,5], Domenico Di Sante[6], Giorgio Sangiovanni[2], Hendrik Bentmann[1] und Friedrich Reinert[1] — [1]EP VII, Universität Würzburg, Germany — [2]TP I, Universität Würzburg, Germany — [3]NHMFL, Tallahassee, FL, US — [4]Nuclear Nonproliferation Division, ORNL, Oak Ridge, Tennessee 37831, US — [5]FAMU-FSU, Tallahassee, FL, US — [6]Center for Computational Quantum Physics, Flatiron Institute, New York 10010, US

In contrast to the more conventional Dirac nodal lines, Weyl nodal lines (WNL) are only twofold-degenerate and this degeneracy is robust against strong spin-orbit coupling [1]. The occurance of WNL is guaranteed by the combination of a glide plane and time-reversal symmetry (TRS).

In this talk we will present a soft x-ray angle-resolved photoemission (ARPES) study where we will trace a WNL in 3D k-space throughout the Brillouinzone over a wide range of photon energies. The experimental results are compared to density functional and photoemission calculations. By utilizing dichroic ARPES experiments, we will moreover address the characteristics of the electronic wave functions around the WNL.

[1] M. Hirschmann et al.; Symmetry-enforced band crossings in tetragonal materials: Dirac and Weyl degeneracies on points, lines, and planes; Phys. Rev. Mat **5**; 054202 (2021)

O 4.2 Mon 10:45 H6

Momentum space signatures of Berry flux monopoles in a Weyl semimetal — •Maximilian Ünzelmann[1,3], Tim Figgemeier[1,3], Philipp Eck[2,3], Begmuhammet Geldiyev[1,3], Domenico Di Sante[2,3], Giorgio Sangiovanni[2,3], Hendrik Bentmann[1,3], and Friedrich Reinert[1,3] — [1]Experimentelle Physik 7, Universität Würzburg — [2]Theoretische Physik 1, Universität Würzburg — [3]Würzburg-Dresden Cluster of Excellence ct.qmat

An intriguing property of Weyl semimetals (WSM) is that they feature topologically protected crossings of the spin-polarized valence and conduction bands in their three-dimensional bulk band structure. These crossing points – the Weyl points (WP) – can be considered as ’magnetic’ monopoles, which, however, are not located in real space but rather in momentum space. The topological (monopole) character manifests itself in the momentum-dependence of the electronic wave functions, which is investigated in the paradigmatic WSM TaAs, using bulk-sensitive soft X-ray angle-resolved photoelectron spectroscopy combined with spin-resolved measurements and circular dichroism (CD) [1]. The latter addresses the local orbital angular momentum (OAM), which exhibits a non-trivial winding around the WP, similar to the Berry curvature, i.e., the k-space magnetic field. The momentum-dependent modulation of the OAM around the WP – observed in density functional theory calculations and CD-ARPES – thus denotes a direct signature for the Berry flux monopoles in TaAs.

[1] M. Ünzelmann et al., Nat. Commun., 12, 3650 (2021)

O 4.3 Mon 11:00 H6

Lifting the Spin-Momentum Locking in Ultra-Thin Topological Insulator Films — Arthur Leis[1], Jonathan Hofmann[1], Michael Schleenvoigt[2], Vasily Cherepanov[1], Felix Lüpke[1], Peter Schüffelgen[2], Gregor Mussler[2], Detlev

Grützmacher[2], •Bert Voigtländer[1], and F. Stefan Tautz[1] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Peter Grünberg Institut (PGI-9), Forschungszentrum Jülich, 52425 Jülich, Germany

3D topological insulators are known to carry 2D Dirac-like topological surface states in which spin-momentum locking prohibits backscattering. When thinned down to a few nanometers, the hybridization between the topological surface states at the top and bottom surfaces results in a topological quantum phase transition, which can lead to the emergence of a quantum spin Hall phase. Here, the thickness-dependent transport properties are studied on the example of BiSbTe3 films, with a four-tip scanning tunneling microscope. The findings reveal an exponential drop of the conductivity below the critical thickness. The steepness of this drop indicates the presence of spin-conserving backscattering between the top and bottom surface states, effectively lifting the spin-momentum locking and resulting in the opening of a gap at the Dirac point. Moreover, we probe the edge state conductance these films. The experiments provide a crucial step toward the detection of quantum spin Hall states in transport measurements.

O 4.4 Mon 11:15 H6

Simultaneous AFM and STM measurements of native point Defects in the topological insulator Bi$_2$Se$_3$ — •Christoph Setescak, Alexander Liebig, Adrian Weindl, and Franz J. Giessibl — Institute of Experimental and Applied Physics, University of Regensburg, Regensburg, Germany

The main properties of topological insulators (TIs) can be derived from the fact that their band structure is "twisted" compared to normal insulators, leading to the creation of gapless states at the surface. To observe these states and make them viable for application in novel technologies it is crucial that the materials delicate electronic structure is not disturbed by defects. So far, experimental characterization of the native point defects relies on scanning tunneling microscopy (STM) and accompanying density functional theory [1]. Here, we present simultaneous atomic force microscopy (AFM) and STM measurements of surface and subsurface defects in Bi$_2$Se$_3$. We find not only rarely-observed single Se surface vacancies, but also larger defects that are composed of multiple surface vacancies. The AFM channel furthermore allows us to determine the relative relaxation of surface atoms in proximity of subsurface defects and to determine the polarity of surface defects by means of Kelvin probe force spectroscopy. This is especially valuable as surface Se vacancies contribute to unwanted charge doping of the TI. [1] J. Dai et al., PRL 117, 106401 (2016)

O 5: Organic Molecules at Surfaces 1: Substrate Effects

Time: Monday 10:30–13:00 Location: S051

Topical Talk O 5.1 Mon 10:30 S051

Molecular nanostructures on metals vs. graphene — •Meike Stöhr — Zernike Institute for Advanced Materials, University of Groningen, Netherlands

To preserve the (functional) properties of either individual adsorbates or well-ordered molecular assemblies upon adsorption on solid surfaces, the molecule substrate interactions have to be generally relatively weak. This can be achieved by introducing a decoupling layer between (metallic) surface and molecules. Among others, graphene has been shown to be a good choice towards this end due to its low density of states around the Fermi level [1]. For two different organic molecules, the similarities and changes will be discussed when adsorbed on metals and graphene, respectively [2]. In the second part, the formation of chevron-like graphene nanoribbons (GNRs) using Ullmann-type coupling will be presented in dependence of the substrate used. [3] While for Cu(111) and Ag(110) 1D metal-coordinated polymers were obtained GNR formation was successful on Au(111). The electronic properties of the GNRs were observed to display both a length and symmetry dependence. References: [1] S. Maier et al., Beilstein J. Nanotechnol. 12 (2021) 950. [2] J. Li et al., J. Phys. Chem. C 123 (2019) 12730; J. de la Rie et al., J. Phys. Chem. C accepted. [3] T.A. Pham et al., Small 13 (2017) 1603675; R.S.K. Houtsma et al., submitted.

O 5.2 Mon 11:00 S051

Emergence of a singly-occupied state of *p*-terphenyl-based thiols bound to sulphur defects on MoS$_2$/Au(111) — •J. Rika Simon[1], Dmitrii Maksimov[2], Juan Pablo Guerrero Felipe[1], Paul Wiechers[1], Christian Lotze[1], Ana M. Valencia[3,4], Caterina Cocchi[3,4], Björn Kobin[4], Stefan Hecht[4], Mariana Rossi[2], and Katharina J. Franke[1] — [1]Freie Universität Berlin, Germany — [2]MPI for the Structure and Dynamics of Matter, Hamburg, Germany — [3]Carl von Ossietzky Universität Oldenburg, Germany — [4]Humboldt-Universität zu Berlin, Germany

The combination of transition-metal dichalcogenides (TMDCs) and organic molecules into hybrid inorganic-organic systems is a field gathering much interest in recent years. The use of submonolayers of the TMDC MoS$_2$ as a decoupling layer in an STM junction is already well established and allows highly resolved dI/dV spectra. But 2D materials also have drawbacks: Their properties are highly dependent on their local structure, because defects influence their properties severely. Here we utilise these defects by anchoring the thiol-based molecule CF3-3P-SH (trifluoromethyl-*p*-terphenyl-thiol) into purposely created top-layer sulphur point defects in MoS$_2$ on Au(111). One end-group of the anchored molecule is bound to the defect, allowing it to rotate around the anchoring point. On such molecules we observe a Kondo resonance. *Ab initio* molecular dynamics simulations show the emergence of a singly-occupied molecular state near E_F depending on the configuration of the molecule with respect to the surface, which in turn gives rise to the observed Kondo resonance.

O 5.3 Mon 11:15 S051

Electron spin resonance of iron-phthalocyanine molecules on a surface — •Christoph Wolf[1,2], Xue Zhang[2,3], Yu Wang[1,2], Philip Willke[4], Andreas J. Heinrich[1,5], and Taeyoung Choi[5] — [1]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic of Korea — [2]Ewha Womans University, Seoul, Republic of Korea — [3]Institute of Spin Science and Technology, South China University of Technology Guangzhou, China — [4]Physikalisches Institut, Karlsruhe Institute of Technology, Karlsruhe, Germany — [5]Department of Physics, Ewha Womans University, Seoul, Republic of Korea.

The combination of the high energy resolution of electron spin resonance (ESR) and high spatial resolution of the scanning tunneling microscope (STM) resulted in a novel probe with unparalleled capabilities for the study of surface physics.

In this work, we present the first application of this tool to molecules by ESR-STM spectroscopy of iron-phthalocyanine (FePc) molecules on thin layers of the insulator magnesium-oxide deposited on silver. Here, I will focus on the insight gained by combining density functional theory (DFT) and ESR-STM experiment. I will highlight successes and shortcomings of DFT by discussing electronic states of single FePc molecules as well as the interaction between FePc dimers at the neV energy scale.

Finally, I will give an outlook on quantum coherent properties of FePc based on pulsed ESR measurements and a non-equilibrium Green's function model.

O 5.4 Mon 11:30 S051

Mapping of resonant excitation channels of C$_{60}$/Cu(111) sample system through multiphoton polychromatic momentum microscopy — •Martin Mitkov[1], Ralf Hemm[1], Tobias Feuerbach[1], Aaron Gebert[1], Florian Haag[1], Ka Man Yu[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]TU Kaiserslautern and Research Center OPTIMAS, Erwin-Schrödinger Str. 46, 67663 Kaiserlsautern, Germany — [2]Institut für Physik, Johannes-Gutenberg-Universität Mainz, Mainz 55128, Germany

Mapping the excited states of organic molecular thin films is of crucial importance for understanding the optical and transport properties of organic materials. Here, we present a multiphoton photoemission study of the excited states of the C$_{60}$/Cu(111) interface for photon energies between 1.80 eV and 3.35 eV. Characteristic changes in the photoemission spectra for different photon energies allow us to determine the energetic positions of the excited states. In addition, we focus on the momentum-dependent photoemission pattern of the excited molecular states. The latter reveals distinct periodicities and symmetries based on the molecular orbitals involved in the optical excitation process. Altogether, this information will allow us to disentangle resonant intramolecular excitations from the Cu surface state. This allows us to uncover the photon energy-dependent excitation pathways of charge carriers at the C_{60}/Cu(111) interface.

O 5.5 Mon 11:45 S051

Disentangling the Complex Electronic Structure of an Adsorbed Nanographene: Cycloarene C108 — •Jose Martinez-Castro[1], Rustem Bolat[1], Qitang Fan[2], Simon Werner[2], Hadi Arefi[1], Taner Esat[1], Jörg Sundermeyer[2], Christian Wagner[1], J. Michael Gottfried[2], Ruslan Temirov[1,3], Markus Ternes[1,4], and F. Stefan Tautz[1] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Department of Chemistry, Philipps-Universität Marburg, 35032 Marburg, Germany — [3]II. Physikalisches Institut, Universität zu Köln, 50937 Köln, Germany — [4]II. Institute of Physics, RWTH Aachen University, D-52074 Aachen, Germany.

We combine low-temperature scanning tunneling spectroscopy, CO functionalized tips and algorithmic data analysis to investigate the electronic structure of the molecular cycloarene C108 (graphene nanoring) adsorbed on a Au(111) surface. We demonstrate that CO functionalized tips enhance the visibility of molecular resonances, both in differential conductance spectra and in real-space topographic images. Comparing our experimental data with ab-initio density functional theory reveals a remarkably precise agreement of the molecular orbitals and enables us to disentangle close-lying molecular states only separated by 50 meV at an energy of 2 eV below the Fermi level. We propose this combination of techniques as a promising new route for a precise characterization of complex molecules and other physical entities which have electronic resonances in the tip-sample junction.

O 5.6 Mon 12:00 S051

Tuning of structure of cyano-porphyrin self-assemblies on surfaces: from metals to bulk insulators — Maximilian Ammon[1], Mirunalini Devarajulu[1], Yi Liu[1], Martin Gurrath[2], Dominik Lungerich[3], •Binbin Da[1], Norbert Jux[3], Bernd Meyer[2], and Sabine Maier[1] — [1]Department of Physics, FAU Erlangen-Nürnberg, Erlangen, Germany. — [2]Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Erlangen, Germany. — [3]Lehrstuhl für Organische Chemie II, FAU Erlangen-Nürnberg, Erlangen, Germany.

We discuss the adsorption and binding motifs in self-assembled Zn-*p*CNTPP networks on Au(111), KBr(001), and MgO(001) using low-temperature STM and nc-AFM combined with DFT. Zn-CNTPPs adopt a planar adsorption geometry with the macrocycle parallel to the surface on all three surfaces and assemble in well-ordered islands. While a global minimum structure is found on KBr due to a strong CN$\cdots$K interaction, multiple and energetically nearly equivalent adsorption sites occur on MgO and Au. Therefore, commensurate adsorption is suggested on KBr, while optimizing the molecule-molecule interactions over molecule-surface interactions is more important on MgO and Au, which the STM and nc-AFM data experimentally evidence.[1] However, since the interaction of porphyrins with Au(111) is stronger than on MgO(001), the phenyl rings are more inclined toward the surface, resulting in larger unit cells on Au(111) and also the binding motif of the cyanophenyl groups changes.

[1] Ammon, M. et al., Surface Science, 723, 122097, 2022.

O 5.7 Mon 12:15 S051

Comparing adsorption of triphenylene derivatives: metallic vs. graphitic surfaces — •Joris de la Rie[1], Mihaela Enache[1], Qiankun Wang[1], Wenbo Lu[1], Nico Schmidt[1], Milan Kivala[2], and Meike Stöhr[1] — [1]Zernike Institute for Advanced Materials, University of Groningen — [2]Institute of Organic Chemistry, University of Heidelberg

Thin films of organic molecules show great promise for applications in future (nano)electronic devices, such as solar cells, light emitting diodes and transistors. A major factor in the performance of these films is the interface between substrate and molecular film. The interfacial properties depend on the balance between intermolecular and molecule-substrate interactions. Herein, we present a comparative study on self-assembled monolayers (SAMs) of a triphenylene-based donor molecule (HAT) on three substrates: Ag(111), graphene/Ir(111) and graphene/Ni(111). We studied the structure of the SAMs by means of scanning tunneling microscopy and low-energy electron diffraction, and their interaction with the substrates by X-ray and ultraviolet photoelectron spectroscopy. On each substrate, HAT formed a close-packed hexagonal network that is commensurate with the substrate. From the photoelectron spectroscopy measurements we only found a weak (physisorptive) interaction between molecules and substrates. This goes against the established belief for SAMs on metal surfaces, where commensurate networks are principally formed on strongly interacting surfaces where the molecules chemisorb.

O 5.8 Mon 12:30 S051

The Number of KCl layers counts: Thickness dependent Growth of Quinacridone on KCl on the Ag(100) surface — •Niklas Humberg[1], Rémi Bretel[2], and Moritz Sokolowski[1] — [1]Institut für Physikalische und Theoretische Chemie der Universität Bonn, Germany — [2]University of Paris-Saclay, Institut des Sciences Moléculaires D'Orsay, France

The epitaxial growth of the prochiral molecule quinacridone (QA) on a insulating KCl layer on the Ag(100) surface was investigated by STM. If QA is deposited onto a complete KCl layer, then the growth of QA structures is strongly dependent on the thickness of the KCl layer. For small amounts of QA on thin KCl layers small domains of parallel molecular chains were observed. The structure of these chains is identical to the chains that were previously observed on the Ag(100) surface. However, the repulsive substrate mediated interaction between neighboring chains that was observed on the Ag(100) surface cannot be observed on such KCl layers. Interestingly, depositing larger amounts of QA on such KCl layers leads to the formation of three-dimensional chain-like structures that show some similarities with the bulk crystal structures of QA.

Contrary to this, on thick KCl layers on the Ag(100) surface that are five or more monolayers high dewetting of the molecules and a formation of three-dimensional QA clusters was already observed for very small amounts of QA. These effects are explained by the interaction between the QA molecules and the silver substrate which still plays a role for thin layers but is suppressed by thick KCl layers.

O 5.9 Mon 12:45 S051

Growth of chiral heptahelicene molecules on ferromagnetic Co and Fe thin-film substrates — •Mohammad Reza Safari, Frank Matthes, Daniel E. Bürgler, and Claus M. Schneider — Peter Grünberg Institut (PGI-6), Forschungszentrum Jülich, 52425 Jülich, Germany

The discovery of chirality-induced spin selectivity effects [1,2], which result from an interaction between the electron spin and the handedness of chiral molecules, has sparked interest in surface-adsorbed chiral molecules due to potential applications in spintronics, enantioseparation, and chemical sensing. Here, we report growth studies of chiral heptahelicene molecules on two monolayers Fe on W(110), Co bilayer nano-islands on Cu(111), and for comparison on Cu(111) by low-temperature spin-polarized scanning tunneling microscopy. In all cases, the molecules remain intact, adsorb in a flat configuration, and exhibit specific in-plane orientations that reflect the symmetry of the substrate lattices. The Co and Fe layers remain out-of-plane magnetized after molecular adsorption. In addition, we are able to determine the helicity of individual molecules adsorbed on the highly reactive Fe and Co surfaces in a similar manner as previously reported for the less reactive Cu(111) surface [3]. These observations pave the way for further investigations of CISS effects in well-defined molecule-substrate systems on the single-molecule scale as a basis for theoretical modeling.

[1] B. Göhler *et al.*, Science **331**, 894 (2011). [2] K. Banerjee-Ghosh *et al.*, Science **360**, 1331 (2018). [3] K.-H. Ernst *et al.*, Nano Lett. **15**, 5388 (2015).

O 6: Nanostructures at Surfaces 1

Time: Monday 10:30–13:00 Location: S052

O 6.1 Mon 10:30 S052

Autonomous non-contact molecular manipulation of nanocars based on reinforcement learning — •Bernhard R. Ramsauer[1], Grant J. Simpson[2], Leonhard Grill[2], Oliver T. Hofmann[1], and Andreas Jeindl[1] — [1]Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria — [2]Institute of Chemistry, NAWI Graz, University Graz, Heinrichstraße 28/IV, 8010 Graz, Austria

At the world's first race of nanocars at the CEMES-CNRS, in France, participants had to direct a nanocar across a specific *racetrack* [1]. In order to control their nanocar, they have to manipulate it via an STM-tip, without being in direct contact with the nanocar. The physics that govern the molecule*s movement and rotation is complex and involves the interaction between the tip and the molecule as well as the molecule and the substrate [2]. Thus, it requires time and effort for humans to be able to maneuver a molecule with a reasonable success rate. However, predicting the outcome of a performed action is unintuitive and often hard to predict for humans. Therefore, we developed an artificial intelligence (AI) based on reinforcement learning (RL) and show how it can be implemented to manipulate single molecules. The AI utilizes an off-policy learning algorithm known as Q-Learning. Our results can be the basis for more sophisticated techniques of non-contact molecular manipulations. This allows to identify and manoeuvre single molecules at will, building the basis for future bottom-up constructions of nanotechnology. [1] G. Rapenne et al., Nature Rev. Mater. 2, 17040 (2017) [2] G. J. Simpson et al., Nature Nanotech. 12, 604 (2017)

O 6.2 Mon 10:45 S052

Constructing covalent organic nanoarchitectures molecule by molecule via scanning probe manipulation — Qigang Zhong[1,2], Alexander Ihle[1,2], Sebastian Ahles[2,3], Hermann A. Wegner[2,3], Andre Schirmeisen[1,2], and •Daniel Ebeling[1,2] — [1]Institute of Applied Physics, Justus Liebig University Giessen, Germany. — [2]Center for Materials Research, Justus Liebig University Giessen, Germany. — [3]Institute of Organic Chemistry, Justus Liebig University Giessen, Germany

Constructing low-dimensional covalent assemblies with tailored size and connectivity is challenging yet often key for applications in molecular electronics where optical and electronic properties of the quantum materials are highly structure dependent. We present a versatile approach for building such structures block by block on bilayer sodium chloride (NaCl) films on Cu(111) with the tip of an atomic force microscope, while tracking the structural changes with single-bond resolution. Covalent homo-dimers in *cis* and *trans* configurations and homo-/hetero-trimers were selectively synthesized by a sequence of dehalogenation, translational manipulation and intermolecular coupling of halogenated precursors. Further demonstrations of structural build-up include complex bonding motifs, like carbon-iodine-carbon bonds and fused carbon pentagons. This work paves the way for synthesizing elusive covalent nanoarchitectures, studying structural modifications and revealing pathways of intermolecular reactions.
Nature Chemistry 13, 1133-1139 (2021)

O 6.3 Mon 11:00 S052

Electronic and magnetic properties of an on-surface synthesized 2D metal organic framework — •Amina Kimouche[1,2], Roberto Robles[3], Noemi Contreras[1], Daniel Ruiz[1], and Aitor Mugarza[1,4] — [1]Institut Català de Nanociència i Nanotecnologia, Barcelona, Spain — [2]Institut für Physik und Astronomie, Universität Potsdam, Potsdam, Germany — [3]Centro de Fisica de Materiales CFM/MPC (CSIC-UPV/EHU), Donostia-San Sebastián, Spain — [4]ICREA Institució Catalana de Recerca i Estudis Avançats, Barcelona, Spain

2D MOFs constitute a new class of designer materials where the coexistence of Dirac electrons and flat bands can lead to rich physical phenomena and to the realization of quantum phases such as topological or quantum anomalous Hall insulators.

Following the concepts of coordination chemistry, based on a surface-assisted self-assembly of the metal and organic components, we have carried out synthesis of Iron-hexaiminotriphenylene (HITP) MOFs on Au(111) substrate and characterized their electronic structure using scanning tunnelling spectroscopy (STS). On the other hand, ab-initio calculations of the observed structures indicate the presence of strong ferromagnetic interactions that persist under the influence of the Au substrate, indicating that the interaction with the Au substrate stabilizes even further the FM state on Fe3(HITP)2. We relate such strong magnetic interactions to the formation of a radical spin at the ligand that mediate the inter-ionic interactions.

O 6.4 Mon 11:15 S052

Host guest chemistry and supramolecular doping in triphenylamine-based covalent frameworks on Au(111) — Christian Steiner[1], Lukas Fromm[2], Julian Gebhardt[2], Yi Liu[1], Alexander Heidenreich[2], Natalie Hammer[2], •Hexia Shi[1], Andreas Görling[2], Milan Kivala[3], and Sabine Maier[1] — [1]Department of Physics, Friedrich-Alexander Universität Erlangen-Nürnberg, Erlangen, Germany — [2]Department of Chemistry and Pharmacy, Friedrich-Alexander Universität Erlangen-Nürnberg, Erlangen, Germany — [3]Organisch-Chemisches Institut & Centre for Advanced Materials, Ruprecht-Karls-Universität Heidelberg, Heidelberg, Germany

We report the host-guest interaction in triphenylamine-based covalently-linked macrocycles and networks on Au(111) using low-temperature scanning tunneling microscopy in combination with density-functional theory. Triphenylamine precursors formed macrocycles and 2D networks featuring carbonyl- and hydrogen-functionalized pores, creating preferred adsorption sites for trimesic acid (TMA) and halogen atoms. TMA binds through hydrogen bonds to the carbonyl sites while halogens selectively adsorb between two carbonyl groups at Au hollow sites. Band structure calculations reveal that TMA adsorption reduces the electronic band gap of the triphenylamine covalent frameworks due to charge transfer, while the interaction of the halogens leads to a slight downshift of the bands.[1].

[1] Steiner, C. et al. *Nanoscale*, 2021, **13**, 9798-9807.

O 6.5 Mon 11:30 S052

Carbon-carbon coupling on inert surfaces via a radical deposition source - proof-of-concept, challenges and perspective — •Gianluca Galeotti[1] and Markus Lackinger[1,2] — [1]Deutsches Museum, Museumsinsel 1, 80538 Munich, Germany — [2]Technische Universität München, James-Franck-Str. 1, 85748 Garching, Germany

The realization of one-atom thin C-C-bonded nanostructures is an ongoing challenge of nanotechnology. Those are, however, almost exclusively synthesized on metal surfaces, taking advantage of the catalytic activity to lower the required temperature for monomer activation below the desorption threshold. The development of methods for the direct synthesis on inert and insulating surfaces would be a milestone in the field, enabling studies of nearly unperturbed covalent nanostructures with unique electronic properties, such as graphene nanoribbons and-conjugated 2D polymers. Here, we will describe the development of a Radical Deposition Source (RaDeS) for the direct deposition of radicals onto inert surfaces for subsequent coupling into C-C bonded polymers. The radicals are generated en route by indirect deposition of halogenated precursors through a heated reactive tube, where the dehalogenation reaction proceeds. As a model system for inert surfaces, we use Ag(111) passivated with a closed monolayer of chemisorbed iodine. We will first illustrate the proof-of-concept with the synthesis of poly-para-phenylene from iodinated terphenyl precursors. Subsequently, we show how this approach can be used to overcome limitations of the conventional on-surface synthesis imposed by desorption, opening additional pathways for the synthesis of nanostructures.

O 6.6 Mon 11:45 S052

Carbon-based low-dimensional materials from first principles — •Niklas Enderlein, Roland Gillen, Sabine Maier, and Janina Maultzsch — Friedrich-Alexander-Universität Erlangen-Nürnberg

Bottom-up synthesis of carbon-based networks from molecular precursors offers a promising path to material design, where the structural, electronic, optical and vibrational properties, in principle, can be tailored through the choice of the precursor structures and functionalization. Prominent examples for this method are chevron-type graphene nanoribbons with defined topology [1] and highly controllable nitrogen incorporation [2], or multifunctional nanoporous graphene [3]. Considering the vast number of possible carbon-based networks, density functional theory (DFT) is a powerful tool for the preselection and design of promising molecular precursors based on the computational prediction of their intrinsic physical properties.

In this spirit, we present the results of recent DFT simulations on potentially interesting carbon-based networks that lend themselves to on-surface bottom-up synthesis through Ullmann coupling of well-defined molecular precursors. We show how the electronic properties are significantly affected by the network topology and the structure of the molecular building blocks, potentially giving rise to novel phenomena. Our results inspire further efforts in the direction of molecular precursor design.

[1] Y. Lee et al., Nano letters 18.11, 7247-7253 (2018)
[2] C. Bronner et al., Angewandte Chemie 125.16, 4518-4521 (2013)
[3] C. Moreno et al., Science 360.6385, 199-203 (2018)

O 6.7 Mon 12:00 S052

Covalently linked molecules as 1D materials — •Samuel Vasconcelos and Michael Rohlfing — University of Münster

We performed first-principles calculations to address the problem of the formation and characterization of covalently linked structures with molecules as building blocks. We show that upon pressure a rehybridization process takes place which leads to one-dimensional compounds resembling nanothreads, in which carbon atoms are all 4-fold coordinated and possess remarkable mechanical properties. We show that for porphyrins, the resulting 1D nanostructures have metallic character. Moreover, in the case of porphyrin-metal complexes, we find that the covalently linked structures may be a platform for the stabilization of straight metallic wires. We extend the methodology for the kekulene family, that throughout the same processes forms sp3 nanotubes.

O 6.8 Mon 12:15 S052

Peierls distortion and charge density waves in novel exfoliable 1D materials — •Chiara Cignarella[1], Davide Campi[2], and Nicola Marzari[1] — [1]THEOS and MARVEL, EPFL, Lausanne, Switzerland — [2]Universita di Milano Bicocca, Milano, Italy

One-dimensional materials are extremely attractive due to their unique electronic properties and potentialities in next-generation applications. A high-throughput screening has provided a portfolio of more than 800 novel 1D/quasi-1D materials exfoliable from the 3D Van der Waals compounds, out of which we select a dataset of metallic chains as possible candidates for vias and interconnects. Often, their low-dimensional nature leads to dynamical instabilities in the form of Peierls distortions or charge density waves (CDW), which drive structural phase transitions at finite wavevectors. Here, we analyse the stability of this novel class of materials, identifying the reconstructed stable superstructure from the phonon modes. In order to get more insight into the mechanism of the CDW, we then investigate the nesting function and the critical role of the electron-phonon coupling, still unexplored in real quasi-1D systems.

O 6.9 Mon 12:30 S052

Emulating organic molecular orbitals with artificial atoms on a surface — •E. Sierda, D. Badrtdinov, B. Kiraly, E. J. Knol, X. Huang, M. I. Katsnelson, G. C. Groenenboom, D. Wegner, M. Rösner, and A. A. Khajetoorians — Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

Bottom-up strategies to emulate the orbital structure of organic compounds is an exciting prospect, especially for molecules that are complex, unstable or hard to isolate, e.g. cyclobutadiene or triangulene. A successful implementation of such an emulator requires creating and coupling artificial atoms with multi-orbital character and possibility for orbital hybridization, on a platform that does not couple to the emulated structure. We emulate the electronic structure of planar organic molecules, using coupled, bottom-up constructed quantum dots (QDs) composed of atomic ions. We illustrate that compact clusters of ions, created via atom manipulation, exhibit a well pronounced state, localized within the semiconductor bulk band gap. For a pair of such QDs, we observe two states and identify them as bonding and antibonding via spatial maps. Linear chains of QDs exhibit emulated linear combinations of atomic orbitals with both s- and p-like character. Furthermore, we construct artificial structures resembling sp2-hybridized organic molecules. A rich electronic structure with pronounced states is found in tunneling spectroscopy. By comparing their spatial maps with quantum-chemical simulations of the organic compound, we can identify the states as emulated organic orbitals, providing evidence for sp2 hybridization present in the artificial structures.

O 6.10 Mon 12:45 S052

Substrate-mediated polymorphism in monolayer self-assembly at liquid-solid interfaces — •Arash Badami Behjat[1], Wolfgang Heckl[1,2], Michael Schmittel[3], and Markus Lackinger[1,2] — [1]Technical University of Munich, James-Franck-Str. 1, 85748 Garching, Germany — [2]Deutsches Museum, Museumsinsel 1, 80538 Munich, Germany — [3]Universität Siegen, Adolf-Reichwein-Str. 2, 57068 Siegen, Germany

Scanning-Tunneling Microscopy (STM) studies of molecular self-assembly at liquid-solid interfaces require atomically flat, chemically inert, and electrically conductive substrates, hence are almost exclusively carried out on graphite. Here, we demonstrate that Au (111) passivated with a monolayer of chemisorbed iodine (I-Au(111)) constitutes a viable alternative for fundamental studies. By using aromatic homologues of tricarboxylic acids as a versatile model system, and by a direct comparison between graphite and I-Au (111), we experimentally study and demonstrate the decisive influence of molecule-surface interactions. We present three cases of a novel substrate-mediated polymorphism. On I-Au(111), we consistently find polymorphs with lower packing density and optimized intermolecular binding-a clear indication for diminished molecule-surface interactions. This hypothesis was corroborated by probing the monolayer's thermodynamic stability in variable temperature STM experiments.

O 7: New Methods and Developments 1: Scanning Probe Techniques 1 (joint session O/KFM)

Time: Monday 10:30–13:00 Location: S053

Topical Talk O 7.1 Mon 10:30 S053

Identification of active electrocatalytic centers using EC-STM under reaction conditions — •Aliaksandr Bandarenka — Technical University of Munich, Department of Physics, James-Franck-Str 1, 85748 Garching bei München, Germany

Identification of so-called active electrocatalytic centres can be very complicated under reaction conditions. In many cases, electrochemical scanning tunnelling microscopy can be efficiently used to do so by comparing the tunnelling noise in the presence and the absence of the electrocatalytic reactions. I the presentation, I will discuss examples, which deal with finding the active sites at the surface of various electrodes for hydrogen evolution, oxygen reduction, and oxygen evolution reactions. Pt, HOPG, Pt-alloys, and transition metal oxides are used as the model systems.

O 7.2 Mon 11:00 S053

Coherent Noise Removal for Scanning Probe Microscopy — Jens Oppliger, Danyang Liu, and •Fabian Donat Natterer — Department of Physics, University of Zurich, Winterthurerstrasse 190, CH-8057, Switzerland

Despite the best efforts to isolate the weak signals in scanning probe microscopes from sources of noise, white and coherent noise remain major nuisances. While the presence of Gaussian white noise can be handled with temporal averaging, the influence of high-Q coherent noise, such as coming from ground-loops or mechanical resonances, is less straightforward to delete when rastering along the surface. Such noise leads to characteristic streaks and spurious Bragg peaks in the Fourier transform of two-dimensional data. Here we demonstrate a straightforward method to remove coherent noise using data-labelling and exemplify its working for quasiparticle interference and topographic imaging.

O 7.3 Mon 11:15 S053

General, Strong Impurity-Strength Dependence of Quasiparticle Interference — •Seung-Ju Hong[1], Jae-Mo Lihm[1,2,3], and Cheol-Hwan Park[1,2,3] — [1]Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea — [2]Center for Correlated Electron Systems, Institute for Basic Science, Seoul 08826, South

Korea — [3]Center for Theoretical Physics, Seoul National University, Seoul 08826, South Korea

Quasiparticle interference patterns induced by impurities contain information about electronic structures in momentum space. In this presentation, we show that the interpretation of quasiparticle interference patterns is not trivial and needs special care. Even in the simple case of a single-site impurity on the square lattice, the pattern is strongly dependent on the strength of impurity potential. For example, the wave vector with the strongest scattering differs by about 16% in spin-dependent JDOS and exact QPI computations. We also showed that this dependence can be analyzed by decomposing the pattern into the impurity-dependent T-matrix part and momentum-dependent Green function part. We applied our formalism to TaAs, an archetype Weyl semimetal with first-principles calculations. We find that the strong dependence on impurities is also present in TaAs. Thus, our work demonstrates that these quasiparticle interference patterns must be analyzed with care and needs more attention.

Reference [1] S.-J. Hong, J.-M. Lihm, and C.-H. Park, J. Phys. Chem. C 2021, 125, 13, 7488-7494

O 7.4 Mon 11:30 S053

Real-space sub-femtosecond imaging of quantum electronic coherences in molecules — Manish Garg[1], •Alberto Martin-Jimenez[1], Michele Pisarra[2], Yang Luo[1], Fernando Martin[2,3,4], and Klaus Kern[1,5] — [1]Max Planck Institute for Solid State Research, Stuttgart, Germany — [2]Instituto Madrileño de Estudios Avanzados en Nanociencia (IMDEA Nano), Madrid, Spain — [3]Universidad Autonoma de Madrid (UAM), Madrid, Spain — [4]Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Madrid, Spain — [5]Institut de Physique, Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland

Tracking electron motion in molecules is the key to understand and control chemical transformations. Contemporary techniques in attosecond science have the capability to generate and trace the consequences of this motion in real time, but not in real space. Scanning tunnelling microscopy (STM), on the other hand, can locally probe the valence electron density in molecules, but cannot provide by itself dynamical information at this ultrafast time-scale. Here we show that, by combining STM and attosecond technologies, quantum electronic coherences induced in molecules by $<$ 6 femtosecond long carrier-envelope-phase (CEP) stable near-infrared laser pulses can be directly visualized with angstrom-scale spatial and sub-femtosecond temporal resolutions. We demonstrate concurrent real-space and real-time imaging of coherences involving the valence orbitals of perylenetetracarboxylic dianhydride (PTCDA) molecules, and full control over the population of the involved orbitals.

O 7.5 Mon 11:45 S053

Femtosecond Tip-Enhanced Coherent Anti-Stokes Raman Spectroscopy of a Single Graphene Nanoribbon — •Yang Luo[1], Alberto Martin-Jimenez[1], Manish Garg[1], and Klaus Kern[1,2] — [1]Max Planck Institute for Solid State Research, Stuttgart, Germany — [2]Institut de Physique, Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland

By integration of ultrashort laser pulses with a scanning tunneling microscope (STM) one can study the electronic and carrier dynamics with very high spatial and temporal resolution. Nevertheless, molecular vibrational modes at the single-molecule level are difficult to track, owing to the lack of energy resolution. To overcome this barrier, we have now integrated a local spectroscopic tool, combining ultrafast laser pulses with an STM-based tip-enhanced Raman spectroscopy (TERS). By performing TERS with femtosecond laser pulses, we have tracked vibrational coherences and phonon dephasing dynamics in a single graphene nanoribbon (7-GNR). The decoherence time (T2/2 ~ 440 fs) of the phonons in a GNR has been obtained from the time-resolved coherent anti-Stokes Raman spectra. Temporal evolution of vibrational coherences (beatings) between different phonon modes in the GNR has been measured, which evolve on time scales as short as ~ 100 fs. This work lays the foundation for investigating intramolecular vibrational coherences and vibronic dynamics with utmost spatial, temporal and energy resolutions, simultaneously.

O 7.6 Mon 12:00 S053

Coherent phonon spectroscopy on the nanoscale — Shuyi Liu[1], Adnan Hammud[1], Ikutaro Hamada[2], Martin Wolf[1], •Melanie Müller[1], and Takashi Kumagai[3] — [1]Fritz-Haber-Institut, Berlin, Germany — [2]College of Materials Science and Engineering, Hunan, China — [3]Institute for Molecular Science, Okazaki, Japan

Coherent phonon (CP) spectroscopy is a powerful tool to monitor ultrafast lattice dynamics under nonequilibrium conditions, providing insight into microscopic interactions that dictate macroscopic material properties. In imperfect crystals, the excitation and relaxation of CPs will be susceptible to the nanoscale environment, calling for real-space observation of ultrafast lattice dynamics. We demonstrate nanoscale coherent phonon spectroscopy by means of ultrafast laser-induced scanning tunneling microscopy (STM) in a plasmonic junction. Comparison of the CP spectra with tip-enhanced Raman spectroscopy allows us to identify the involved phonon modes. In contrast to the Raman spectra, the relative CP intensities exhibit strong nanoscale spatial variations, which correlate with changes in the local density of states recorded via scanning tunneling spectroscopy. Our work introduces a new approach to study the ultrafast structural response at solid surfaces using optical STM.

O 7.7 Mon 12:15 S053

Construction of a dry low temperature STM — •Simon Gerber[1] and Wulf Wulfhekel[2] — [1]Physikalisches Institut, Karlsruhe Institute of Technology — [2]Physikalisches Institut, Karlsruhe Institute of Technology

Driven by rising helium prices, we design a dry, low temperature Scanning Tunneling Microscope with a closed helium cycle. We designed a compact dry four stage cryostat with an integrated dilution refrigerator which is cooled using helium from a 400 mW cold head. The STM is connected to the dilution refrigerator and allows measurements down to millikelvin temperatures. The system is mechanically decoupled at several points to minimize vibrations from the cold head reaching the STM. The microscope is positioned inside a split-coil magnet with magnetic fields up to 4T. Optical Access to the STM is possible in the parked position and allows fast tip and sample exchange. The tips and samples can then be prepared under UHV conditions. The complete cryostat and and the STM are home-built.

O 7.8 Mon 12:30 S053

Probing tunneling processes into YSR states with microwaves — •Janis Siebrecht[1], Haonan Huang[1], Piotr Kot[1], Sujoy Karan[1], Ciprian Padurariu[2], Björn Kubala[2], Joachim Ankerhold[2], Alfredo Levy Yeyati[3], Juan Carlos Cuevas[3], and Christian R. Ast[1] — [1]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany — [2]Institut für Komplexe Quantensysteme and IQST, Universität Ulm, Ulm, Germany — [3]Departamento de Física Teórica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Madrid, Spain

Microwaves are an important tool in the manipulation of multi-level systems such as single spins on a surface, nitrogen vacancies in diamond or double quantum dots. Here we use a scanning tunneling microscope (STM) at a base temperature of 0.56 K to probe the intrinsic YSR states in a Vanadium tip in contact with a V(100) surface. The addition of an E-Band (60-90 GHz) microwave antenna at the junction opens the possibility to study the behavior of YSR states with AC driving- a scenario which has been subject to many theoretical but very few experimental studies. Using microwave-assisted tunneling, we gain insight into how the excited state participates in the tunneling process and how this is related to Andreev processes and parity conservation. Our results point at a new path, namely microwave manipulation of YSR states, which could be an important step towards using YSR states as qubits.

O 7.9 Mon 12:45 S053

Compressed fingerprint spectroscopy based on scanning microscopy — •Bernd Kästner[1], Manuel Marschall[1], Arne Hoehl[1], Andrea Hornemann[1], Gerd Wübbeler[1], Selma Metzner[1], Piotr Patoka[2], Eckart Rühl[2], and Clemens Elster[1] — [1]Physikalisch-Technische Bundesanstalt, Berlin, Germany — [2]Freie Universität Berlin, Germany

The infrared spectral region between 400 and 4000 cm^{-1} is called the fingerprint region, because the absorption features are unique to individual organic substances. Such a spectrum usually contains many peaks, making it difficult to link individual peaks to the substance. Consequently, the spatial mapping of substances requires spectral imaging, where at each point in space a complete spectrum needs to be recorded. Usually this can be achieved by spectrometers equipped with array detectors. Recently, scanning methods based on the optical nearfield and local thermal expansion with nanoscale spatial resolution have been developed allowing sub-diffraction spectral imaging. However, the inherently serial recording severely limits their imaging application due to long acquisition times involved and the resulting stability issues. In this work we demonstrate different strategies to significantly reduce the measurement time in spectral imaging measurement by compressing the measurement combined with a low-rank matrix reconstruction. Several examples from different fields of application will be discussed.

O 8: Solid-Liquid Interfaces 1: Reactions and Electrochemistry

Time: Monday 10:30–12:45 Location: S054

Topical Talk O 8.1 Mon 10:30 S054

Dynamic structure changes of bare and modified Cu(111) during CO and water activation — •Andrea Auer[1], Nicolas Hörmann[2], Mie Andersen[3], Karsten Reuter[2], and Julia Kunze-Liebhäuser[1] — [1]Institute of Physical Chemistry, University of Innsbruck, Austria — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [3]Aarhus Institute of Advanced Studies, Denmark

CO is a key intermediate in the electro-oxidation of energy carrying fuels. Single-crystal Cu(111) model catalysts efficiently electro-oxidize CO in alkaline media, under strong and continuous surface structural changes that lead to simultaneous strengthening of the CO and weakening of the OH binding, which makes the observed high activity possible.

Cu(111) modified with $Ni(OH)_2$ and $Co(OH)_2$ reveals strong morphological changes upon adatom deposition, which lead to a significant enhancement in the rate of the alkaline hydrogen evolution reaction (HER), one of the most important processes in the development of hydrogen-based energy conversion devices. Adatom modification influences the charge distribution at the solid/liquid interface by a decrease of the electric field strength negative of the potential of zero charge. This implies an easier reorganization of the interfacial water molecules facilitating charge transfer trough the double layer. The tendency of Cu(111) to restructure is found to dominate its electrochemical properties. The structural changes of the electrode surface are intimately related to the electric field at the solid/liquid interface and to its electrocatalytic activity, in general.

O 8.2 Mon 11:00 S054

Cu(111) reconstruction and oxidation imaged in oxygen free alkaline media with electrochemical scanning tunneling microscopy — •Toni Moser, Andrea Auer, and Julia Kunze-Liebhäuser — Department of Physical Chemistry, University of Innsbruck, Innrain 52c, Innsbruck, Austria

Cu has recently gained attention due to its capability to efficiently oxidize CO at low overpotentials[1]. However, the structural evolution of the surface at anodic potentials causing initiation of oxide formation under complete exclusion of oxygen has yet to be fully understood. While Cu(111) oxidation has previously been studied with electrochemical scanning tunneling microscopy (EC-STM), investigations without atmospheric oxygen and without contamination from dissolved glassware in alkaline media are sparse. Recent results indicate a significant delay in the anodic formation of Cu_2O in completely deaerated alkaline electrolyte, which indicates a significant impact of dissolved O_2 on the oxidation of copper surfaces. In this work, we focus on the in-situ investigation of hydroxide adsorption on and oxidation of Cu(111) via EC-STM, eliminating atmospheric oxygen by conducting the experiments inside an Ar-filled glove box. The delayed formation of an amorphous oxide layer at anodic potentials and subsequent reduction processes at cathodic potentials, where a smoothening of the surface can be observed, as well as new insights into the hydroxide adsorption structure are presented.

[1] A. Auer, M. Andersen, E.-M. Wernig, N. G. Hörmann, N. Buller, K. Reuter & J. Kunze-Liebhäuser, Nat Catal 3, 797-803 (2020).

O 8.3 Mon 11:15 S054

Enhanced Field Effects at Protruding Defect Sites in Electrochemistry? – A Theoretical Evaluation — •Simeon D. Beinlich[1,2], Nicolas G. Hörmann[1], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Technical University of Munich, Munich, Germany

Does electrochemistry at protruding surfaces sites differ significantly from that at ideal low-index surfaces? Intuitively, classical electrostatics suggest a local field enhancement at protruding sites. In this case, a dipole-field-like picture would suggest a pronounced potential-dependence of adsorption energies at such sites.

Here, we evaluate these dependencies for various adsorbates on vicinal Pt(111) surfaces using first-principles calculations in combination with a fully grand canonical approach [1]. Our results show an enhancement of the local electric field at pristine surfaces. However, it is lifted upon adsorption and hence does not cause the anticipated stronger field-effects. Nevertheless, we observe dramatic variations in the potential-dependence which can be rationalized from the differences in surface dipoles that form upon adsorption. These correlate with site coordination showing a consistent trend across adsorbates and adsorption sites.

We rationalize these findings and discuss how the adsorption behavior changes on defect-rich, undercoordinated surfaces in an electrochemical environment.

[1] S.D. Beinlich *et al.*, ACS Catal. **12** 6143–6148 (2022)

O 8.4 Mon 11:30 S054

Vapor adsorption on carbon nanomembranes (CNMs) — •Enneita Khayya, Petr Dementyev, and Armin Gölzhäuser — Faculty of Physics, Bielefeld University, 33615 Bielefeld, Germany

Intrinsically porous carbon nanomembranes (CNMs) demonstrate promising mass transfer properties with respect to separation of liquids and gases. To complement the permeation studies with CNMs, we introduce a spectroscopic experiment for probing adsorption of vaporous substances on their surface under ambient conditions. Polarization-modulation infrared reflection absorption spectroscopy (PM IRAS) is used to quantify the number of adsorbed species with the help of innovative calibration approaches, including azeotropic mixtures and immobilization in a polymer matrix. Water and alcohols are found to readily condense on supported CNMs yielding liquid-like interfaces. The results are consistent with the vapor permeation rates measured in free-standing CNMs.

O 8.5 Mon 11:45 S054

Investigating Zirconium Nitride Cathodes for the Electrochemical Nitrogen Reduction Reaction — •Teodor Apetrei, Saswati Santra, Verena Streibel, and Ian D. Sharp — Walter Schottky Institut, Technische Universität München, Garching, Germany

The electrochemical nitrogen reduction reaction (NRR) can convert nitrogen to ammonia at ambient conditions. The most critical factor to activate N2 is to cleave the N-N triple bond. Transition metal nitrides have been proposed as electrocatalysts for the NRR, with computational studies predicting that N-N bond cleavage can be facilitated via a Mars-van-Krevelen mechanism. Herein, we experimentally test this prediction by investigating sputter-deposited ZrN thin films for the NRR. Our investigations indicate that small amounts of ammonia are indeed produced when pristine ZrN thin films are used as NRR cathodes. However, we also observe nitrogen loss and electrochemical instabilities, which could indicate a sacrificial rather than catalytic role of ZrN. Hence, to accelerate the rate-limiting N-N bond splitting step and facilitate nitrogen replenishment, we anchor Fe SACs onto ZrN. Our preliminary results indicate that high-temperature attachment of Fe SACs leads to overall smaller current densities. However, electrochemical measurements and comparative XPS and XRD studies of the pre- and post-NRR samples suggest an increased nitrogen stability within the Fe-modified ZrN films and improved electrochemical stability. Whether this Fe-modification also facilitates N-N bond dissociation and boosts the NRR activity is currently investigated.

O 8.6 Mon 12:00 S054

Towards Understanding Platinum Degradation: Modelling the Growth of Nanoislands — •Francesc Valls Mascaro[1], Marc T.M. Koper[1], and Marcel J. Rost[2] — [1]Leiden Institute of Chemistry, Leiden University — [2]Huygens-Kamerlingh Onnes Laboratory, Leiden Institute of Physics, Leiden University

Platinum is the catalyst of choice in many electrochemical energy conversion systems like fuel cells due to its superior activity. However, the stability of platinum catalysts is limited under fuel cell operation conditions. This degradation process has been extensively studied by cyclic voltammetry and inductively coupled plasma mass spectrometry (ICP-MS) [1, 2]. The origin of this degradation is most likely linked to the roughening of the surface due to the nucleation and growth of platinum nanoislands [3, 4, 5, 6]. In this work, we model, fully analytically and with the support from electrochemical data, the growth of these nanoislands on Pt(111) in perchloric acid. The model here presented successfully describes the surface growth taking place during the oxidation-reduction cycling. Different parameters such as the flux of adatoms and vacancies were obtained from the fittings between the model and the experimental data.

References: [1] Topalov, A. et al., Chem. Sci., 5, 631 (2014) [2] Sandbeck D.J.S. et al., ACS Appl. Mater. Interfaces, 12, 25718 (2020) [3] Jacobse, L. et al., Nat. Mater. 17, 277 (2018) [4] Jacobse, L. et al., ACS Cent. Sci. 5 (12), 1920 (2019) [5] Rost, M.J. et al., Nat. Commun. 10, 5233 (2019) [6] Ruge, M. et al., J. Am. Chem. Soc., 139, 4532 (2017)

O 8.7 Mon 12:15 S054

Exploring the Limits of Mean-Field Theory in Modeling Thermodynamic Cyclic Voltammograms — •Nicolas Bergmann, Nicolas G. Hörmann, and Karsten Reuter — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Deutschland

Mean-field theory (MFT) is at the heart of many approaches to simulate materials. Recently, MFT has been used to model thermodynamic cyclic voltammograms (CVs) [1], a standard electrochemical experiment. However, its accuracy limitations in this context remain unclear.

Here, we outline our general ansatz to derive mean field models for thermodynamic CVs based on *ab initio* DFT calculations of a wide variety of adsorbate configurations [1]. To derive continuous MFT expressions, we use nonparametric Gaussian process regression. We apply our method to assess the fingerprint CV of Cu(100) in iodine-containing, alkaline solutions. The simulations offer new insights into the competitive adsorption between I and OH. Additionally, we benchmark our method by comparing a mean-field model to grand canonical lattice Monte Carlo simulations for the well-studied system Ag(100) in Br-containing electrolyte [2,3].

We analyze in detail the respective (dis-)advantages of both methods.

[1] N.G. Hörmann *et al.*, *J. Chem. Theory Comput.* **2021**, 17, 1782
[2] M.T.M. Koper, *J. Electroanal. Chem.* **1998**, 450, 189-201
[3] M. Nakamura *et al.*, *Phys. Rev. B* **2011**, 84, 165433

O 8.8 Mon 12:30 S054

Neural network surrogates for kinetic Monte Carlo models of electrocatalytic surfaces — •Younes Hassani Abdollahi[1,2], Jürgen Fuhrmann[3], and Sebastian Matera[1,2] — [1]Institut f. Mathematik, Freie Universität Berlin, Arnimallee 6, 14195 Berlin, Germany — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — [3]Weierstraß-Institut f. Angewandte Analysis u. Stochastik, Mohrenstr. 39 10117 Berlin, Germany

The kinetic Monte Carlo method (kMC) is the physically most sound approach for addressing the kinetic interplay of elementary processes at electrocatalytic surfaces but also comes at high computational costs. Therefore, computationally efficient surrogate models are highly desirable which allow the utilization of kMC simulation results in coarser scale simulations.

Using the oxygen reduction reaction on Pt(111) as a prototypical example, we investigate regression neural networks as surrogates to reproduce the stationary TOF as a function of all reaction conditions, i.e. electrostatic potential, concentrations, and temperature. We found that a relatively shallow perceptron with 2 layers of 32 and 32 neurons, respectively, and SiLU activation functions serve as an appropriate choice. We demonstrate the performance of this model with a varying number of kMC data points. Finally, we discuss how this model can be incorporated into a multiscale modeling approach, which addresses the interaction of transport and kinetics.

O 9: Ultrafast Electron Dynamics at Surfaces and Interfaces 2

Time: Monday 15:00–18:15 Location: H3

O 9.1 Mon 15:00 H3

Two distinct 4f states in mixed-valent TmSe$_{1-x}$Te$_x$ — •Chul hee Min[1], Michael Heber[2], Simon Müller[3], Lukas Wenthaus[2], Markus Schoz[2], Dmytro Kutnyakhov[2], Lenart Dudy[4], Hendrik Bentmann[3], Federico Pressacco[2], Matthias Kalläne[1], Woojae Choi[5], Yong Seung Kwon[5], Friedrich Reinert[3], and Kai Rossnagel[1] — [1]IEAP, CAU Kiel, Germany — [2]DESY, Hamburg, Germany — [3]EP7 and ct.qmat, University of Würzburg, Germany — [4]Synchrotron SOLEIL, Saint-Aubin, France — [5]Dep. of EMS, DGIST, South Korea

From the strong electron correlation effects in rare earth compounds, rich phase diagrams emerge with tunable ground states, which underlie a series of unique physical phenomena and quantum states, including quantum criticality, topological Kondo insulators, and diverse charge-neutral quasiparticle formations. However, despite their importance, the nature of such composite quasiparticles is difficult to characterize because all coherent spectral features develop in a similar way. Using angle-resolved photoemission spectroscopy (ARPES) together with time-resolved PES, we have addressed distinct coherent 4f features in unique mixed-valence TmSe$_{1-x}$Te$_x$. Our findings open the path for future investigations of small energy-scale excitations and may provide a framework for understanding the dynamics and entangled nature of correlated electrons.

O 9.2 Mon 15:15 H3

Time- and angle-resolved photoemission study of magnetic topological insulators MnBi$_2$Te$_4$ and MnBi$_4$Te$_7$ — •Paulina Majchrzak[1], Klara Volckaert[1], Deepnarayan Biswas[1], Denny Puntel[2], Wibke Bronsch[2], Federico Cilento[2], Xing-chen Pan[3], Yong Chen[1,3], and Søren Ulstrup[1] — [1]Dept. of Physics and Astronomy, Aarhus University, DK — [2]Elettra - Sincrotrone Trieste, IT — [3]Advanced Institute for Materials Research, Tohoku University, JP

Van der Waals heterostructures comprising layers of intrinsically antiferromagnetic topological insulator (TI) MnBi$_2$Te$_4$ and non-magnetic TI Bi$_2$Te$_3$ offer a rich toolbox for engineering exotic quantum phenomena. Magnetic and transport properties of these materials are strongly affected by the interplay between bulk and surface states with divergent topologies resulting from hybridisation between the top layers.

Here, we disentangle those complex interactions in the time domain with TR-ARPES. We discuss the interband dynamics in bulk and surface states as a function of stacking and surface terminations for MnBi$_2$Te$_4$ and MnBi$_4$Te$_7$. Our results fill a knowledge gap in understanding of interlayer coupling in MnBi$_2$Te$_4$-based heterostructures.

O 9.3 Mon 15:30 H3

Electronic and phonon dynamics in 1T-TiSe$_2$ with ultrafast core-level transient absorption spectroscopy in the extreme ultraviolet — •Tobias Heinrich[1], Hung-Tzu Chang[1], Sergey Zayko[1], Murat Sivis[1,2], and Claus Ropers[1,2] — [1]Max Plank Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute - Solids and Nanostructures, University of Göttingen, Germany

Extreme ultraviolet (XUV) transient absorption spectroscopy with high harmonic sources is ideally suited to investigate the complex interplay between lattice and electronic degrees of freedom on ultrafast timescale [1]. Here, we compare the experimental result of highly sensitive XUV transient absorption spectroscopy on the charge density wave (CDW) compound 1T-TiSe2 with density functional theory (DFT) simulations to disentangle electronic and phonon contributions to the transient absorption spectra. In addition to photo excited carriers we observe two coherently excited phonon modes that can be assigned to the A_{1g} optical mode (6 THz) and the A_{1g}* (3.3 THz) amplitude mode associated with the CDW formation [2]. The modes show distinct spectral fingerprints which are reproduced by DFT calculations such that their dynamics can be individually analyzed. It is found that only the amplitude mode vanishes at higher fluences, corroborating the proposed mechanism of non-thermal CDW melting [3].

[1] A. R. Attar *et al.*, ACS Nano **14**, 11, 15829-15840 (2020)

[2] H. Hedayat *et al.*, New J. Phys. **23**, 033025 (2021)
[3] E. Möhr-Vorobeva *et al.*, Phys. Rev. Lett. **107**, 036403 (2011)

O 9.4 Mon 15:45 H3

Coherent phonon-driven transient modulation of a Dresselhaus-type spin splitting in *Td*-WTe$_2$ — Petra Hein[1], Stephan Jauernik[1], Hermann Erk[1], Lexian Yang[2,3], Yangpen Qi[4,5], Yan Sun[5], Claudia Felser[5], and •Michael Bauer[1] — [1]Institute of Experimental and Applied Physics, University of Kiel, Germany — [2]State Key Laboratory of Low Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing, China — [3]Frontier Science Center for Quantum Information, Beijing, China — [4]School of Physical Science and Technology, ShanghaiTech University, China — [5]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany

Time- and angle-resolved photoemission spectroscopy is used to study transient changes of the electronic structure in the Weyl-semimetal *Td*-WTe$_2$ in response to the excitation of coherent phonons. A Fourier-transform of the three-dimensional experimental data yields phonon-mode resolved insights into such coupling processes. Results of our analysis reveal a transient modulation of a Dresselhaus-type spin splitting of electronic bands that is selectively driven by the excitation of an interlayer shear mode of the layered compound [1]. The results provide real-time insights into electron-phonon coupled processes that are of vital importance for a light-driven topological phase transition in Td-WTe$_2$.

[1] P. Hein, et al., Nat. Commun. 11, 2613 (2020).

O 9.5 Mon 16:00 H3

Coherent Control of a Metastable Hidden Phase — J. Maklar[1], S. Dong[1], •J. Sarkar[1], Y. A. Gerasimenko[2], T. Pincelli[1], S. Beaulieu[1], P. S. Kirchmann[3], J. A. Sabota[3], S.-L. Yang[3,4], D. Leuenberger[3,4], R. G. Moore[3], Z.-X. Shen[3,4], M. Wolf[1], D. Mihailovic[2], R. Ernstorfer[1,5], and L. Rettig[1] — [1]Fritz-Haber-Institut der MPG, Berlin, DE — [2]Jožef Stefan Institute, Jubljana, Sl — [3]SLAC National Accelerator Laboratory, California, USA — [4]Stanford University, California, USA — [5]Technical University Berlin, DE

In materials with multiple competing orders, ultrashort light pulses can induce metastable states that are not accessible at thermodynamic equilibrium. One of such, the metallic hidden (H) phase of 1T-TaS$_2$, is of particular interest as it features an order-of-magnitude change in resistivity, promising for novel energy-efficient high-speed memory devices. We use time- and angle-resolved photoemission spectroscopy (trARPES) to investigate the electronic band structure and formation dynamics of the metastable H-state in 1T-TaS$_2$. The band structure mapping of H-state reveals suppression of correlation effects and metallization, suggesting a critical role of interlayer stacking order of the TaS$_2$ sheets in the phase transition. The fluence-dependent dynamics provides strong evidence that the charge density wave amplitude mode governs a collective, ultrafast switching pathway to the H-state. This is further corroborated by demonstrating coherent control of the switching efficiency into the H-phase using a multi-pump-pulse excitation scheme.

O 9.6 Mon 16:15 H3

Influence of carbon buffer layer on non-equilibrium carrier dynamics of epitaxial graphene and WS2/graphene heterostructures — •Lukas Bruckmeier[1], Niklas Hofmann[1], Leonard Weigl[1], Johannes Gradl[1], Neeraj Mishra[2,3], Stiven Forti[2], Camilla Coletti[2,3], and Isabella Gierz[1] — [1]Faculty for Physics, University of Regensburg, Regensburg, Germany — [2]Center for Nanotechnology Innovation@NEST, Istituto Italiano di Tecnologia, Pisa, Italy — [3]Graphene Labs, Istituto Italiano di Tecnologia, Genova, Italy

The non-equilibrium photocarrier dynamics of epitaxial graphene on SiC(0001) have been studied in detail in the past. The fact that the graphene layer rests on a second two-dimensional carbon buffer layer (BL) - the well-known $(6\sqrt{3}\times 6\sqrt{3})$R30° reconstruction - was believed to be of minor importance for the interpretation of the time-resolved data. We use time- and angle-resolved photoemission spectroscopy to show that photoexcitation of the graphene/BL heterostructure is followed by a short-lived transient decrease in binding energy of the Dirac cone indicating a transient charging of the graphene layer with excess electrons. We attribute the transient n-doping of the graphene layer to the resonant excitation of a direct electronic transition between the non-dispersive states of the BL located 0.5eV below the Fermi level and the Dirac cone. We further show that this direct electronic transition affects the charge transfer dynamics in epitaxial WS2/graphene/BL heterostructures.

O 9.7 Mon 16:30 H3

Ultrafast phonon thermalization in a monolayer crystal — •Hyein Jung — Fritz-Haber-Institut der MPG

Understanding energy flow in semiconductors following a perturbation is key for future applications. Observing dynamic processes on their fundamental time scales enables studying energy transfer processes between intrinsic subsystems, and subsequently understanding the coupling between them. Here we study ultrafast lattice dynamics in monolayer WSe2, carried out using femtosecond (high-energy) electron diffraction (FED).

We studied the lattice response to laser excitation by probing laser-induced variations in Bragg peak intensities, which reflect changes in incoherent lattice vibrations (quantified by atomic mean squared displacements, MSD). We implement a novel approach to analyze such diffraction data, by which we disentangle element-specific vibrational responses in the sub-picosecond time domain.

Through this analysis, we observe a series of steps in the evolution of the lattice response through varying trends of the two elements (W, Se). These observations suggest a cascade of electron-phonon and phonon-phonon scattering processes occurring on short picosecond time scales. We interpret these by means of energy transfer between phonon groups using on ab-initio calculations of the partial phonon density of states phonon. These results demonstrate that our element-specific approach enables a deeper understanding of the cascade of e-ph and ph-ph energy transfer processes following excitation.

O 9.8 Mon 16:45 H3

k-dependent band gap renormalization in monolayer WS2 revealed by tr-ARPES — •Niklas Hofmann[1], Alexander Steinhoff[2], Leonard Weigl[1], Johannes Gradl[1], Tim Wehling[2,3], Sivan Refaely-Abramson[4], Neeraj Mishra[5,6], Stiven Forti[5], Camilla Coletti[5,6], and Isabella Gierz[1] — [1]University of Regensburg, 93040 Regensburg, Germany — [2]University of Bremen, 28359 Bremen, Germany — [3]University of Hamburg, 22607 Hamburg, Germany — [4]Weizmann Institute of Science, 7610001 Rehovot, Israel — [5]Center for Nanotechnology Innovation @NEST, Istituto Italiano di Tecnologia, 56127 Pisa, Italy — [6]Graphene Labs, Istituto Italiano di Tecnologia, 16163 Genova, Italy

Monolayer transition-metal dichalcogenides show strong enhancement of Coulomb interactions due to their reduced dimensionality with immediate effects on both the optical as well as the single-particle band gap. Photogenerated electron-hole pairs have been shown to result in a giant band gap renormalization that has been attributed to efficient screening of the Coulomb interaction. The corresponding band structure changes are predicted to show a pronounced momentum dependence that we resolve using time- and angle-resolved photoemission spectroscopy on monolayer WS2 supported by a graphene substrate. Excellent agreement with ab initio calculations allows us to disentangle the intricate interplay of different many-body contributions to the observed transient band structure renormalization with important implications for the optoelectronic properties of 2D semiconductors.

O 9.9 Mon 17:00 H3

Pump helicity-dependent anisotropic population dynamics in the topological insulator Sb2Te3 — •Jan Böhnke[1], Haydar Altug Yildirim[1,3], Stephan Schmutzler[1], Jaime Sánchez-Barriga[2], Oliver Rader[2], Cornelius Gahl[1], and Martin Weinelt[1] — [1]Fachbereich Physik, Freie Universität Berlin, Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany — [3]Leibniz-Institut für Astrophysik, Potsdam, Germany

The specific spin texture of Dirac cone like topologically protected surface states (TSS) and the long electron mean free path allow for spin-polarized currents at the surface of topological insulators. Optical control of such currents has been discussed controversially.

We investigated the role of direct and indirect population channels for the unoccupied TSS on Sb2Te3 in 2D momentum space by time and angle-resolved two-photon photoemission spectroscopy. Excitation with 1.55 eV photons leads to an initially anisotropic population of the Dirac cone, dependent on the helicity of the excitation pulse and the azimuthal orientation of the sample. The strongest anisotropy is found in the energy range of the warped Dirac cone. It exhibits predominantly a 3-fold symmetry originating from the symmetry group of the bulk material. This contribution accordingly does not correspond to a macroscopic current in the TSS. On a time scale of 100 fs the population anisotropy is masked by electrons scattering from the bulk conduction band into the TSS.

O 9.10 Mon 17:15 H3

Spatio-temporal imaging of bright and dark excitonic quasiparticles in twisted TMD heterostructures — •Jan Philipp Bange[1], David Schmitt[1], Wiebke Bennecke[1], AbdulAziz AlMutairi[2], Guiseppe Meneghini[3], Daniel Steil[1], R. Thomas Weitz[1], Sabine Steil[1], G. S. Matthijs Jansen[1], Samuel Brem[3], Ermin Malic[3], Stephan Hofmann[2], Marcel Reutzel[1], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Germany — [2]Department of Engineering, University of Cambridge, U.K. — [3]Fachbereich Physik, Philipps-Universität Marburg, Germany

In two-dimensional van-der-Waals semiconductors, the weak Coulomb screening of charge carriers leads to exciting new material properties, such as bright and dark excitons with large binding energies. Consequently, when creating a heterostructure from two transition-metal dichalcogenide (TMD) monolayers with a type II band alignment, interlayer excitons can be formed [1]. Because dark excitons are not directly accessible with optical techniques, the spatial and lateral dynamics on the fundamental nanometer length scale remain largely unexplored. How do dark and interlayer quasiparticles form, relax and diffuse in the presence of a heterojunction, stress fields and inhomogeneities? Here, we address this question using time-resolved momentum and dark-field photoemission microscopy which enables us to study the ultrafast formation dynamics of different excitonic species in twisted WSe$_2$/MoS$_2$ heterostructures.

[1] Schmitt *et al.*, arXiv2112.05011 (2021).

O 9.11 Mon 17:30 H3

SHG imaging microscopy of ultrafast charge-transfer dynamics in twisted TMDC heterostructures — •Marleen Axt, Jonas E. Zimmermann, Gerson Mette, and Ulrich Höfer — Fachbereich Physik, Philipps Universität Marburg, Germany

Two-dimensional heterostructures of transition metal dichalcogenides (TMDC) represent very well-defined model systems of van-der-Waals interfaces. Many material combinations feature a type-II band alignment, which can separate photoexcited electrons and holes into different layers through ultrafast charge transfer leading to the formation of so-called interlayer excitons or interface excitons.

We investigate the ultrafast charge-transfer dynamics in TMDC heterostructures as a function of the stacking angle using time-resolved second-harmonic generation (SHG) imaging microscopy. This experimental technique combines the advantages of SHG with high temporal and spatial resolution. For differently twisted MoS$_2$/WSe$_2$ heterostructures the electron transfer from WSe$_2$ to MoS$_2$ after resonant excitation (1.70 eV) was found to depend considerably on the twist angle. The transfer time is reduced from 85 fs down to 12 fs when going from a larger rotational mismatch (16°) towards 2H-stacking (52°).

O 9.12 Mon 17:45 H3

Spin and charge carrier dynamics at a CuPc/WSe$_2$ heterostructure — •Sebastian Hedwig[1], Gregor Zinke[1], Benito Arnoldi[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Erwin-Schroedinger-Str. 46, 67663 Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Staudingerweg 7, 55128 Mainz, Germany

2D-Van-der-Waals systems are a highly intriguing class of low dimensional materials with promising spin functionalities for future nanoscale spintronic applications. Here, we show our approach to control the spin and charge carrier dynamics of the Van-der-Waals material WSe$_2$ by the adsorption of CuPc molecules. We conduct time-, angle- and spin-resolved photoemission experiments to investigate the optically excited carrier dynamics at the K- and Σ-points of WSe$_2$. After an initial spin-selective excitation at the K-point, depending on the pump light polarization [1], we observe that the subsequent intraband scattering from the K- to the Σ-point of the bare WSe$_2$ conduction band coincides with a change of the carriers spin polarization. Both, the optical excitation and the subsequent relaxation process can be actively modified by appropriate adsorption of CuPc. In particular, the dominant optical excitation at the K-point of WSe$_2$ is replaced by a direct interlayer excitation from the CuPc into the WSe$_2$ layer. We will show that the strength of the interlayer excitation can be tuned and controlled by the polarization of the exciting light field.

[1] Bertoni et al.; Phys. Rev. Lett. 117, 277201 (2016)

O 9.13 Mon 18:00 H3

Subcycle lightwave-ARPES in the strong-field regime — Suguru Ito[1], Manuel Meierhofer[2], Josef Freudenstein[2], Dmytro Afanasiev[2], •Jens Güdde[1], Rupert Huber[2], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Fakultät für Physik, Universität Regensburg, Germany

Angle-resolved photoemission spectroscopy (ARPES) combined with THz excitation enables the investigation of lightwave-driven Dirac currents in the surface state of topological insulators with sub-cycle time resolution [1]. At low THz frequencies and moderate effective field strengths of a few kV/cm at the surface, we have shown that the current dynamics is dominated by intraband acceleration of the electrons within the surface band of Bi$_2$Te$_3$.

Here, we will show how such experiments can be extended to the strong-field regime at driving frequencies of 25-40 THz. This is challenged by strong energy and momentum streaking of the photoelectrons after photoemission as well as the requirement of ultrashort pulses for photoemission which also introduce considerable energy broadening of the ARPES spectra. We will discuss how these challenges can be successfully overcome in order to enable lightwave-ARPES at field strength of $\sim$1 MV/cm despite surface screening. This paves the way for visualizing strong-field phenomena such as high-harmonic generation (HHG) [2] and the emergence of Floquet-Bloch states directly in the band structure on a sub-optical-cycle time scale.

[1] J. Reimann *et al.*, Nature 562, 396 (2018).

[2] C. P. Schmid *et al.*, Nature 593, 385 (2021).

O 10: Focus Session: Single Atom Catalysis 2

Time: Monday 15:00–17:30 Location: H4

Topical Talk O 10.1 Mon 15:00 H4

Atomically-precise design of low-nuclearity catalysts — •Sharon Mitchell and Javier Pérez-Ramírez — ETH Zurich, Zurich, Switzerland

Nanostructured catalysts incorporating supported metal atoms or small clusters of defined size and chemical composition attract considerable attention because of their potential to maximize resource efficiency. When optimally assembled, all the metal nuclei can participate in the catalytic cycle with properties tailored to deliver high specific activity and stable performance. Over the past decade, the number and diversity of reported systems have exploded as researchers attempted to control the nanostructure with increasing atomic precision. Nonetheless, spatially resolving the architecture and properties of supported low-nuclearity catalysts using existing analytical methods remains challenging. This talk will discuss approaches to prepare and characterize catalytic materials integrating low-nuclearity metal species. Topical case studies will introduce recent achievements and challenges, including the synthesis of single-atom catalyst libraries with controlled density, the precision synthesis of low-nuclearity species, tools for metal speciation analysis in electron-beam-sensitive materials, and an automated image analysis approach for atom detection and classification.

O 10.2 Mon 15:30 H4

Synthesis of single-atom model catalysts via atomic layer deposition for CO oxidation — •Chunlei Wang[1], Hélois Tissot[1], Joakim Halldin Stenlid[2], Markus Soldemo[1], Sarp Kaya[3], and Jonas Weissenrieder[1] — [1]Materials and Nano Physics, KTH Royal Institute of Technology, SE-100 44 Stockholm, Sweden — [2]Department of Physics, Stockholm University, SE-106 91 Stockholm, Sweden — [3]Department of Chemistry, Koc University, 34450 Istanbul, Turkey

Single-atom model catalysts, with individual metal atoms anchored on well-defined single crystals under ultra-high vacuum conditions, can provide an atomic-scale insight into active sites and reaction mechanisms for applied catalysis, thus promoting the design of better industrial catalysts. The metal growth of model catalyst is usually synthesized by physical vapor deposition method. Here, we applied a novel atomic layer deposition strategy to model systems for the synthesis of single-atom Fe1Ox catalysts on Cu2O(100) and Pt(111). The coordination configuration was determined through a combination of scanning tunneling microscopy, synchrotron radiation X-ray photoelectron spectroscopy (XPS), and density functional theory calculations. The redox properties of single atoms were investigated using ambient-pressure XPS under mbar level of reactant gas.

O 10.3 Mon 15:45 H4

A Customized IRAS System for Investigations of Adsorbates on Metal-Oxide Single Crystals — •David Rath, Jiri Pavelec, Ulrike Diebold, Michael Schmid, and Gareth S. Parkinson — Institute of Applied Physics, TU Wien, Austria

The IRAS system GRISU (**GR**azing incident **I**nfrared absorption **S**pectroscopy **U**nit) was developed to investigate adsorbates on metal oxide single crystals in the research field of single-atom catalysis [1]. It combines the commercially available FTIR spectrometer Bruker Vertex 80v with an UHV chamber [2]. The compact design requires only one CF150 port for the main optical components, features five mirrors for beam guidance placed in HV and UHV environment and optimises the system's performance, flexibility, and usability. The result is a small controllable focal-spot diameter (max. 3 mm) on the sample, motorised optical components, and an aperture limiting the incidence angle range (variable, 49° to 85°) on the sample. The simulated system (done with a ray-tracing program and a simplified spectrometer model) shows an efficiency of 13 %, i.e., 13 % of the radiation passing through the first aperture (Ø 6 mm) after the IR source in the FTIR spectrometer reaches the detector after being reflected from the molecular beam spot (Ø 3.5 mm) on the sample. Compared to a commercially available system with two parabolic mirrors with a focal length of 250 mm, the efficiency is about 20× higher. First measurements demonstrate the performance of the system.

[1] G. S. Parkinson, Catal. Lett. 149, 1137 (2019)

[2] J. Pavelec, et al., J. Chem. Phys. 146, 014701 (2017).

Topical Talk O 10.4 Mon 16:00 H4

Design of Model Single-Atom Catalysts: Metal Adatoms, Monomeric Oxide Units, and Mixed Surface Layers on Oxide Surfaces — •Zdenek Dohnalek — Physical and Computational Sciences Directorate and Institute for Interfacial Catalysis, Pacific Northwest National Laboratory, Richland, WA 99354, USA

Single-atom catalysts have attracted significant attention due to their ultimate metal efficiency and the promise of novel properties. The sublimation of oxides and metals is employed to design monodispersed model systems with supported metal adatoms, monomeric oxide units, and ordered mixed oxide surfaces. Scanning tunneling microscopy, ensemble-averaged electron spectroscopies, and density functional theory are employed to achieve an atomic-level understanding. Specifically, the deposition of (MgO)1 monomers and (MoO3)n oligomers is studied on anatase TiO2(101) via direct evaporation of MgO and MoO3 powders. While gas phase (MgO)1 is readily immobilized at room temperature, (MoO3)n transiently diffuse, agglomerate, and spontaneously decompose into the (MoO3)1 monomers. The transient mobility of the oligomers is the key to the self-assembly of the ordered overlayers of (MoO3)1. Metal adatoms and mixed oxide surfaces are synthesized by the sublimation of Rh onto Fe3O4(001). Higher substrate temperatures facilitate Rh incorporation into the surface, leading to ordered mixed Rh-Fe3O4(001). Cryogenic deposition temperatures stabilize Rh on the surface and allow for the preparation of the pure Rh adatom phase. The effect of temperature and adsorbates on the stability of such model catalysts is explored.

O 10.5 Mon 16:30 H4

Rh and Ir single atoms on Fe_3O_4(001): local structure affecting catalytic properties — •Matthias Meier[1,2], Zdenek Jakub[1], Jiri Pavelec[1], Michael Schmid[1], Ulrike Diebold[1], Cesare Franchini[2,3], and Gareth S. Parkinson[1] — [1]Institute of Applied Physics, Technische Universität Wien, Vienna, Austria — [2]Faculty of Physics and Center for Computational Materials Science, University of Vienna, Vienna, Austria — [3]Department of Physics and Astronomy, Alma Mater Studiorum, Università di Bologna, Bologna, Italy

Single-atom catalysts are often supported by cheap oxides, such as iron oxides. As a model system [1], magnetite (Fe_3O_4), specifically its (001) facet has been used because it offers stable sites for single-atom adsorption up to high temperatures. Here, I will demonstrate how important the support is for the stability of single-atoms, as well as their catalytic properties. Rh and Ir [2] single atoms utilize Fe vacancies in the subsurface of the reconstructed Fe_3O_4(001) unit cell to incorporate into the surface layer. Changing the positions of Fe atoms in the support as part of the incorporation process enables the single atoms to be accommodated in a more favorable configuration than if they were adsorbing on top of the surface. Their catalytic properties are drastically affected by changes in the atomic environment. Incorporation temperatures vary depending on both coverage and the presence or absence of adsorbates, such as CO.

[1] R. Bliem et al., Science **346**, 1215 (2014). [2] Z. Jakub et al., Angew. Chemie Int. Ed. **58**, 13961 (2019).

O 10.6 Mon 16:45 H4

Atomic-Level Studies of C_2H_4 on Clean and Rh_1 Single-Atom Decorated Fe_3O_4(001) — •Panukorn Sombut[1], Lena Puntscher[1], Chunlei Wang[1], Manuel Ulreich[1], Jiri Pavelec[1], Ali Rafsanjani-Abassi[1], Matthias Meier[2], Ulrike Diebold[1], Cesare Franchini[2,3], Michael Schmid[1], and Gareth S. Parkinson[1] — [1]Institute of Applied Physics, TU Wien, Austria — [2]Faculty of Physics, Center for Computational Materials Science, University of Vienna, Austria — [3]Alma Mater Studiorum, Università di Bologna, Bologna, Italy

The local binding environment of metal-oxide supported single-atom catalysts (SACs) determines how reactants adsorb and therefore plays a decisive role in catalysis. Here, we study how Fe_3O_4(001)-supported Rh_1 adatoms interact with ethylene (C_2H_4) using DFT, combined with experimental surface science techniques (TPD, XPS, and STM). We show that C_2H_4 physisorbs on the clean Fe_3O_4(001). We also identify and model different molecule orderings at different coverages that agree nicely with STM images and TPD data. Then, we study C_2H_4 adsorption at 2- and 5-fold coordinated Rh sites at the Fe_3O_4(001) surface, and show that the local environment has a strong effect on the adsorption properties: 2-fold Rh can adsorb two C_2H_4 molecules, while 5-fold Rh can only host a single C_2H_4 molecule. Finally, we investigate coadsorption of C_2H_4 with CO, a vital step towards enabling the hydroformylation reaction, and show that this is feasible only at 2-fold coordinated Rh sites.

Topical Talk O 10.7 Mon 17:00 H4

Model catalysis of single atoms on ultrathin solid films — •Kai Wu — College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, China

Metal atoms at surfaces play a key role in catalysis and related disciplines. A new strategy is introduced to prepare ultrathin films like oxides on surfaces which are further utilized to prepare stablized metal atoms. Since the surface free energy of a bulk oxide is much lower than the chemical potential or free energy of the metal atoms, one can play the game by reducing the thickness of the oxide film, even down to one atomic monolayer, which is grown on a bulk metal substrate. In such a way, the chemical potential of the oxide thin film can be tweaked by the underlying bulk metal substrate. Once the chemical potential or free energy of the ultrthin oxide support is tuned to such an extent that it becomes comparable with those of the metal atoms, can one then stablize these metal atoms without additional measures. Such an approach may be termed as surface free energy strategy to prepare uncoordinated metal atoms at surfaces. Scince the metal atoms are truly unprotected and therefore their physicochemical properties could be intrinsic, in sharp contract to those of coordinated metal atoms prepared by surface coordination and crystal engineering strategies. In this presentation, several systems of the unprotected metal atoms including alkali and transition metal atoms on ultrathin films such as metallic oxides and carbides grown on bulk metal substrates are employed to explore surface catalysis.

O 11: Electronic Structure Theory

Time: Monday 15:00–16:30 Location: H6

O 11.1 Mon 15:00 H6

Quantum Nuclear Effects in Thermal Transport of Semiconductors and Insulators — •Hagen-Henrik Kowalski[1], Matthias Scheffler[1], Mariana Rossi[2], and Christian Carbogno[1] — [1]The NOMAD Laboratory at the FHI-MPG and HU, Berlin, Germany — [2]MPI for Structure and Dynamics of Matter, Hamburg, Germany

Accounting for the nuclear motion is essential for the prediction of various material properties, from thermal conductivity to the relative stability of different polymorphs. Often, it is assumed that quantum nuclear effects (QNEs) are decisive at low temperatures, but that anharmonic effects can be neglected in this limit. Conversely, it is often presumed that anharmonicity is influential at elevated temperatures, but that QNEs are not active in this limit. In this work, we investigate the interplay of QNEs and anharmonicity by extending a recently proposed anharmonicity metric [1] to path integral molecular dynamics (PIMD). Our *ab initio* MD and PIMD calculations for solid Argon, Silicon, Lithium Hydrid, and Pentacene further substantiate that QNEs can have a massive impact even at room temperature and beyond, especially in weakly bonded systems [2]. Furthermore, we show that QNEs can induce strong anharmonic effects –beyond the applicability realm of perturbation theory– even at 0K. We discuss the underlying microscopic mechanisms and hence elucidate why QNEs and strong anharmonicity often go hand in hand in real materials.
[1] F. Knoop, et.al., *Phys. Rev. Mat.* **4**, 083809, (2020).
[2] M.Rossi, *J. Chem. Phys.* **154**, 170902 (2021)

O 11.2 Mon 15:15 H6

Volume Dependence of Excitation Energies of Sodium Clusters in GW — •Štěpán Marek and Richard Korytár — Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic

Prediction of molecular junction transport properties is a challenging task. One particularly complicated aspect of the problem is accurate description of binding between the molecule and the electrodes. Effect of image charges and non-ground state properties of the molecule-electrode system are expected to induce significant error when using DFT to evaluate the junction properties. GW is a post-DFT method that is assumed to fix some of the problems of bare DFT approach. In this contribution, we explore the (size) convergence properties of spectrum of sodium clusters using GW, and compare it to predictions by DFT and HF. We discuss the strategies to remove quasi-degeneracies induced by symmetries of the clusters, and their impact on critical properties of the spectrum, namely gap and average level spacing. Our analysis serves as a guide towards convergence studies of molecular junctions using GW.

O 11.3 Mon 15:30 H6

***Ab initio* phonon self-energies: To screen, or not to screen** — •Jan Berges[1], Nina Girotto[2], Tim Wehling[3], Nicola Marzari[4,1], and Samuel Poncé[5] — [1]University of Bremen, Germany — [2]Institute of Physics, Zagreb, Croatia — [3]University of Hamburg, Germany — [4]EPFL, Switzerland — [5]UCLouvain, Belgium

First-principles calculations of phonons are often based on the adiabatic approximation and a Brillouin-zone sampling that is not sufficient to capture Kohn anomalies. These shortcomings can be remedied through corrections to the phonon self-energy arising from the low-energy electrons. A well-founded correction method exists [Calandra, Profeta, and Mauri, Phys. Rev. B **82**, 165111 (2010)], which only relies on readily available (adiabatically) *screened* quantities. However, many-body theory suggests to use one *bare* electron–phonon vertex in the phonon self-energy [Giustino, Rev. Mod. Phys. **89**, 015003 (2017)] to avoid double counting. This can be seen as a limiting case of downfolding to *partially screened* phonons and interactions [Nomura and Arita, Phys. Rev. B **92**, 245108 (2015)]. We compare these approaches using the examples of TaS_2, MgB_2, n-doped MoS_2, and p-doped diamond. We confirm the robustness of the former method, while the latter allows for systematic improvements to describe correlations or metal–insulator transitions.

O 11.4 Mon 15:45 H6

Implementation of DFT+U+J and the minimum-tracking linear response method for polaron formation modeling — •Ziwei Chai[1,3], Karsten Reuter[3], Harald Oberhofer[1,2], and Limin Liu[4] — [1]Chair for Theoretical Chemistry, Technische Universität München — [2]Chair for Theoretical Physics VII, Universität Bayreuth, Germany — [3]Fritz-Haber-Institut der Max-Planck-Gesellschaft — [4]School of Physics, Beihang University

In many oxides charge carriers localize as small polarons. However, treating them with semi-local first-principles density-functional theory (DFT) tends to be a challenge usually addressed by hybrid DFT or Hubbard-corrected DFT+U. We present our implementation of DFT+U+J based on a "tensorial" representation of the subspace and the "minimum-tracking linear response method" which can determine U and J parameters from first-principles in the CP2K package. We performed systematic tests to prove the validity of the implementation.

Finally, the formation of polarons can be modeled by either breaking the symmetry of the initial structure or imposing an implicit or explicit constraining potential on the local orbital occupation. On top of our DFT+U+J implementation, we thus present the subspace occupancy-constraining potential (SOCP) approach to simulate the formation of polarons by constraining the occupancy number of the relevant local orbitals. Any polaronic configuration can thus straightforwardly be accessed without the need to explicitly break the system's symmetry.

O 11.5 Mon 16:00 H6

Signatures of molecular conformation in the evolution of DFT-based single molecule conductance — •Hector Vazquez — Inst. of Physics, Czech Academy of Sciences

Single molecules placed between two nanoelectrodes represent the ultimate limit in downscaling of electronic components. Electron transport simulations based on DFT-NEGF have enabled the understanding of conducting junctions and the interpretation of experiments [1]. However, these computationally costly calculations are often carried out for a small number of representative junction structures. In contrast, in room temperature experiments, the geometry of the molecule is expected to change significantly.

Here we describe and apply an approximate method to calculate molecular conductance within DFT for thousands of geometries. By combining it with room-temperature molecular dynamics (MD) simulations of the junction [2], we obtain the evolution of conductance for thousands of structure-conductance points. We analyze several geometric parameters and their effect on conductance, including quantum interference. This analysis on large datasets of DFT-based calculations reveals the signatures of molecular structure on junction conductance.

[1] F. Evers, R. Korytar, S. Tewari and J.M. van Ruitenbeek, Rev. Mod. Phys. 92, 35001 (2020)

[2] H. Vazquez, R. Skouta, S. Schneebeli, M. Kamenetska, R.Breslow, L. Venkataraman and M.S. Hybertsen, Nature Nanotechnol. 7, 663 (2012)

O 11.6 Mon 16:15 H6

Spectral properties and thermodynamics of correlated metals via the algorithmic inversion of dynamical potentials — •Tommaso Chiarotti[1], Andrea Ferretti[2], and Nicola Marzari[1] — [1]Theory and Simulations of Materials (THEOS) and National Centre for Computational Design and Discovery of Novel Materials (MARVEL), École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland — [2]Centro S3, CNR–Istituto Nanoscienze, 41125 Modena, Italy

Dynamical potentials are needed to predict accurate spectral, transport, and in general embedding properties of materials. The nonlinearity introduced by the frequency changes at a fundamental level the problem to address, moving from the diagonalization of an operator, e.g., the Kohn and Sham Hamiltonian in density-functional theory, to the Dyson inversion of a self-energy. Here, we propose a novel treatment of frequency-dependence able to solve Dyson-like equations via an exact mapping to an effective non-interacting problem, extending to the non-homogeneous case the algorithmic inversion method (Chiarotti et al., PRR, 2022). A sum-over-poles representation for the self-energy, together with the static one-particle Hamiltonian, are used to build a (larger) effective Hamiltonian having the excitation energies of the system as eigenvalues and the Dyson orbitals as projections of the eigenvectors. As a case study, we consider the paradigmatic system of S_rVO_3 to compute accurate spectra and energetics of the material.

O 12: Organic Molecules at Surfaces 2: Characterization of Organic Monolayers

Time: Monday 15:00–18:00 Location: S051

O 12.1 Mon 15:00 S051

Quantifying Interactions in Organic Monolayers — •Pierre-Martin Dombrowski, Stefan Renato Kachel, Leonard Neuhaus, Tobias Breuer, J. Michael Gottfried, and Gregor Witte — Philipps-Universität Marburg, Germany

The formation of molecular nanostructures is determined by the interplay of intermolecular and molecule-substrate interactions, whose experimental determination is challenging. Temperature-programmed desorption (TPD) is a well-established technique capable of quantifying these interactions, but its analysis is by no means trivial and therefore rarely done quantitatively for large adsorbates. In the present study, we analyse the desorption kinetics of the two organic semiconductors pentacene and perfluoropentacene from Au(111) and MoS_2 surfaces to show the potential of TPD and highlight challenges for larger adsorbates. [1,2] Combining TPD with scanning tunnelling microscopy, work function measurements and theoretical modelling, we show that intermolecular interactions are dominated by the intramolecular charge distribution. We are further able to determine the coverage-dependent prefactor of desorption with unprecedented precision, which enables a correlation of the desorption signal with the activation of specific degrees of freedom of motion of adsorbed molecules. Lastly, we compare the differences in molecule-substrate interactions for the two substrates at hand, which reveals that entropy can stabilize organic monolayer films on MoS_2 despite a weak molecule-substrate bond.

[1] S. R. Kachel et al., *Chem. Sci.* (2021), **12**, 2575-2585.

[2] P.-M. Dombrowski et al., *Nanoscale* (2021), **13**, 13816-13826.

O 12.2 Mon 15:15 S051

Toward Understanding Thermal and Electric Properties of Single Molecular Junctions and Self-Assembled Monolayers — •Mohamed Ibrahim[1], Philipp Wiesener[1], Luke O'Driscoll[2], Martin Bryce[2], and Achim Kittel[1] — [1]Oldenburg University, Oldenburg, Germany — [2]Durham University, Durham, England

Over the last years, implanting organic molecules in devices is continuously attracting large attention because of their small size (nm scale), tunable electronic and thermal properties by manipulating individual atoms. Therefore, it is obvious to extend the field of thermoelectrics using molecules to cool devices and sensors very locally. A single layer of well organized molecules is formed and realized by the self assembly mechanism, which allows molecular moieties to be adsorbed spontaneously on a surface producing large domains. This motivated us to report here about the characterisation of selfassembled oligo phenylene ethynylene dithiol molecules (OPE3), and some specifically modified forms by mainly adding side groups to it on gold surfaces by means of X-ray photoelectron spectroscopy (XPS), reflected electron energy loss (REELS), and ultraviolet photoelectron spectroscopy (UPS). The results show that the unsubstituted OPE3 has a high densely packed SAM with a thickness 1.7 nm, while the presence of substituents, attached to the middle ring, led to variation of the SAM film thickness. This indicates changes in the geometric configuration of the π stacking of OPE3 especially, tilt angle and packing densities. Parent OPE3 REELS spectrum shows a band gap value of 2.01 eV which is totally different than the reported value in the literature.

O 12.3 Mon 15:30 S051

Molecular orientation and phase transitions of DHTAP — •Claudia López-Posadas[1], Michael Györök[1], Antony Thomas[2], Thomas Leoni[2], Olivier Siri[2], Conrad Becker[2], and Peter Zeppenfeld[1] — [1]Institute of Experimental Physics, Johannes Kepler University Linz, Altenberger Str. 69, 4040 Linz, Austria — [2]Aix-Marseille University, CNRS, CINaM, UMR 7325, F-13288 Marseille, France

The structure and orientation of 5,14-dihydro-5,7,12,14-tetraazapentacene (DHTAP) layers deposited on Cu(110), Cu(110)-(2x1)O and the Cu(110)/Cu(110)-(2x1)O stripe phase was studied using reflectance difference spectroscopy (RDS), Scanning Tunneling Microscopy (STM) and Low Energy Electron Diffraction (LEED). The evolution of the RDS signal allows to identify the sequential formation of up to three monolayers as well as a phase transition upon completion of the first one. On Cu(110), DHTAP molecules in the first monolayer are always lying flat with their long molecular axis aligned parallel to the [-110]-direction of the Cu(110) surface. However, for subsequent layers the orientation critically depends on the deposition temperature T. At T=240K the DHTAP molecules are mostly aligned parallel to the ones in the first layer, whereas at room temperature and above their preferential orientation is orthogonal to the molecules in the first layer, the coexistence of the two orientations can be observed at a critical temperature of T=270K. Finally, the main optical transitions and the orientation of the transition dipole moments of the DHTAP layers were extracted from the RDS spectra.

O 12.4 Mon 15:45 S051

Nickel(II) Porphyrins on Metal Surfaces: Oxidation-State Tuning and Formation of a Supramolecular Mixed-Valence Adsorbate Structure — •Jan Herritsch, Qitang Fan, Marie-Irène Albus, Lukas Ruppenthal, Lukas J. Heuplick, Leonard Neuhaus, Tobias Wassermann, and J. Michael Gottfried — Philipps-Universität Marburg

Nickel tetrapyrrole complexes are structurally related to biologically relevant molecules (e.g., F430 cofactor, tunichlorin, nibalamine) and are promising precursors for novel functional interfaces. Here, we report on the influence of the interaction at the metal/organic interface on the electronic structure of a nickel octaethyl porphyrin (Ni(OEP)) monolayer on different coinage metal surfaces studied by XPS, UPS, STM, NEXAFS and LEED. On Ag(111), Ni(OEP) forms a mixed-valent adsorbate phase in which the Ni centers occur in two different oxidation states. The two separate peaks in the Ni 2p XP spectrum indicate that about 40% of the Ni centers are reduced. STM and LEED show an incommensurate superstructure with an ordered arrangement of the metal centers in different oxidation states. Further insights into the valence electronic structure were obtained by UPS and NEXAFS. On the more reactive Cu(111) surface, Ni(OEP) forms a long-range ordered structure in which nickel centers are uniformly reduced; whereas on the most inert Au(111) surface, the oxidation state of the Ni centers remains unaffected by adsorbate interactions.

O 12.5 Mon 16:00 S051

Electronic and structural properties at the NiTPP/O-Cu(100) interface — •Jonah Elias Nitschke[1], Henning Sturmeit[1], Iulia Cojocariu[2], Vitaliy Feyer[2], Alessandro Sala[2], Andreas Windischbacher[3], Peter Puschnig[3], Stefano Ponzoni[1], Giovanni Zamborlini[1], and Mirko Cinchetti[1] — [1]Department of Physics, TU Dortmund University, Germany — [2]Peter Grünberg Institut (PGI-6), Forschungszentrum Jülich, Germany — [3]Institute of Physics, University of Graz, Austria

Depending on the strength of the molecule-substrate interaction, charge transfer, chemical reactions or a redistribution of the electronic cloud may occur at organic-metal interfaces. Here, we investigate the structural and electronic properties of Nickel tetraphenylporphyrin molecules (NiTPP) deposited on oxygen-passivated Cu (100) surface. By using a multi-technique approach, which combines LEED and STM, we reveal a coverage dependent superstructure with multiple domains that ultimately reduces into a single unit cell.

In the latter configuration, STM measurements show that the NiTPP molecules adsorb either with the macrocycle planar to the surface on in a saddle-shape configuration, with the pairs of opposite pyrrole rings tilted upwards. STS and photoemission orbital tomo-graphy measurements show that, contrary to the NiTPP adsorbed on the bare Cu(100) surface, where a significant charge transfer is observed, the oxygen overlayer quenches the charge transfer at the interface, thus offering a simple approach to physically decouple of the molecular film from the underlying substrate.

O 12.6 Mon 16:15 S051

Surface Chemical Bond and Molecular Topology of Polycyclic Aromatic Systems — •Lukas Ruppenthal[1], Florian Münster[1], Benedikt P. Klein[1], Jan Herritsch[1], Leonard Neuhaus[1], Stefan R. Kachel[1], Pengcai Liu[2], Xing-Yu Chen[2], Jiawen Cao[2], Lars E. Sattler[3], Sebastian M. Weber[3], Qitang Fan[1], Gerhard Hilt[3], Xiao-Ye Wang[2], and J. Michael Gottfried[1] — [1]Department of Chemistry, Philipps-University Marburg, Germany — [2]College of Chemistry, Nankai University Tianjin, China — [3]Institute of Chemistry, Carl von Ossietzky University Oldenburg, Germany

Metal/organic interfaces have a large impact on the performance of organic (opto-)electronic devices. Therefore, the detailed understanding of their chemical, electronic and geometric structure is important for the further technological development. Many common organic semiconductors contain π-electron systems with alternant topologies, whereas non-alternant alternatives have only recently found increasing attention due to their unusual electronic properties. Here, we compare the alternant polycyclic aromatic molecule pyrene with its non-alternant isomers acepleiadylene and azupyrene regarding their interaction with Cu(111) surface, using PES, NEXAFS, TPD and STM. The non-alternant isomers are also interesting as molecular models of graphene defects, e.g. azupyrene for the Stone-Wales defects. In all cases, the non-alternant isomers show increased metal/molecule-interaction due to their reduced HOMO-LUMO gap, which brings the LUMO energetically closer to the Fermi energy of the metal, causing stronger hybridization with electronic states of the metal surfaces.

O 12.7 Mon 16:30 S051

Tailoring the organic-semiconductor/metallic interface: From self-assembly to heteromolecular phases of carboxylic acids on Ag surfaces — •Matthias Blatnik, Veronika Stará, Anton Makoveev, Jakub Planer, Thomáš Krajňak, Pavel Procházka, and Jan Čechal — CEITEC BUT, Brno, CZ

Atomic-level understanding of the metal/organic-semiconductor interface has become paramount in the strife for the development and fabrication of more efficient organic electronic devices in recent years. Interfacial properties are crucially linked to molecule-molecule and molecule-substrate interactions, molecular functionalization (e.g., deprotonation), a precise energy level alignment, substrate termination or the formation of multi- or heterolayers with a different organic compound. All these have to be well understood through the study of model systems in ultra-clean conditions before technological advances can be achieved. Here, we present a model system of self-assembled aromatic carboxylic acids (e.g., 4,4' Biphenyl Dicarboxylic Acid, 1,3,5-Benzenetribenzoic Acid) on Ag surfaces (orientations (100) and (111)). We explore the effects of the metal substrate's orientation on formation and growth of single molecular phases, introduce a monolayer thick layer as a charge injection layer and study the formation of a heteromolecular compound with an additional organic material (e.g., pentacene). We employ low energy electron microscopy (LEEM) and diffraction (μLEED), as well as STM and XPS to give a real-time view and detailed information on nucleation and growth of the molecular phases and transformations and the chemical composition.

O 12.8 Mon 16:45 S051

Highly ordered commensurate structures of merocyanines on Ag(100) — •Anna Juliana Kny[1], Max Reimer[2], Noah Al-Shamery[1], Ritu Tomar[1], Thomas Bredow[1], Selina Olthof[2], and Moritz Sokolowski[1] — [1]Institut für Physikalische und Theoretische Chemie der Universität Bonn, Germany — [2]Department für Chemie der Universität zu Köln, Germany

Vacuum deposited films of the merocyanine molecule 2-[5-(5-dibutylamino-thiophen-2-yl-methylene)-4-*tert*-butyl-5*H*-thiazol-2-ylidene]-malononitrile (HB238) and its derivatives have been investigated in the context of organic solar cells [1].

Although the specific structural order in these films is important for the optical properties, only very little is known about the adsorption and ordering of merocyanines on surfaces. Therefore, we investigated monolayers of HB238 on a Ag(100) surface by SPA-LEED, STM, XPS, UPS, and DFT calculations.

Upon deposition onto the Ag(100) surface at room temperature the formation of a commensurate superstructure is observed. It is composed of homochiral HB238 aggregates of four molecules. We discuss the surface bonding and structure formation in dependence of the specific functional groups and, furthermore, the sterically demanding donor substituents. For the role of the latter one, we also investigated derivatives of HB238 on Ag(100). The merocyanines were synthesized and kindly provided by the group of Prof. K. Meerholz (Cologne).

Supported by the DFG through the research training group 2591. [1] JACS 137 (2015) 13524.

O 12.9 Mon 17:00 S051

Van der Waals Heteroepitaxy: Intrinsic Epitaxial Alignment of Perfluoropentacene Films on Transition Metal Dichalcogenides — •Maximilian Dreher, Darius Günder, and Gregor Witte — Philipps-University Marburg, Germany

In this work, we have studied the formation and azimuthal alignment of crystalline adlayers of the organic semiconductor (OSC) perfluoropentacene (PFP) on the basal plane of several transition metal dichalcogenides (TMDC) single crystals, which are further compared to graphite and hBN. The quite inert basal planes of TMDCs enable an unrestricted growth of OSCs without the requirement of relaxation in terms of commensurability at the interface, since molecules exhibit a stronger interaction among each other than with the substrate. Nevertheless, the crystalline PFP multilayers exhibit distinct azimuthal twist angles relative to the substrate surface, which we could rationalize by so called 'on-line coincidences' introduced by Forker et al. recently. [1] Here, the molecules do not favor a specific adsorption site at the interface as it is often described by commensurate superstructures. Instead the bulk crystal structure remains unperturbed down to the interface, but a specific twist angle is adopted, where molecules avoid unfavored adsorption sites. The extreme sensitivity of the resulting twist angles by small deviations in the crystal structure enabled us further to use the large thermal expansion of the OSC to control the twist angles by changing the substrate temperature during deposition. [1] Forker et al., Soft Matter 13, 1748-1758 (2017) [2] Dreher et al., Chem. Mater. 32, 20, 9034-9043 (2020)

O 12.10 Mon 17:15 S051

Steering Self-Assembly of Three-Dimensional Iptycenes on Au(111) by Tuning Molecule-Surface Interactions — Lukas Grossmann[1,2], •Eva Ringel[1,2], Wolfgang Heckl[1,2], and Markus Lackinger[1,2] — [1]Deutsches Museum, Museumsinsel 1, 80538 München — [2]Technische Universität München, Physics Department, James-Franck-Strasse 1, 85748 Garching

Three-dimensional organic molecules have been neglected in studies on surfaces, even though their self-assemblies exhibit a far greater variability than their planar counterparts. While planar molecules adsorb mostly flat on surfaces, three-dimensional molecules can adopt vastly different adsorption geometries. This additional degree of freedom can result in self-assembly of entirely different supramolecular structures. Moreover, adsorption geometries can be steered by tuning molecule-surface interactions, thereby providing a new means for 2D crystal engineering. In this respect, iptycenes are highly stable, but currently underexplored model compounds. Here, we study self-assembly of threefold symmetric triptycene derivates with extended anthracene blades on Au(111) surfaces. Additionally, the influence of intrinsic dipole moments was investigated by comparing analogs with peripheral fluorine substitution. All structures were resolved by Scanning-Tunneling-Microscopy under ultra-high vacuum conditions. On pristine Au(111), the molecules maximize the area of contact, whereas iodine-passivation affords hexagonal porous structures that feature optimized molecule-molecule interactions through face-to-face stacking of all anthracene blades.

O 12.11 Mon 17:30 S051

Adsorption of Submonolayer Coverages of Phenylphosphonic Acid on Rutile TiO2(110) — •Alexander Wolfram[1], Maximilian Muth[1], Julia Köbl[1], Nataliya Tsud[2], Sascha Mehl[3], Hans-Peter Steinrück[1], and Ole Lytken[1] — [1]Friedrich-Alexander Universität Erlangen-Nürnberg, Lehrstuhl für Physikalische Chemie 2, Egerlandstr. 3, 91058 Erlangen, GER — [2]Charles University, Faculty, of Mathematics and Physics, Department of Surface and Plasma Science, Holešovičkách 2, Prague, 18000, Czech Republic — [3]Elettra-Sincrotrone Trieste SCpA, Strada Statale 14, km 163.5, Trieste, Basovizza, 34149, Italy

The interfaces of oxide surfaces with organic molecules are crucial for the performance of devices, such as dye-sensitized solar cells, sensors and organic electronics. All these devices contain interfaces where organic molecules are bound to surfaces, often using covalent anchoring groups. These anchoring groups strongly influence the interface and thus the device performance. An interesting anchoring group is the phosphonic acid group, which is well known to bind strongly to oxide surfaces. At ELETTRA sinchrotrone in Trieste we investigated submonolayer coverages of phenylphosphonic acid on a rutile TiO_2 (110) surface with high-resolution X-ray photoelectron spectroscopy (XPS) and near-edge X-ray absorption fine structure (NEXAFS) spectroscopy. Based on the changes in the O 1s and P 2p core levels, we are able to identify the different binding motifs present on the surface at different temperatures, while the NEXAFS intensities gives us information about the orientation of the phenyl moiety.

O 12.12 Mon 17:45 S051

Accurate determination of adsorption-energy differences of metalloporphyrins on TiO2(110) 1x1 — •Maximilian Muth, Alexander Wolfram, Elmar Kataev, Julia Köbl, Hans-Peter Steinrück, and Ole Lytken — Univ. Erlangen-Nürnberg

A deeper knowledge of the behavior between porphyrin molecules and the rutile TiO2(110) surface and is of crucial importance for the development of new applications. Especially the usage of such systems in photocatalysis or solar cells are promising possibilities. Therefore, we investigated the adsorption energy of three different metallotetraphenylporphyrins (MTPP M = Mg, Co, Zn). Unfortunately, temperature programmed desorption, a typical method for the determination of adsorption energies is not applicable for this particular system because of the irreversible adsorption of the monolayer in direct contact to the TiO2-surface. For this reason, instead, we compared the adsorption-energy of the three MTPPs relatively to each other by using a layer exchange experiment. We adsorb mixtures of always two different MTPPs on top of each other and allow the molecules to diffuse during a heating ramp. Eventually, an equilibrium state forms in which the MTPP with the higher adsorption-energy will enrich in the monolayer in direct contact with surface while the weaker adsorbing MTPP will be enriched in the multilayer. After further heating for multilayer desorption we determine the concentrations of the MTPPs found in the remaining monolayer by using XPS and use them to calculate the difference in adsorption-energy of MTPPs via the equilibrium constant.

O 13: Nanostructures at Surfaces 2

Time: Monday 15:00–17:30 Location: S052

O 13.1 Mon 15:00 S052

A new setup for dimensional nanometrology using soft X-rays — •Leonhard Lohr[1], Richard Ciesielski[1], Analía Fernández Herrero[2], Andreas Fischer[1], Alexander Grothe[1], Frank Scholze[1], and Victor Soltwisch[1] — [1]Physikalisch-Technische Bundesanstalt (PTB), Abbestraße 2-12, 10587 Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie, Albert-Einstein-Straße 15, 12489 Berlin, Germany

Measuring nanostructured surfaces on small test patterns on dies from semiconductor fabs is an important metrology challenge. The geometrical dimensions of metrology test structures, such as linear gratings, must be determined with uncertainties in the sub-nm range. By using methods such as small angle X-ray scattering or X-ray fluorescence under grazing incidence, the photon beam spot size becomes too large for sufficient accurate measurements.

We present a new setup, mounted on the soft X-ray beamline in PTB's laboratory at the electron storage ring BESSY II. This setup works with synchrotron radiation in an ultra-high vacuum and with lubricant-free mechanics. Its small and compact design enables to detect scattered monochromatic soft X-rays under angles of incidence up to 30 degrees. The setup reduces photon beam spot size and covers diffraction patterns like them from scattered hard X-rays under grazing incidence. Depending on the incident photon energy, fluorescence from small excited regions of the sample can be measured simultaneously.

We present first measurement results from small test patterns which are located by imaging the surface using a large photon beam.

O 13.2 Mon 15:15 S052

Synthesis of metal sulfide nanoribbons on graphene by self-assembly — •Xuejiao Zhang, Kelvin Anggara, Vesna Srot, Xu Wu, Peter A. van Aken, and Klaus Kern — Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany

Bottom-up synthesis of nanostructures on surface have relied on the self-assembly of nanoscale building blocks. The diversity of accessible nanostructures however have been constrained by the limited choice of atomic and molecular building blocks that can be evaporated on surface. Here we bypass this limitation by using complex inorganic ions generated from electrospray ionization as building blocks to synthesize nanostructures on surfaces. We deposited HMonS3n+1** (n = 4-6) ions onto a freestanding single-layer graphene by Electrospray Ion-Beam Deposition (ESIBD), and imaged the resulting nanostructures by aberration-corrected Scanning Transmission Electron Microscopy (STEM). The molecules were observed to form anisotropic, single-layered, crystalline MoS2 nanoflakes (< 100 nm2), which in turn self-assembled into MoS2 nanoribbons extending as far as 1 μm. The first observation of such nanostructures evidences the potential of this approach to prepare previously inaccessible nanomaterials on surfaces.

O 13.3 Mon 15:30 S052

Ni kagome lattice on Pb(111) — •Gustav Bihlmayer[1], Yen-Hui Lin[2], Stefan Blügel[1], and Pin-Jui Hsu[2] — [1]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich, D-52425 Jülich, Germany — [2]Department of Physics, National Tsing Hua University, Hsinchu, 30013, Taiwan

Deposition of atomically thin films on well-ordered surfaces sometimes allows realizing new structural motives that have no correspondence in the bulk phases. Here we observe that, at low temperatures, submonolayer growth of Ni on a Pb(111) substrate leads to well-ordered, hexagonal islands. In scanning tunneling microscopy (STM) two types of edges with very different apparent height and scattering properties are observed. From density functional theory (DFT) calculations and STM images with atomic resolution we conclude that the Ni atoms form a kagome lattice and the islands show saw-tooth edges. Comparison of STM and DFT data suggests that some Ni atoms are incorporated in the subsurface layer and give rise to the low observed height of the islands and the strong difference observed at their edges. From the DFT calculations we conclude that the Ni island is non-magnetic and the electronic structure shows some characteristics of the kagome lattice, i.e. flat bands also visible in scanning tunneling spectra.

O 13.4 Mon 15:45 S052

DNA origami as reference systems for nano-metrology — •ziba akbarian[1,2], birka lalkens[1,2], michelle weinert[3], ingo busch[3], harald bosse[3], and uta schlickum[1,2] — [1]Technische Universität Braunschweig, Braunschweig, Germany — [2]Laboratory for Emerging Nanometrology (LENA), Braunschweig, Germany — [3]Physikalisch Technische Bundesanstalt (PTB), Braunschweig, Germany

A key aspect of quantitative measurements is the traceability of all measurements to the international system of units (SI). DNA origami is capable of being traced back to SI units thanks to its precisely defined internal structures in 2D and 3D. Here, we will describe the possibilities of building reference systems for calibrating scanning probe instruments with μm resolution only that can be traced back to atomically precise structures. In order to accomplish this goal, we plan to fabricate DNA origami structures that include marks at well-defined positions to obtain measurable protrusions within the internal structures. The controlled fabrication of DNA origami structures that do not distort upon adsorption onto surfaces, in both liquid and air conditions will be surveyed as the first step in this research.

O 13.5 Mon 16:00 S052

How covalent chemistry affects the surface dipole of metallic nanostructures — Rustem Bolat[1,2], •Jose M. Guevara[1], Marvin Knol[1,2], Philipp Leinen[1], Ruslan Temirov[1,2,3], Oliver T. Hofmann[4], Reinhard J. Maurer[5], F. Stefan Tautz[1,2], and Christian Wagner[1,2] — [1]Forschungszentrum Jülich, Germany — [2]RWTH Aachen University, Germany — [3]Universität zu Köln, Germany — [4]Graz University of Technology, Austria — [5]University of Warwick, UK

Scanning probe experiments often encounter nanostructures of undercoordinated metal atoms, either as the investigated system or at the tip apex. While the electrostatic properties of extended planar surfaces are textbook material, the interaction of metal adatoms with the surface and with each other opens an interesting playground in which our intuition for the covalent chemistry of such metal structures is limited.

Here we investigate Ag and Au adatoms, monoatomic chains and small clusters, on the Ag (111) surface using quantum dot microscopy. We image the electrostatic potential above these nanostructures and quantify the respective surface dipoles. The two species of adatoms behave antagonistic, as the surface dipoles are positive for Ag and negative for Au structures. The measured dipoles are in excellent agreement with density functional theory calculations. We disentangle the influence of individual adatom-surface and adatom-adatom bonds down to individual atomic orbitals.

O 13.6 Mon 16:15 S052

Gold nanostructures with varying physicochemical surrounding examined with surface second harmonic generation circular dichroism spectroscopy — •Natalie Fehn[1], Ehsan Vahidzadeh[2], Karthik Shankar[2], Ueli Heiz[1], and Aras Kartouzian[1] — [1]Physical Chemistry, Department of Chemistry and Catalysis Research Center, Technical University of Munich — [2]Electrical and Computer Engineering Deptartment, Faculty of Engineering, University of Alberta

The sophisticated second-order spectroscopic methods of surface second harmonic generation (s-SHG) and s-SHG circular dichroism spectroscopies (s-SHG-CD) provide additional and complementary structural information in regards to their linear counterparts, absorbance and CD spectroscopy. Because of their surface sensitivity, s-SHG and s-SHG-CD represent an excellent choice for the investigation of molecular thin films and supported plasmonic structures, such as nanoparticles and clusters. We are especially interested in the interaction of such nanostructures with their surroundings, e.g. chiral molecule adsorbates or oxide layers, with the goal of observing a chiral response from the originally achiral particles. Those particles may then serve as catalysts in asymmetric heterogeneous catalysis. This contribution will focus on our research with emphasis on supported gold nanostructures.

O 13.7 Mon 16:30 S052

Elucidating the chirality of polycrystalline films with fundamental and SHG-CD — •Kevin Liang[1], Florian Ristow[2], Jakob Scheffel[2], Natalie Fehn[1], Reinhard Kienberger[2], Ueli Heiz[1], Aras Kartouzian[1], and Hristo Iglev[2] — [1]Physical Chemistry, Department of Chemistry and Catalysis Research Center, Technical University of Munich — [2]Physics Department E11, Technical University of Munich

Preferential desorption of enantiomers of 1,1'-Bi-2-naphthol (BINOL) has been observed, depending on the handedness of circularly polarised light, which is of interest for enantioenrichment processes. Therefore, characterisation of the enantiomeric excess of the sample and the understanding of its chirality is key. However, that requires further studies and greater clarification to be carried out and obtained, respectively. This contribution discusses the optical properties of BINOL thin films with fundamental and second harmonic generation (SHG) circular dichroism spectroscopy (CD) with different sample preparation methods, enantiomeric compositions and under varying experimental conditions. Depending on the previous conditions mentioned, the observed CD and anisotropy (g) value can be even of opposite sign for the same thin film.

O 13.8 Mon 16:45 S052

AFM studies of ion-exchange treated glass surfaces — •Fabian Höhn[1,3], Fabian Ullmann[1,3], Norbert Arndt-Staufenbiel[2], and Stefan Krischok[1,3] — [1]Institute of Physics, TU Ilmenau, Weimarerstraße 25, 98693 Ilmenau, Germany — [2]Fraunhofer Institute for Reliability and Microintegration, Gustav-Meyer-Allee 25, 13355 Berlin, Germany — [3]Center of Micro-and Nanotechnologies, TU Ilmenau, Gustav-Kirchhof-Straße 7, 98693 Ilmenau, Germany

There is a big difference between the bond strength of glass and its respective tensile strength. The reason for this discrepancy is assumed to be so-called "microcracks", this surface damage is considered to massively reduces the tensile strength. We assume that these damages widen by stretching the surface and become thus more visible. Another point that will be investigated, regarding this matter, is micro waveguides in glass. These are created by silver ion exchange in glass. It is assumed that the mentioned surface damages are a cause for reduced quality of these waveguides. For this purpose, we examine the difference in topography between untreated Gorilla V1 glass from Corning, a sample stretched using the three-point method and a sample in which a silver ion exchange was carried out on the surface. The difference in topography is first described qualitatively and then determined quantitatively with roughness parameters. Finally, a comparison of the three samples should provide an insight into the topography of glasses under stress and a contribution to the optimization of the mentioned waveguides.

O 13.9 Mon 17:00 S052

Processing copper surfaces with ultrashort laser pulses to reduce secondary electron yield — •Elena Bez[1,2], Marcel Himmerlich[1], Ana Karen Reascos Portilla[1], Pierre Lorenz[3], Klaus Zimmer[3], Mauro Taborelli[1], and André Anders[2,3] — [1]CERN, European Organization for Nuclear Research, 1211 Geneva 23, Switzerland — [2]University of Leipzig, Linnéstr. 5, 04103 Leipzig, Germany — [3]Leibniz Institute of Surface Engineering (IOM), Permoserstr. 15, 04318 Leipzig, Germany

Ultrashort-pulse laser processing in air is employed to engrave micro- and nanostructures on copper surfaces aiming to reduce secondary electron emission. Parameters such as the laser power and scanning speed are varied to investigate their influence on the resulting structures. The morphology, as well as the chemical composition of the laser-treated surfaces, are analyzed by scanning electron microscopy (SEM) and X-ray photoelectron spectroscopy (XPS), respectively. At low power and high scanning speed, only slight changes to the surface topography occur, whereas compact, cauliflower-like nanostructures and micrometer deep trenches are generated at high power and low scanning speed. The higher the accumulated laser fluence, the more material is ablated and the more oxidized particles are redeposited. A clear correlation exists between the accumulated fluence for processing with 355, 532 and 1064 nm photons, and the resulting secondary electron yield of the surface. Its maximum can be reduced from 2.2 to 0.7. Mastering these dependencies helps to develop a system that enables laser processing of beam pipes of selected magnets in the Large Hadron Collider.

O 13.10 Mon 17:15 S052

Highly ordered three-dimensional Ni-TiO2 nanopore arrays as sodium-ion battery anodes — •Mo Sha, Huaping Zhao, and Yong Lei — Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, 98693 Ilmenau, Germany

Sodium-ion batteries (SIBs) represent an effective energy storage technology with potentially lower material costs than lithium-ion batteries. Here, we show the electrochemical performance of SIBs with electrode design at the nanoscale. Highly ordered three-dimensional (3D) self-supported Ni-TiO2 nanopore arrays (NiNPA@TiO2) with highly oriented nanoporous structures are fabricated using nanoimprited AAO templating technique and applied as nanostructured anodes for SIBs applications. Their large specific surface area can ensure a high capacity, and their highly oriented and stable nanoporous structure can facilitate ion transport. The NiNPA@TiO2 nanoarrays delivered a reversible capacity of 240 mAh g-1 after 100 cycles at the current density of 50 mAh g-1 and were able to retain a capacity of 105 mAh g-1 at the current density as high as 5 A g-1. Their large active sites, high ion accessibility, fast electron transport, and excellent electrode integrity were shown as great merits to obtain the presented electrochemical performance. Not limited to the SIBs electrodes, the highly ordered 3D heterostructured nanoarrays as a promising electrode design for other electrochemical energy conversion and storage devices.

O 14: New Methods and Developments 2: Scanning Probe Techniques 2 (joint session O/KFM)

Time: Monday 15:00–16:15 Location: S053

O 14.1 Mon 15:00 S053

The importance of the dipole at the metal tip apex when approaching closer with a CO tip — •Shinjae Nam, Oliver Gretz, Thomas Holzmann, Alfred John Weymouth, and Franz J. Giessible — University of Regensburg, Regensburg, Germany

By functionalizing the tip with a single CO molecule, the resolution of atomic force microscope (AFM) can be drastically increased. The contrast enhancement produced by a CO tip has been explained in terms of strong Pauli repulsion and the associated tilting of the probe molecule. Although these two interactions play a dominant role at very close distances, recent experiments show that other interactions, especially electrostatic forces, are also important to understand the observed contrast. Here, we used Lateral Force Microscope, a variant of frequency modulation atomic force microscopy, to quantify the interaction between a CO tip and a CO on the Cu (111) surface. Interestingly, one more feature appeared in the measurement when we measured closer to the surface at the side of the surface CO. Following the results of other investigations, we include the electrostatic force in our simulations. We modeled our tip as a quadrupole, including a dipole at both the metal tip and on the CO molecules. We found that the dipole of the metal apex of the tip becomes a much greater influence as we approach closer to the surface.

O 14.2 Mon 15:15 S053

Chemical bond imaging using torsional and flexural higher eigenmodes of qPlus sensors — •Daniel Martin-Jimenez[1], Michael G. Ruppert[2], Alexander Ihle[1], Sebastian Ahles[3], Hermann A. Wegner[3], André Schirmeisen[1], and Daniel Ebeling[1] — [1]Institute of Applied Physics, Justus Liebig University Giessen, Heinrich-Buff-Ring 16, 35392 Giessen (Germany). — [2]University of Newcastle, Callaghan, NSW, 2308 (Australia). — [3]Institute of Organic Chemistry, Justus Liebig University Giessen, Heinrich-Buff-Ring 17, 35392 Giessen (Germany).

Non-contact atomic force microscopy (AFM) with CO-functionalized tips allows to visualize the chemical structure of adsorbed molecules and identify individual inter- and intramolecular bonds. Herein, we analyze the suitability of qPlus sensors, which are commonly used for bond imaging, for the application of modern multifrequency AFM techniques. Two different qPlus sensors were tested for submolecular resolution imaging via actuating torsional and flexural higher eigenmodes and via bimodal AFM. The torsional eigenmode of the first sensor is perfectly suited for performing lateral force microscopy (LFM) with single bond resolution. The advantage of using a torsional eigenmode is that the same molecule can be imaged either with a vertically or laterally oscillating tip without replacing the sensor simply by actuating a different eigenmode. Submolecular resolution is also achieved by actuating the 2nd flexural eigenmode of our second sensor. With laser Doppler vibrometry measurements and AFM simulations we can rationalize the image contrast mechanism of the 2nd eigenmode.

O 14.3 Mon 15:30 S053

3D Force mapping of single organic molecules at room temperature — •Timothy Brown, Philip Blowey, Jack Henry, and Adam Sweetman — University of Leeds, Leeds, UK

Scanning probe microscopy has established itself as a highly effective technique in the study of surfaces and molecules. In particular, non-contact atomic force microscopy has yielded enormous progress in our ability to characterise materials at the atomic scale, including the ability to resolve the chemical structure of individual molecules, and to acquire 3D force-maps with intramolecular resolution.

Intramolecular imaging is almost exclusively performed using qPlus sensors at cryogenic temperatures, as the functionalisation of the tip via a CO molecule (required for intra-molecular imaging) is only stable at near liquid helium temperatures. Although it has been shown that intramolecular imaging may be performed at higher temperatures, via use of semi-conducting, rather than metallic substrates, acquisition of high density 3D data sets generally requires long acquisition times. Hence the lack of thermal equilibrium between the tip and sample at room temperature makes acquisition of these datasets at elevated temperatures extremely challenging.

In this talk we present the first demonstration of high resolution 3D force mapping of a single organic molecule at room temperature using conventional silicon cantilevers. We show how the challenges of operating in a room temperature experimental environment can be overcome to acquire reproducible 3D force maps of a resolution and quality previously only demonstrated at low temperature.

O 14.4 Mon 15:45 S053

Monitoring of molecular configurations during manipulation with a scanning probe microscope — •Joshua Scheidt[1,2], Alexander Diener[1,2], Michael Maiworm[3], Rolf Findeisen[3], Kurt Driessens[2], F. Stefan Tautz[1], and Christian Wagner[1] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Jülich, Germany — [2]Maastricht University, Data Science and Knowledge Engineering, Maastricht, Netherlands — [3]Control and Cyber-Physical Systems Laboratory, Technische Universität Darmstadt, Darmstadt, Germany

A bold vision of nanofabrication is the assembly of functional molecular structures with a scanning probe microscope (SPM). Such an approach allows the quick variation of conformation and composition of (supra)molecular systems and an assessment of these parameters on the envisioned functionality. However, monitoring the molecular conformations during manipulations remain elusive due to the dual role of the SPM tip as an actuator and an imaging probe. We present an approach which enables monitoring based on continuously gathered force gradient data using a particle filter approach, which solves the inverse problem of conformation monitoring by comparing current force gradient data to a structured set of simulations stored in the form of a finite state automaton. This allows using molecular simulations with wall-times for completion much longer than the time scale of the experiments. Our proof-of-principle investigations are based on the vertical SPM manipulation of a PTCDA (3,4,9,10-perylene-tetracarboxylic dianhydride) molecule on the Au(111) surface.

O 14.5 Mon 16:00 S053

Real-space imaging of σ-hole by means of Kelvin probe force microscopy. — •Aurelio Gallardo[1], Benjamín Mallada[2], Bruno de la Torre[2], and Pavel Jelínek[1] — [1]FZU of the CAS, Prague, Czech Republic — [2]RCPTM-CATRIN, Palacký University, Olomouc Czech Republic

Anisotropic charge distributions on individual atoms, such as σ-holes, are crucial for the structural properties of certain systems. Nevertheless, the existence of σ-holes has only been demonstrated indirectly, either observing the interaction between halogenated molecules or by theoretical calculations. However, there was no experimental technique that would allow the spatail resolution of anisotropic atomic charges.

To tackle this problem, we employed Kelvin probe force microscopy (KPFM) which imaging mechanism relays on the electrostatic tip-sample interaction. To achieve the requested resolution, we developed a theoretical description of the KPFM imaging mechanism on atomic scale, which enables optimize the experimental setup. Namely we demonstrated both theoretically and experimentally that probe tip functionalization by a single Xe atom enhances the spatial resolution to directly visualize the anisotropic charge of the σ-hole, as well as the quadrupolar character of the carbon monoxide molecule. [1]

We believe that this work large already outstanding imaging capabilities of scanning probe techniques. In particular, this KPFM technique will enable better description of charge distribution in molecular complexes as well as on surfaces.

References: [1] Mallada et al., Science 374, 863-867 (2021)

O 15: Solid-Liquid Interfaces 2: Structure and Spectroscopy

Time: Monday 15:00–18:00 Location: S054

Topical Talk O 15.1 Mon 15:00 S054

Hydration Layer Mapping at Solid-Liquid Interfaces — •Angelika Kühnle — Physical Chemistry I, Department of Chemistry, University Bielefeld, Universitätsstraße 25, 33615 Bielefeld, Germany

Solid-liquid interfaces are omnipresent in nature and technology. Under ambient conditions, the properties of many materials are governed by a thin layer of water at the interface. Understanding processes occurring at the solid-liquid interface thus almost always requires a detailed knowledge of the hydration structure at the interface. Recent improvements in atomic force microscopy (AFM) instrumentation now enable molecular-level insights into the three-dimensional (3D) solvation structure at the interface.

In this talk, the capability of 3D AFM will be presented by discussing the hydration structure at the gypsum-water interface. Gypsum, the dihydrate of calcium sulfate, is an abundant rock-forming mineral in the Earth's crust. It is composed of alternating bilayers of calcium sulfate and water. Upon cleavage, the crystal water is exposed. What is the fate of this crystal water at the aqueous interface? Comparing 3D AFM data with water density maps derived from molecular dynamics simulations allows for elucidating molecular-level details of the gypsum-water interface. Our findings indicate that the crystal water at the interface remains tightly bound, even when in contact with bulk water. Thus, the interfacial chemistry of gypsum is governed by the crystal water rather than the calcium or sulfate ions.

O 15.2 Mon 15:30 S054

Self-assembly and thin film growth dynamics of an ionic liquid on Au(111) investigated in real space — Manuel Meusel[1], Matthias Lexow[1], Afra Gezmis[1], Simon Schötz[1], Margareta Wagner[2], •Simon Jaekel[1], Andreas Bayer[1], Florian Maier[1], and Hans-Peter Steinrück[1] — [1]Chair of Physical Chemistry II, University of Erlangen-Nürnberg (FAU), Germany — [2]Institute of Applied Physics, Technical University of Vienna, Austria

Ionic liquids (IL) are organic salts with low melting points, often at or even below room temperature. They have shown promise as solvents and electrolytes, but have also become part of novel catalytical concepts involving solid metal catalysts.

In this context, our group studied the self-assembly and growth dynamics of thin films of 1,3-dimethylimidazolium bis[(trifluoromethyl)sulfonyl]imide ($[\mathrm{C_1C_1Im}][\mathrm{Tf_2N}]$) on the model surface of Au(111) using scanning probe microscopy[1,2,3]. Our experiments show that the film undergoes distinct phases between the growth of the wetting layer and subsequent multilayers. Further, 2D film growth is shown to be in competition with the growth of a metastable 3D droplet phase, with the dominant growth mode determined by a combination of temperature and nucleus formation.

[1] Meusel et al. ACS Nano 14 (2020) 9000-9010

[2] Meusel et al. Langmuir 36 (2020) 13670-13681

[3] Meusel et al. J. Phys. Chem. C 125 (2021) 20439-20449

O 15.3 Mon 15:45 S054

Hydration layers at the graphite-water interface: Attraction or confinement? — Hagen Söngen[1], Ygor Morais Jaques[2], Lidija Zivanovic[2], Sebastian Seibert[1], •Ralf Bechstein[1], Peter Spijker[2], Hiroshi Onishi[3], Adam S. Foster[2,4], and Angelika Kühnle[1] — [1]Physical Chemistry I, Bielefeld University, Germany — [2]COMP Centre of Excellence, Department of Applied Physics, Aalto University, Helsinki, Finland — [3]Department of Chemistry, Kobe University, Japan — [4]Division of Electrical Engineering and Computer Science, Kanazawa University, Japan

Water molecules at solid surfaces typically arrange in layers. The physical origin of the hydration layers is usually explained by (1) the attraction between the surface and the water and/or (2) the confinement of water due to the presence of the surface. While attraction is specific for the particular solid-solvent combination, confinement is a general effect at surfaces. A differentiation between the two effects is critical for interpreting hydration structures. At the graphite-water interface, the solid-solvent attraction is often considered to be negligible. Nevertheless, we observe hydration layers using three-dimensional atomic force microscopy at the graphite-water interface. We use Monte Carlo simulations to explain why confinement alone could cause the formation of hydration layers. With molecular dynamics simulations, we show that at ambient conditions, there is a significant graphite-water attraction which is pivotal for the formation of layers at the graphite-water interface.

[1] H. Söngen et al., Physical Review B, 100 (2019) 205410

O 15.4 Mon 16:00 S054

Investigation of the wetting layer of [C1C1Im][Tf2N] on Pt(111) by variable temperature scanning tunneling microscopy — •Afra Gezmis, Simon Jaekel, Manuel Meusel, Andreas Bayer, Florian Maier, and Hans-Peter Steinrück — Lehrstuhl für Physikalische Chemie II, Universität Erlangen-Nürnberg, Egerlandstr. 3, 91058 Erlangen, Germany

With the introduction of Ionic liquids (IL) novel catalytic concepts like the Solid Catalyst with Ionic Liquid Layer (SCILL) approach have emerged. In a SCILL system, a high surface area solid substrate is covered with a thin IL film, and this film modifies catalytically active surface sites at the support. In order to gain better insights in the underlying effects, it is crucial to obtain a detailed understanding of the IL/solid interface. Due to the low vapor pressure of ILs, these interfaces can be investigated in ultrahigh vacuum by surface science methods. Herein, we present our first study on the adsorption behavior of 1,3-dimethylimidazolium bis[(trifluoromethyl)sulfonyl]imide ([C1C1Im][Tf2N]) on the reactive Pt(111) surface by variable-temperature scanning tunneling microscopy. We investigated the effect of temperature for coverages up to a closed wetting layer, in particular the formation of 2D islands and their temperature-dependent size. Interestingly, we were even able to detect mobile, single ion pairs on the surface, while for the 2D structures only limited mobility was seen.

Supported through an ERC Advanced Grant to H.P.S (#693398 ILID)

O 15.5 Mon 16:15 S054

bias-dependent switching of molecular nanostructures at the liquid-HOPG interface: the influence of concentration — •Baoxin Jia[1], Mihaela Enache[1], Sandra Miguez-Lago[2], Milan Kivala[2], and Meike Stöhr[1] — [1]Zernike Institute for Advanced Materials, University of Groningen, Netherlands — [2]Institute of Organic Chemistry, University of Heidelberg, Germany

Here we discuss the influence of concentration on the bias-dependent switching of a carboxy-functionalized triarylamine derivative at the nonanoic acid-HOPG interface studied by STM. For a fully saturated solution, a porous phase (chickenwire) was observed for negative sample bias and a close-packed phase for positive sample bias. For a 50% saturated solution, a second porous phase (flower) coexisted with the chickenwire phase at negative sample bias, while the close-packed phase was observed at positive sample bias. For a 20% saturated solution, the two porous phases and the close-packed phase coexisted at positive sample bias, while the two porous phases were observed at negative bias. For all concentrations investigated, a reversible phase transformation between the porous phases and the close-packed phase was accomplished by changing the bias polarity. Additionally, the switching behaviour for a 10:1 mixture of the triarylamine derivatives and 1,3,5-tris(4-carboxyphenyl) benzene molecules at the interface was studied. No intermixed structures were observed. Instead, both molecules formed networks separately but still showed a bias-induced phase transformation. However, the switching occurred for each molecule separately and no effect of cooperativity was detected.

O 15.6 Mon 16:30 S054

In-situ investigation of surface band-bending in the ZnO(0001)-OH/electrolyte interface via the excitonic response. — •Luis Rosillo-Orozco, Christoph Cobet, and Kurt Hingerl — Johannes Kepler University, Linz, Austria

In recent years, the effects of adsorbates on the surface band-bending in ZnO have been studied in UHV and characterized by Valence-Band XPS [1],[2]; showing an alteration of the space-charge region due to the electron transfer that occurs in the absorption processes. In this work we aim to understand the surface optical properties of a hydroxide stabilized ZnO(0001)-OH surface, previously obtained by chemical etching by Valtiner et al. [3], in contact with 0.1M NaClO4 as an electrolyte at a certain applied potential. By changing the cell applied potential, we can, in principle, produce any form of band bending at the semiconductor surface [4] including the flat-band condition. We use in-situ spectroscopic ellipsometry while varying the cell applied potential in order to study the response of the discrete excitons due to the inner electrical fields created by the band bending. We can identify the flat-band potential (Vfb) as the one where the imaginary part of the pseudo-dielectric function shows no change at the energy corresponding to the discrete exciton transition. By modulating the potential barrier at the interface we are able to investigate the response of the excitons at any given applied potential. Furthermore, given the significant sensibility of the band-bending to a surface dipole change, it is possible to use this technique to look into the effects of a modification of the surface such as adsorbates.

O 15.7 Mon 16:45 S054

Dynamic Polymorph Formation of a Trimesic Acid Derivative at Solid-Liquid Interface — •Richa Arjariya[1], Vipin Mishra[1], Gagandeep Kaur[1], Sandeep Verma[1], Markus Lackinger[2], and Thiruvancheril G Gopakumar[1] — [1]Department of Chemistry, Indian Institute of Technology Kanpur, Kanpur, UP-208016, India — [2]Department of Physics, Technical University of Munich, James-Franck-Strasse 1, Garching 85748 and Deutsches Museum, Museumsinsel 1, Munich 80538, Germany

In this work we show the self-assembly of a tricarboxylic acid derivative of trimesic acid (BTA) at heptanoic acid-, nonanoic acid-graphite interface. At both interfaces BTA forms a trimer based-self-assembly, super-flower (SF) pattern. The spontaneously formed SF pattern is observed to be dynamically converting to a dimer-based assembly, chicken-wire pattern (CW), at heptanoic acid-graphite interface while scanning. Interestingly, at nonanoic acid-graphite interface, SF pattern remains stable and not converted to CW pattern. We attribute that the formation energy of both SF and CW patterns is comparable. The difference in the stability of different patterns in heptanoic acid and nonanoic acid is most likely related to the solubility of BTA in these solvents.[1,2]

1) T. N. Ha, T. G. Gopakumar, M. Hietschold, J. Phys. Chem. C, 2011, 115, 21743.

2) M. Lackinger, S. Griessl, W. M. Heckl, M. Hietschold and G. W. Flynn, Langmuir, 2005, 21, 11, 4984-4988

O 15.8 Mon 17:00 S054

in-situ optical probe of chloride-induced surface states in Cu(110)/liquid interfaces — •Saul Vazquez-Miranda[1], Kurt Hingerl[1], and Christoph Cobet[1,2] — [1]Johannes Kepler Universität Linz Altenberger Straße 69 4040 Linz, Austria — [2]Johannes Kepler Universität, Linz School of Education, A-4040 Linz, Austria

While surface states (SSs) appearance and role in metal-electrolyte interfaces are still a controversial debate. The existence of SSs, permits control and tunability of electronic properties of metal-electrolyte interfaces. Resonant excitations among them could enhance, e.g. photocatalytic reactions. SSs and other properties are readily adjustable via an applied electrical potential that is, by promoting changes in the adsorption of ionic species. The electrolyte induces additional scattering and screening effects, so that the electron charge distributions can differ considerably in the presence of high electric fields. We report, by means of electrochemical impedance spectroscopy (EIS) jointly with in-situ reflectance anisotropy spectroscopy (RAS), which aimed to assess the evolution of surface properties and SSs occurring at Cu (110) in contact with an HCl solution. Thereafter, by modeling the RAS response and in comparison with EC-STM measurements, specific surface structures have been identified and ascribed to the optical response. In a specific potential range, three additional resonances are detected in RAS that can be explained by two-dimensional confined SSs.

O 15.9 Mon 17:15 S054

In-situ electrochemical X-ray photoelectron spectroscopy as laboratory technique to study the electrified interface — •Christoph Griesser, Daniel Winkler, Toni Moser, and Julia Kunze-Liebhäuser — Department of Physical Chemistry, University of Innsbruck, Innrain 52c, Innsbruck, Austria

The interface between a charged metal and an aqueous electrolyte is the most commonly studied in electrochemical surface science, as its properties determine the reactivity of many systems relevant for technological applications. More specifically, in the electrocatalytic conversion of energy, the activity, selectivity and efficiency are determined by the charge transfer between electrolyte and electrode, which is governed by the interfacial properties of the system. Therefore, a fundamental understanding of the interplay between applied potential and surface/interface chemistry is pivotal to further advance energy conversion and storage technologies. While there are several in-situ methods, to characterize the surface structure under reaction conditions, the surface chemistry (i.e. the elemental composition of the surface and the oxidation state of the components) itself is still most often investigated via ex-situ X-ray photoelectron spectroscopy (XPS). This work presents first in situ electrochemical XPS results obtained with a laboratory near ambient pressure (NAP-) XPS system. We show, that it is possible to track the oxidation state of a bulk gold (Au) electrode under reaction conditions, i.e. during anodic oxidation.

O 15.10 Mon 17:30 S054

Structure dependent product selectivity of the CO electroreduction on Au(111) electrodes modified with Cu adatoms — •Daniel Winkler, Toni Moser, Christoph Griesser, Matthias Leitner, and Julia Kunze-Liebhäuser — University of Innsbruck, Innrain 52c, 6020 Innsbruck, Austria

The efficient conversion of carbon dioxide (CO_2) into valuable hydrocarbons could be a promising solution for storage of excess energy and carbon neutral transportation. The electrochemical reduction of CO_2 has been extensively studied on different monometallic surfaces, where Cu remains the only metal providing a sufficient formation activity to value-added products, such as methane or ethylene. Despite this beneficial behavior, further strategies to increase the product selectivity must be found. This can be accomplished by understanding the exact CO_2 reduction reaction (CO_2RR) mechanism, which remains one of the most challenging problems. Here we focus on the carbon monoxide (CO) reduction on Au(111) electrodes modified with different coverages of Cu adatoms to address this issue. Differential electrochemical mass spectrometry (DEMS) shows an increase in selectivity for the formation of ethylene at low Cu coverages. *In situ* electrochemical scanning tunneling microscopy (EC-STM) and X-ray photoelectron spectroscopy results suggest that two different neighboring CO adsorption sites present at the interface between the metallic Cu islands and the Au(111) surface, which is maximized at low coverages, provide ideal conditions for a facilitated CO-CO coupling reaction resulting in an enhanced formation of ethylene.

O 15.11 Mon 17:45 S054

Electrochemical reflection anisotropy spectroscopy for time-resolved interface structures in aqueous and non-aqueous electrolytes — •Matthias M. May[1,2], Margot Guidat[1,2], Mario Löw[2], Florian Keller[2], Justus Leist[2], and Jongmin Kim[1,2] — [1]Universität Tübingen, Institute of Physical and Theoretical Chemistry, Tübingen, Germany — [2]Universität Ulm, Institute of Theoretical Chemistry, Ulm, Germany

The microscopic structure of electrochemical interfaces determines many properties that are decisive for the performance of applications in catalysis or batteries. Yet access to this solid–liquid interface at sufficient temporal and spatial resolution is challenging and limits the understanding of this complex interface. Electrochemical reflection anisotropy spectroscopy (RAS) is a powerful emerging tool in spectroelectrochemistry [1], which we apply to a number of systems relevant for energy and matter conversion. Here, we present initial results on InP and Au in aqueous systems as well as select post-Li battery systems. For InP, we can directly observe stability windows with respect to applied potentials and electrolyte composition. We highlight the potential and challenges of electrochemical RAS from a perspective of both computational and experimental spectroscopy.

[1] May and Sprik, New. J. Phys. 20 (2018) 033031.

O 16: Poster Monday: Ultrafast Processes 1

Time: Monday 18:00–20:00 Location: P4

O 16.1 Mon 18:00 P4

Probing photo-induced, ultra-fast dynamics by time-resolved spectroscopic ellipsometry — •Felix-Florian Delatowski[1,3], Krishna Khakurel[1], Jörg Rappich[2], Shirly Espinoza[1], Mateusz Rebarz[1], Martin Zahradnik[1], and Jakob Andreasson[1] — [1]ELI Beamlines, Institute of Physics, Czech Academy of Sciences, Czech Republic — [2]Institute Silicon Photovoltaics, Helmholtz-Zentrum Berlin für Materialien und Energie, Germany — [3]Semiconductor Physics Group, Felix Bloch Institute for Solid State Physics, Universität Leipzig, Germany

Photo-induced phenomena, such as charge carrier and structural dynamics, charge transfer and relaxation processes on sub-picosecond time scales are of great interest for a profound understanding of light-matter interaction. Time-resolved spectroscopic ellipsometry is a promising surface sensitive tool to investigate these phenomena. In this work, we explore the potential of this measurement technique exemplary on germanium and gold and present preliminary experimental results.

In addition to the experimental observations, we also present an approach to predict ellipsometry spectra through ab-initio DFT calculations using the program SIESTA. In order to extend this approach, some preliminary measurements of organic molecules (Pyrimidine) have been performed. We discuss the benefits that the synergy of ab-initio based spectrum predictions and experiments bring in the interpretation of the dynamics in the sample.

O 16.2 Mon 18:00 P4

Tailoring the carrier dynamics of WSe$_2$ by the adsorption of CuPc — •Gregor Zinke[1], Sebastian Hedwig[1], Benito Arnoldi[1], Martin Aeschlimann[1], and Benjamin Stadtmueller[1,2] — [1]Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Erwin-Schroedinger-Str. 46, 67663 Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Staudingerweg 7, 55128 Mainz, Germany

Tailoring the electronic properties and carrier dynamics of 2D-Van-der-Waals materials is a highly promising way to design spin functionalities in low dimensions. This is often achieved by the formation of heterostructures with other Van-der-Waals materials.

Here, we functionalize the carrier dynamics of the prototypical Van-der-Waals material WSe$_2$ by the adsorption of the aromatic molecule CuPc. Using time- and angle-resolved photoemission with XUV-radiation, we investigate the temporal evolution of the excited states at the K- and Σ-points of WSe$_2$ after an optical excitation. For the bare WSe$_2$, we find a spin selective excitation at the K-point depending on the pump light polarization [1], which is followed by a spin-flip scattering from the K- to the Σ-point. After the adsorption of CuPc, the excitation scheme is completely altered. In particular, we uncovered a direct interlayer excitation from the CuPc into the WSe$_2$ layer that dominates the carrier dynamics of the CuPc/WSe$_2$ heterostructure.

Reference: [1] Bertoni et al.; Phys. Rev. Lett. 117, 277201 (2016)

O 16.3 Mon 18:00 P4

Numerous Improvements on Ultrafast Pump-Probe RHEED Experiment — •Jonas Fortmann[1], Christian Brand[1], Thorben Groven[1], Mohammad Tajik[1], Michael Horn-von Hoegen[1], and Thomas Duden[2] — [1]Department of Physics and Center for Nanointegration CENIDE, University of Duisburg-Essen, Lotharstraße 1, 47057 Duisburg — [2]Construction office, Mustangweg 17, D-33649, Bielefeld

During the last year, major improvements have been made to our ultrafast time-resolved reflection high energy electron diffraction (tr-RHEED) experiment. In particular, the previous detection unit consisting of a multichannel plate and a cooled CCD camera has been replaced by a single electron sensitive CMOS based camera (TVIPS TemCam XF-416), which allows spot profile analysis with its superior resolution and signal-to-noise ratio and no blooming. Flatfielding of the detector is achieved with a home-built electron source at 20 keV accompanied by an external EM-deflection unit. In addition, the 80 fs-laser pump pulse ($\lambda = 800\,\mathrm{nm}$) has been upgraded with an optical parametric amplifier (Topas Prime) that allows excitation of the sample's surface at $1.16 - 2.60\,\mu\mathrm{m}$ in future experiments. Re-routing of the beamline has shortened the beam paths by $\sim 1.5\,\mathrm{m}$ and saved half of the mirrors, which provides higher beam stability. Also the third harmonic generation (THG) stage for electron generation in the Au film photocathode has been optimized. A new lab software has been established on the basis of TANGO controls. First experimental data with the new setup are shown.

O 16.4 Mon 18:00 P4

Ultrafast optical spectroscopy of few-layer TMDCs under ultrahigh-vacuum conditions — •Maximilian Franz, Kilian Kuhlbrodt, Jan Gerrit Horstmann, and Claus Ropers — Max-Plank-Institut für Multidisziplinäre Naturwissenschaften, Göttingen, Germany

Optical pump-probe spectroscopy (OPP) has enabled detailed investigations of the nonequilibrium optical properties of atomically thin materials, with prominent examples in graphene or transition metal dichalcogenide (TMDCs) heterostructures [1]. However, these materials are often susceptible to oxidation and contamination under atmospheric conditions, resulting in strongly altered physical and chemical properties [2]. Here, we present the development of an OPP setup for the investigation of dynamics in few-layer TMDCs and heterostructures under ultrahigh vacuum (UHV) conditions. We present first results on the ultrafast phase transition in $1T$-TaS$_2$ samples using time-resolved reflectance and transmittance measurements with fs temporal and μm spatial resolutions. Furthermore, we discuss possible applications, e.g., layer-selective optical excitation of TMDC heterostructures, and the combination of OPP with structure-sensitive techniques like ultrafast low-energy electron diffraction [3].

[1] C. Jin et al., Nature Nanotechnology 13, 994-1003 (2018); [2] Y. Yu et al., Nature Nanotechnology 10, 270-276 (2015); [3] S. Vogelgesang et al., Nature Physics 14, 184-190 (2018)

O 16.5 Mon 18:00 P4

Description for the electronic non-equilibrium after ultrashort laser excitation of metals — •Markus Uehlein, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany

When a metal is excited by a femtosecond laser pulse in the visible range, electrons absorb the energy, resulting in a non-equilibrium energy distribution. This absorption behavior and the following thermalization to a hot Fermi distribution can be simulated using complex and numerically expensive Boltzmann collision integrals [1]. After the thermalization, the two-temperature model (TTM) can describe the relaxation of the heated electrons and the phonons in a simple way. It is, however, unable to trace non-thermal electrons.

We present an intermediate model, called extended TTM (eTTM). It was developed in Refs. [2, 3] and adds a system of non-equilibrium electrons to the TTM. Besides some improvements to the published versions, we compare the eTTM to the Boltzmann model as well as to the TTM [4]. In particular, we compare the spectral particle dynamics to a time-resolved two-photon photoemission measurement [5].

[1] B. Y. Mueller and B. Rethfeld; PRB **87**, 035139 (2013)
[2] E. Carpene; PRB **74**, 024301 (2006)
[3] G. D. Tsibidis; Appl. Phys. A **124**, 311 (2018)
[4] M. Uehlein, S. T. Weber and B. Rethfeld;
Nanomaterials **12**, 1655 (2022)
[5] Y. Beyazit *et al.*; Phys. Rev. Lett. **125**, 076803 (2020)

O 16.6 Mon 18:00 P4

A setup for interferometrically time-resolved multi-photon photoemission benchmarked on Ag(111) — •Hannah Strauch, Marco Merboldt, Jan Philipp Bange, Daniel Steil, G. S. Matthijs Jansen, Sabine Steil, Marcel Reutzel, and Stefan Mathias — I. Physikalisches Institut, Georg-August-Universität Göttingen, Göttingen, Germany

Time- and angle-resolved photoemission is the method of choice to directly study the electron dynamics of condensed matter systems with energy and momentum resolution. After the system is excited by a pump pulse, a delayed second pulse induces photoemission and thus probes the materials response. To complement the obtained insight with information on the coherence of the response, single-color phase-locked pulse pairs can be employed. Here, we present a setup for interferometric time-resolved multi-photon photoemission based on a passively stabilized Mach-Zehnder interferometer which produces phase-locked pulse pairs and provides an optical delay precision below 60 as. We show interferometric time- and angle-resolved multi-photon photoemission data from the Shockley surface state and the first image potential state of the Ag(111) surface as a benchmark for our setup. Supported by an optical Bloch equation model we explore possibilities to interpret such a data set.

A promising application of the interferometer is in combination a high harmonic generation EUV beamline. This enables double-pump-probe experiments with access to the full Brillouin zone, and allows a comprehensive investigation of optical excitations.

O 16.7 Mon 18:00 P4

Time-resolved ARPES probing *Rabi* Oscillations and *Landau-Zener-Stückelberg* Interferences in Graphene - a Proposal — •Eduard Moos, Hauke Beyer, and Michael Bauer — Institute of Experimental and Applied Physics, Kiel University, Germany

At sufficiently high intensities, the interaction of few-cycle laser fields with solids gives rise to strong-field effects envisioning novel and exciting strategies for controlling optical and electronic properties via the electric field waveform on sub-femtosecond timescales. A striking example is the CEP-control of light-field-driven currents in graphene due to *Landau-Zener-Stückelberg* (LZS) interferences [1] resulting from the complex interplay of field-driven adiabatic intraband and diabatic interband transitions. Simulations show that the LZS-interferences give rise to characteristic asymmetries in the momentum-distribution of the residual conduction band population in the Dirac cone on top of a symmetric quasi-periodic pattern indicative for *Rabi* oscillations. TRARPES using few cycle femtosecond pump laser pulses seems to be in ideal tool for the investigation of these processes and there dependence on parameters such as pulse peak electric field strength E0, laser polarization, and CEP phase. Based on preliminary results on graphite using 7 fs few-cycle NIR pump-pulses we will discuss in this presentation the prospects, but also the challenges that arise in such type of TRARPES experiment.

[1] T. Higuchi, *et al.*, Nature **550**, 224 (2017)
[2] G. Rohde, *et al.*, Phys. Rev. Lett. **121**, 256401 (2018)

O 16.8 Mon 18:00 P4

Laser-based low energy photoelectron diffraction of SnPc/graphite — •Hermann Erk, Stephan Jauernik, Petra Hein, and Michael Bauer — IEAP, CAU Kiel, Germany

Tin-phthalocyanine (SnPc) adsorbed on graphite has been studied using laser-based angle resolved photoemission spectroscopy (ARPES) at 5.9 eV photon energy and low energy electron diffraction (LEED). An ordered SnPc monolayer was prepared by thermal evaporation onto single crystalline graphite (SCG) flakes and subsequent annealing at 370 K. LEED data reveal an incommensurate ordered phase of the SnPc overlayer at room temperature. In the ARPES spectra the SnPc long-range order can be seen due to an adsorbate-induced backfolding of the band structure of graphite from the $\overline{\mathrm{K}}$ point to the center of the Brillouin zone. A comparison of ARPES spectra with simulations of the backfolded band structure under consideration of the LEED data will be discussed. First time-resolved ARPES measurements with the aim to address ultrafast changes in the SnPc long range order will be presented.

O 16.9 Mon 18:00 P4

Type II aligned TMD heterostructures: Probing moiré interlayer excitons with energy-, momentum-, and femtosecond time-resolution — •Marcel Reutzel[1], David Schmitt[1], Jan Philipp Bange[1], Wiebke Bennecke[1], AbdulAziz AlMutairi[2], Giuseppe Meneghini[3], Daniel Steil[1], R. Thomas Weitz[1], Sabine Steil[1], G. S. Matthijs Jansen[1], Samuel Brem[3], Ermin Malic[3], Stephan Hofmann[2], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Göttingen, Germany — [2]Department of Engineering, University of Cambridge, Cambridge CB3 0FA, U.K. — [3]Fachbereich Physik, Philipps-Universität, 35032 Marburg, Germany

Transition metal dichalcogenides (TMDs) can be stacked into atomically thin p-n junctions, where an optically excited intralayer exciton can decay into interlayer excitons. Here, the electron and the hole contribution to the quasiparticle reside in the neighbouring TMD layers.

On this poster, we show our recent progress towards the identification and characterization of the ultrafast charge transfer process across a type II interface. Using femtosecond momentum microscopy, we identify the distinct momentum fingerprints of various excitonic states in this system, notably the bright and dark intralayer excitons as well as the interlayer exciton. Most intriguingly, the momentum-resolved measurement provides quantitative access to the interlayer exciton wavefunction that is modulated within the moiré potential.
Schmitt *et al.*, *arXiv:2112.05011 (2021).

O 16.10 Mon 18:00 P4

Influence of the static dielectric permittivity on ultrafast quasiparticles dynamics in $\mathbf{WS_2}$ monolayers — •Subhadra Mohapatra[1,2], Stefano Calati[1,2], Quiyang Li[3], Xiaoyang Zhu[3], and Julia Stähler[1,2] — [1]Humboldt-Universität zu Berlin, Institut für Chemie,Berlin,Germany — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Abt. Physikalische Chemie, Berlin, Germany — [3]Columbia University, New York City, New York

In our recent fluence-and photon energy-dependent studies [1,2] of quasiparticle dynamics in WS_2 monolayers on fused silica (FS) and Si-SiO_2 substrates, we observed that excitonic screening solely reduces the binding energy of the excitons, leading to a transient blue-shift of the exciton resonance while quasi free carrier screening effectively shows a red shift, as the carrier-induced screening leads to a larger band gap renormalization than binding energy reduction. Further investigation of such fluence-dependent quasiparticle dynamics studies using a higher dielectric permittivity of a sapphire substrate, we found that scattering rates, relaxation time constants, and band gap renormalization are not influenced by the dielectric permittivity. On the contrary, the ratio of dynamic screening parameter of the excitons and their Bohr radius is approximately 4 times higher in sapphire than for FS, which must be a direct consequence of the increased dielectric permittivity likely leading to more localized excitons.

References:
[1] Calati et al. PCCP 23(39) (2021).
[2] Calati et al. arXiv:2204.02125 (2022).

O 16.11 Mon 18:00 P4

Ultrafast Transport and Energy Relaxation of Hot Electrons in Au/Fe/MgO(001) Investigated by Linear Time-resolved Photoelectron Spectroscopy — •Florian Kühne[1], Yasin Beyazit[1], Detlef Diesing[2], Ping Zhou[1], Jesumony Jayabalan[1], and Uwe Bovensiepen[1] — [1]University of Duisburg-Essen,Physics — [2]University of Duisburg-Essen, Chemistry

Optically excited electrons and holes are of particular interest in solid-state physics because analysis of their dynamics allows a microscopic understanding of the interactions in non-equilibrium states. Here we want to discern the relaxation by such local inelastic processes and non-local transport effects. To analyze the ultrafast dynamics of charge carriers in the vicinity of the Fermi energy E_F, femtosecond time-resolved linear photoelectron spectroscopy was applied. We report on first experimental results obtained by using 1.55 eV pump and 6 eV probe photons on an Au/Fe/MgO(001) epitaxial heterosystem, complementary to previous work in Beyazit et al., PRL 125, 076803 (2020). By pumping the Fe side, hot electrons are excited in the Fe layer, and subsequently injected into the Au layer and propagate to the surface, where they are probed by photoelectron emission spectroscopy. In the Fe side pumped data, we observe thickness dependent differences in relaxation compared to the Au side pumped data. In Au side pumping we observe efficient transport into the Fe layer. We will present an energy E dependent analysis of the propagation and energy density $U(E, d_{Au})$ of electrons above E_F, which together with a two-temperature model shows a super diffusive transport limit.

O 16.12 Mon 18:00 P4

Investigations of polarons in hematite α-$\mathbf{Fe_2O_3}$(1-102) by means of nc-AFM and KMC — •Jesús Redondo[1,2], Vít Gabriel[1], Giada Franceschi[3], Igor Sokolović[3], Dominik Wrana[1], Florian Kraushofer[3], Erik Rheinfrank[3], Michele Riva[3], Gareth S. Parkinson[3], Michael Schmid[3], Ulrike Diebold[3], Pavel Kocán[1], and Martin Setvin[1,3] — [1]Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic — [2]Faculty of Chemistry, University of the Basque Country, San Sebastián, Spain — [3]Institute of Applied Physics, Vienna University of Technology, Vienna, Austria

Polarons are known to strongly influence the catalytic activity and the electronic, magnetic, and structural properties of transition metal oxides and halide perovskites. The study of polaron formation and dynamics is fundamental to understanding the actual mechanisms and yields of catalytic reactions in these materials. A new method for the investigation of electron and hole polarons is demonstrated. Charge carriers are injected with the AFM/STM tip into the surface of natural, Ti- and Ni-doped α-Fe2O3(1-102). The injected charges form a cloud of electrons or holes trapped in the lattice. This cloud expands due to electrostatic interactions and thermally activated polaron hopping. Controlled annealing of the sample and characterization by Kelvin probe force microscopy (KPFM) provides information on polaron dynamics; these results are compared to kinetic Monte Carlo (KMC) simulations and the dependence of the polaron hopping activation energy on the doping is shown.

O 17: Poster Monday: Organic Molecules at Surfaces 1

Time: Monday 18:00–20:00 Location: P4

O 17.1 Mon 18:00 P4

Influence of a BlueP interlayer on the properties of P2O on Au(111) — •Florentine Friedrich, Maximilian Schaal, Felix Otto, Philip Grimm, Marco Gruenewald, Roman Forker, and Torsten Fritz — Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany

Two-dimensional (2D) materials have attracted much attention in solid state physics in recent years due to their unique properties. Amongst others, one novel representative is the phosphorus allotrope blue phosphorene (BlueP) with its high charge carrier mobility and suitable band gap, that is used here as an interlayer to obtain a decoupling of the functional organic molecule 6,13-pentacenequinone (P2O) from Au(111) surfaces. P2O is deposited in various film thicknesses (1 monolayer equivalent (MLE) up to 4-5 MLE) either on BlueP on Au(111) or directly on the metal substrate to investigate the influence of BlueP as an interlayer on the optical and structural properties of the P2O molecules. The film growth of P2O is monitored by in-situ differential reflectance spectroscopy (DRS), while the BlueP-Au substrate is kept at different temperatures. Further complementary experimental methods were low-electron energy diffraction (LEED), X-ray photoelectron spectroscopy (XPS), and low-temperature scanning tunneling microscopy (LT-STM). It turns out that the BlueP interlayer influences the monolayer structure as well as the growth mode. In contrast to previous studies on C_{60}, the BlueP interlayer stays intact.

O 17.2 Mon 18:00 P4

Navigating the Polymorph-Jungle with Optimal Control techniques — •Simon Hollweger, Anna Werkovits, Richard K. Berger, and Oliver T. Hofmann — Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria

Properties of organic-inorganic interfaces are strongly dependent on the arrangement of the molecules on the surface. However, achieving a controlled growth of a polymorph with preferable physical properties is not straightforward at all - especially for metastable polymorphs.

In principle, starting from a thermodynamically easily accessible polymorph, kinetics can be utilized to stabilize a metastable polymorph. This can be done by promoting certain structural transitions by a-priori unknown sequentially changing growth conditions. Depending on the complexity of the transition network, often only a limited yield of the target polymorph can be reached during growth.

To computationally maximize the yield of the desired structure we use Optimal Control techniques. Therein, process parameters like temperature and pressure are varied to optimize parameter protocols. The transition network of our model system TCNE/Cu(111) is obtained via a combination of Transition State Theory and Density Functional Theory.

O 17.3 Mon 18:00 P4

Adsorption structure of mixed PTCDA derivatives on Ag(111) — •Amin Karimi[1], Jose M. Guevara[1], Veronika Schmalz[2], Ulrich Koert[2], F. Stefan Tautz[1], and Christian Wagner[1] — [1]Peter Grünberg Institut (PGI 3), Forschungszentrum Jülich , Jülich, Germany — [2]Chemistry Department, Philipps Universität Marburg, Marburg, Germany

For molecular manipulation and SPM tip functionalization, the mode of anchoring molecules to the tip is decisive. We recently found that PTCDA binds to a Ag tip with two oxygen-metal bonds after it is retracted from the surface. While this is crucial for its stabilization in a vertical state, the two bonds reduce the degrees of freedom during manipulation in the tip-molecule-surface junction. To overcome this problem, we synthesized a PTCDA derivative (reduced PTCDA), in which one carboxylic oxygen is replaced by two H atoms. For molecular manipulation, this allows choosing between one or two tip-oxygen bonds. Here we present a study on the absorption structures formed by a mixture of PTCDA and reduced PTCDA on the Ag(111) surface with the help of low-temperature NC-AFM/STM. We observe that despite the minimal modification of reduced PTCDA compare to PTCDA, the mixture of both molecules exhibits a strongly different behaviour compared to the regular island growth of pure PTCDA. Particularly striking is the coexistence of zero-, one-, and two-dimensional structures. A likely explanation for such diverse structures is the influence of long-range electrostatic interactions related to the in-plane dipole moment of reduced PTCDA.

O 17.4 Mon 18:00 P4

Single-domain molecular layers on Ag (110) — •Ravi Priya, Weishan Wu, and Peter Jakob — Department of Physics, Philipps-Universität Marburg, Germany

Single domain molecular layers have been explored on the non-hexagonal Ag(110) substrate. The absence of rotational domains for fcc (110) metal substrates allows for growing layers with uniform azimuthal orientation of deposited molecules. In our study we have investigated various molecules and configurations that may form single domain molecular layers (including mirror domains). Specifically, PTCDA, NTCDA, phthalocyanines (CuPc, SnPc, TiOPc) and regio-selectively substituted pentacene species (pentacene, pentacene-quinone, pentacene-tetrone, quinacridone) were deposited and examined in terms of their structure using SPA-LEED, and their vibrational signature using IR - spectroscopy. Among them, PTCDA, NTCDA and pentacene have been found to form single domain structures. In the case of PTCDA two prominent phases, the brick-wall (BW) and the herringbone (HB) phases exist, and they are readily distinguished not only by their LEED patterns but also from their vibrational signatures. Another finding refers to the increased molecule - metal interaction on Ag (110) vs. Ag (111) that leads to an extra energy (down)shift of the LUMO, thereby having a significant impact on interfacial dynamic charge transfer (IDCT) of vibrational modes [1].

[1] P. Jakob, S. Thussing, Phys. Rev. Lett. 126 (2021) 116801, DOI:10.1103/PhysRevLett.126.116801

O 17.5 Mon 18:00 P4

Generation of synthetic chiral structured images for computer vision applications — •Johannes Tim Seifert[1], Peer Kasten[1], Mandy Stritzke[2], Björn Möller[3], Timo de Wolff[2], Tim Fingscheidt[3], and Uta Schlickum[1] — [1]Institut für Angewandte Physik, Technische Universität Braunschweig — [2]Institut für Analysis und Algebra, Technische Universität Braunschweig — [3]Institut für Nachrichtentechnik, Technische Universität Braunschweig

Scanning tunneling microscopy (STM) is a well-established tool to measure surface topographs with atomic precision. Since data generation is slow using STM, we present a tool to create synthetic images with realistic noise and defects, which provides labeled training data for neural networks. We compare two methods evaluating chiral structures in STM images using Deep Learning.

As one approach to classify structures, we are using semantic segmentation based on the U-Net architecture to create maps showing the distribution of both chiralities. Additionally, we use a faster R-CNN architecture for object detection to locate and classify each chiral structure individually.

Since the approaches serve different use cases, they also have separate label requirements. We show that models trained using only our synthetic images perform successfully for real STM Images.

O 17.6 Mon 18:00 P4

Ultrafast dynamics of quantum confined surface electrons in a T4PT-based metal-organic network on noble metals — •Nils Bellenbaum[1], Lu Lyu[1], Eva Walther[1], Tobias Eul[1], Benito Arnoldi[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2]

— [1]Technische Universität Kaiserslautern and Research Center OPTIMAS, Erwin-Schrödinger Straße 46, 67663 Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Staudingerweg 7, 55128 Mainz, Germany

Two-dimensional metal-organic networks have emerged as intriguing architectures to design and control quantum confinment of electrons at surfaces. In our work, we focus on the metal-organic network T4PT/Cu(111) comprised of triangular 2,4,6-tris(4-pyridine)1,3,5-triazine (T4PT) molecules adsorbed on a copper substrate. Using momentum-resolved photoemission, we demonstrate a spatial confinement of the free electron-like surface state of Cu(111) in the pores of the network structure. Similarly, the dispersion of the unoccupied image potential state (IPS) of Cu(111) is modulated by the network's periodic potential, leading to a severe increase of the effective band mass of the IPS. Finally, we discuss the influence of periodic modulation of the IPS on its energy and momentum-dependent population time using timeresolved two photon momentum microscopy. Our findings provide a first glimpse onto tunabilitiy of the electron dynamics at surfaces by spatial confinement in nanoporous metal-organic network structures.

O 17.7 Mon 18:00 P4

Organic Molecules on the Cu(110)-(2x1)O Striped Phase — •Ilias Gazizullin, Christophe Nacci, and Leonhard Grill — Physical Chemistry Department, University of Graz, Heinrichstrasse 28, 8010 Graz, Austria

The deposition of molecules onto single-crystal surfaces allows their investigation at the single-molecule level by scanning tunneling microscopy (STM) and gives access to the controllable on-surface synthesis of 2D materials. Here, we have studied dibromo-p-terphenyl molecules on the Cu(110)-(2x1)O striped phase under ultra-high vacuum conditions with low-temperature STM. The Cu(110)-(2x1)O striped phase is of particular interest since it offers alternating stripes of (metallic) copper areas and of oxygen-covered areas where the adsorbed organic molecules are slightly decoupled from the metal substrate and hence have higher mobility.

Previously, the Cu(110)-(2x1)O striped phase was used as a template for the synthesis of organometallic structures having different sizes and shapes depending on the width of copper stripes [1]. The focus of our study is how annealing affects the molecular adsorption on the surface. It turns out that the molecules form organometallic chains on the copper areas, oriented in three surface directions. Increasing the sample temperature from 300 K to 450 K changes the orientation of the organometallic chains and the shape of the Cu-O areas. Possible interactions leading to such behaviour are discussed.

Reference: [1] Q. Fan, J. Dai, T. Wang, J. Kuttner, G. Hilt, J. M. Gottfried, and J. Zhu, ACS Nano, 3 (2016), 3747-3754

O 17.8 Mon 18:00 P4

Adsorption of phthalocyanine monolayers on Ag(110) — •Gaana Kainikkara, Ravi Priya, and Peter Jakob — Phillips University of Marburg

Vibrational properties and long range ordering of TiOPc, CuPc and SnPc molecular layers on the non-hexagonal Ag(110) surface have been investigated using IR-spectroscopy and SPA-LEED. Special emphasis is put on a comparison with the related Ag(111) substrate surface that is characterized by a somewhat weaker molecule - metal interaction strength. This hypothesis is supported by distinct differences in the vibrational line shapes of modes associated with interfacial dynamical charge transfer [1], the primary cause being an extra shift in the energetic position of the former LUMO with respect to the Fermi energy for the Ag(110) substrate. Another objective in our study was to identify structural changes in the (layered) structural arrangement, both during the growth process, as well as annealing of the layers. Due to their only slightly different footprint, the molecules display similar long range ordered phases. Our primary focus thus concerned the identification of possibly inclined arrangements in the coverage range of 1-2 monolayers [1] P. Jakob, Peter and S. Thussing. "Vibrational Frequency Used as Internal Clock Reference to Access Molecule-Metal Charge-Transfer Times." Phys. Rev. Lett. 126, 116801 (2021).

Part-O: Surface Science Division Type-Poster Presentation Topic-Organic Molecules on Inorganic Substrates: Adsorption and growth

O 17.9 Mon 18:00 P4

Growth of organic crystals on nanoparticle precovered surfaces studied by PEEM — •Katharina Engster[1], Jörg Heller[1], Thorsten Wagner[2], Sylvia Speller[1], and Ingo Barke[1] — [1]University of Rostock, Institute of Physics, 18059 Rostock, Germany — [2]Johannes Kepler University, Institute of Experimental Physics, 4040 Linz, Austria

Nanostructures can be used as plasmonic light sources [1] for local generation of excitons in organic semiconductors. This is a promising pathway to transfer energy in molecular aggregates from a defined starting point over longer distances via exciton propagation. In order to establish a suitable geometry of such a hybrid system, we are interested in the role of metallic clusters during subsequent molecular growth. Mass-selected silver nanoparticles are produced in a gas-phase cluster source and deposited onto a pristine HOPG surface. Afterwards, we use *in-situ* photoemission electron microscopy (PEEM) to monitor the physical vapour deposition of copper phthalocyanine (CuPc) molecules [2,3] on such surfaces. We observe a Stranski-Krastanov growth during deposition of CuPc and post-deposition further morphology changes of the nanocrystallites.

[1] K. Oldenburg et al., J. Phys. Chem. C **123**, 1379 (2019).
[2] Y.-C. Chiu et al., Cryst. Res. Technol. **46**, 295 (2011).
[3] T. Wagner et al., Ultramicroscopy **233**, 113427 (2022).

O 17.10 Mon 18:00 P4

Visualizing self-assemblies of electrosprayed complex molecules on surface: a non-contact force microscopy study — •Gema Navarro[1], Antoine Hinaut[1], Sebastian Scherb[1], Shuyu Huang[1], Yiming Song[1], Klaus Müllen[1], Thilo Glatzel[1], Akimitsu Narita[2], and Ernst Meyer[1] — [1]Department of Physics, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland. — [2]Max Planck Institute for Polymer Research, Ackermannweg 10, D-55128 Mainz, Germany.

Building up functional 2D supramolecular assemblies requires the unit blocks involved in their bottom up formation to be already complex. In the construction of the tailored nanostructures, different approaches can be adopted in order to circumvent the limitations of the traditional wet chemical methods [1,2]. Among them, specifically the Electrospray Deposition (EDS) technique emerges as suitable pathway to explore the adsorption of organic compounds with higher structural complexity [3].

In the present work, we report the adsorption of complex molecules on a noble metal surface. Our study focus on the influence of intermolecular and surface-molecule interactions in the observed assemblies. The deposition of the molecules is carried out by HV-EDS. Furthermore, visualization of the molecules is achieved by means a home-made Atomic Force Microscopy (AFM) set-up, in non-contact operation mode.

[1] Z. Qiu, A. Narita, and K. Müllen, Faraday Discuss. 227,8 (2021).
[2] S. Scherb et al., Commun Mater 1, 1 (2020).
[3] I. C.-Y. Hou, A. Hinaut, Chem. Asian J., e202200220 (2022).

O 17.11 Mon 18:00 P4

C_{60} - PEN Diels - Alder Cycloaddition Reaction on Ag(110) — •Mohammed Suhail Ansari, Ravi Priya, and Peter Jakob — Department of Physics, Philipps - Universität Marburg, Germany

Ultrathin films of C_{60} and pentacene (PEN) have been prepared on Ag(110) and their vibrational properties, as well as thermal evolution investigated by IR spectroscopy. Monolayer species are found to display distinctly different vibrational signatures as compared to higher layers, so the characterization of the grown hetero layers is straightforward. Moreover, the excellent spectral resolution and chemical selectivity allowed us to unambiguously identify the C_{60} - PEN Diels-Alder adduct [1]. In our thermal evolution study, the formation and decomposition of the C_{60} - PEN reaction product as well as the abundances of the C_{60} and PEN compounds have been examined. Production of the adduct species requires a weak thermal activation (annealing to 350 K) and proceeds largely independent of the C_{60} / PEN stacking sequence. We find that this reaction occurs exclusively at the (abrupt) interface of C_{60} and PEN for layered arrangements and is fully reversible, meaning that further annealing to 450 K (and higher) produces again C_{60} and PEN, with the latter desorbing thermally and thus depleting the initial concentration of reactants.

[1] F. Cataldo, D. A. García-Hernández and A. Manchado, On the C_{60} Fullerene Adduct with Pentacene: Synthesis and Stability, Fullerenes, Nanot. Carbon Nanostruct. 23 (2015) 818-823.

O 17.12 Mon 18:00 P4

First Steps Towards Carbon-Based Heisenberg Chains on Au(111) — •Nils Krane[1], Elia Turco[1], Annika Bernhardt[2], Michal Juríček[2], Pascal Ruffieux[1], and Roman Fasel[1] —

[1]nanotech@surfaces Laboratory, Empa - Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland — [2]Department of Chemistry, University of Zurich, Zurich, Switzerland

Carbon based π-magnetism has been a growing field of interest in the recent years. By tailoring nanographenes with atomically price shape and size it is possible to tune their electronic properties and create magnetically non-trivial groundstates. A recent study has demonstrated the formation of triangulene-based Haldene chains, consisting of spin-1 building blocks and featuring gapped spin excitation in the bulk [1]. On the other hand, a Heisenberg chain consisting of coupled spin-1/2 units is predicted to be gapless, due to a continuum of spin excitations above the ground state.

Here we present the first steps towards synthesizing all-carbon spin-1/2 chains. Phenalenyl-based precursers are used for a bottom-up approach to form oligomers of different lengths and spin-excitation gaps. The low activation barrier of hydrogen-passivated phenalenyl was found to be a potential inhibitor of the radical formation, which is required for the covalent coupling of the building blocks.

[1] S. Mishra et al., Nature **598**, 287-292 (2021). https://doi.org/10.1038/s41586-021-03842-3

O 18: Poster Monday: 2D Materials 1

Time: Monday 18:00–20:00 Location: P4

O 18.1 Mon 18:00 P4

Scanning tunneling microscopy and spectroscopy of rubrene on clean and graphene-covered metal surfaces — •Karl Rothe, Alexander Mehler, Nicolas Néel, and Jörg Kröger — TU Ilmenau, Institut für Physik

Rubrene ($C_{42}H_{28}$) was adsorbed with submonolayer coverage on Pt(111), Au(111) and graphene-covered Pt(111). Adsorption phases and vibronic properties of $C_{42}H_{28}$ consistently reflect the progressive reduction of the molecule–substrate hybridization. Separate $C_{42}H_{28}$ clusters are observed on Pt(111) as well as broad molecular resonances. On Au(111) and graphene-covered Pt(111) compact molecular islands with similar unit cells of the superstructure characterize the adsorption phase. The highest occupied molecular orbital of $C_{42}H_{28}$ on Au(111) exhibits weak vibronic progression while unoccupied molecular resonances appear with a broad line shape. In contrast, vibronic subbands are present for both frontier orbitals of $C_{42}H_{28}$ on graphene. They are due to different molecular vibrational quanta with distinct Huang-Rhys factors.

Financial support by the Deutsche Forschungsgemeinschaft through Grant No. KR 2912/12-1 is acknowledged.

O 18.2 Mon 18:00 P4

The effect of morphology of an intercalated Au layer on electronic properties of Graphene — •Amirhossein Bayani and Karin Larsson — Department of Chemistry-Ångström laboratory, Uppsala University, Uppsala, Sweden

Thermal Intercalation processes have recently made it possible to produce large quasi free-standing graphene layers on different substrates. One method, which is based on thermal annealing, uses a 4H-SiC (0001) substrate with an attached carbon buffer layer onto the Si-face of the substrate. Various types of metals have then been used with the purpose to intercalate these metal atoms between SiC and the buffer layers, thereby creating a monolayer (ML) of graphene with an intact Dirac point. Moreover, when positioning heavy metal atoms under graphene, the spin-orbit coupling will increase and thereby enhances the Rashba band splitting. This phenomenon comes from the breakage of a mirror symmetry due to interaction with the substrate. To the knowledge of the authors, the role of the morphology of the intercalated layer on the electronic properties of graphene has not yet been considered and it is worth considering this effect when electronic properties of graphene is wanted to be studied after intercalation. Here we show how the morphology of the intercalated Au layer will affect the electronic properties of a ML of graphene. The possibility to induce a band gap at the Dirac point of graphene by manipulating the staggered potential of a 4H-SiC/Au substrate, is thereby looked for. The calculations are based on density functional theory (DFT) + SOC.

O 18.3 Mon 18:00 P4

Intercalation of epitaxial graphene on SiC(0001) with sulfur — •Susanne Wolff, Niclas Tilgner, Tassilo Rauschendorfer, Florian Speck, and Thomas Seyller — Chemnitz University of Technology, 09126 Chemnitz, Germany

Epitaxial growth of graphene on SiC in argon atmosphere is a well-established method to produce high quality films. There, the first grown carbon layer is partially covalently bound to the substrate and lacks the graphene-like electronically properties. This so-called buffer layer can be decoupled from the substrate by intercalation. Furthermore, the choice of intercalant manipulates the electronic properties of the decoupled graphene.

We investigated the intercalation of a buffer layer with sulfur by X-ray photoelectron spectroscopy (XPS) and angle-resolved photoelectron spectroscopy (ARPES). The intercalation process was performed in a two-zone furnace where the FeS_2 precursor and the buffer layer can be heated separately. An argon gas flow transports the sublimated sulfur to the sample. Partial intercalated samples show two sulfur peaks at different binding energies in XPS. They can be attributed to intercalated sulfur and sulfur on top of the not intercalated buffer layer. ARPES measurement in the vicinity of the Dirac point showed that sulfur intercalation results in p-type doped decoupled graphene layers.

O 18.4 Mon 18:00 P4

twisted graphene on Ir(111) and SiC(0001) studied by SPA- LEED — •mohammad tajik[1], christian brand[1], birk finke[1], karim omambac[1], laurenz kremeyer[1], frank meyer zu heringdorf[1,2], and michael horn-von hoegen[1,2] — [1]Universität Duisburg-Essen — [2]center for nanointegration duisburg-essen

when graphene is placed on a crystalline surface, the periodic structures within the layers superimpose and a moiré superlattices form. Small lattice rotations between the 2D-layer and the substrate strongly modify the moiré superlattice, upon which many electronic, vibrational, and chemical properties depend. Here we report on such structural modification of epitaxial graphene grown on metallic Ir(111) and semiconducting SiC(0001) surfaces. The spontaneous reorientation in the degree- and sub-degree-range of graphene on Ir(111) depends on the substrate temperature during growth. This effect is described by a 2D coincidence network favored by strain reduction together with the dissimilar thermal expansion of the substrate and graphene. For graphene on SiC(0001) only the oriented R0* phase is found due to the higher bonding strength to the substrate. Upon H and Sn intercalation this interaction can be reduced such that the graphene layer is lifted from the substrate. Finally, we present a detailed analysis of an unusually broad diffraction background found for graphene and hex-BN on both substrates.

O 18.5 Mon 18:00 P4

On the Way to Twisted Bilayer Graphene: Formation and Decoupling of 0°-Rotated Epitaxial Graphene — •Hao Yin[1,2], Miriam Raths[1,2], Mark Hutter[1,2], François C. Bocquet[1], and Christian Kumpf[1,2] — [1]Peter Grünberg Institute (PGI-3), Forschungszentrum Jülich and Jülich-Aachen Research Alliance (JARA), Fundamentals of Future Information Technology, Jülich, Germany — [2]Experimentalphysik IV A, RWTH Aachen University, Aachen, Germany

On the way to twisted bilayer graphene with a twisting angle of 30°, we investigated the graphene growth on 6H-SiC(0001) using an unconventional epitaxial growth method named 'surfactant-mediated growth', which is based on annealing the SiC surface in borazine atmosphere. Here, we report a LEEM-based study on two different samples, on which we observed different surface morphologies with varying numbers of stacked graphene layers. A large-scale uniform graphene R0° monolayer is found on the first sample that was annealed to 1330°C. This sample is a promising candidate for producing 30° TBG. Interestingly, some regions of the graphene R0° layer exhibit different brightness contrasts at specific start voltages, without showing significant differences in their LEEM-IV curves. The second sample was annealed 50°C higher, causing the formation of graphene multilayer domains. We utilized LEEM and LEEM-IV in order to determine the number of layers as well as the distribution of multilayer graphene domains.

Furthermore, we discuss the influence of the annealing temperatures in terms of the formation and decoupling of the epitaxial graphene layers.

O 18.6 Mon 18:00 P4

Vertical structure of Sb-intercalated quasi-freestanding graphene on SiC(0001) — You-Ron Lin[1,2,3], Susanne Wolff[4,5], •Mark Hutter[1,2,3], Serguei Soubatch[1,2], Tien-Lin Lee[6], F. Stefan Tautz[1,2,3], Thomas Seyller[4,5], Christian Kumpf[1,2,3], and François C. Bocquet[1,2] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]Jülich Aachen Research Alliance (JARA) — [3]Experimentalphysik IV A, RWTH Aachen University, Germany — [4]Institute of Physics, Faculty of Natural Sciences, TU Chemnitz, Germany — [5]Center for Materials, Architectures and Integration of Nanomembranes (MAIN), TU Chemnitz, Germany — [6]Diamond Light Source Ltd., Didcot, Oxfordshire, UK

Using the normal incidence x-ray standing wave (NIXSW) technique, we have investigated the vertical structure of quasi-freestanding monolayer graphene (QFMLG) obtained by intercalation of antimony under the $(6\sqrt{3}$ x $6\sqrt{3})$ R30° reconstructed graphitized 6H-SiC(0001) surface, also known as zeroth-layer graphene. We found that Sb intercalation decouples the QFMLG very well from the substrate. The distance from the QFMLG to the Sb layer almost equals the expected van der Waals bonding distance of C and Sb. The Sb-intercalation layer itself is mono-atomic, very flat, and located much closer to the substrate, at almost the distance of a covalent Sb-Si bond length. All data is consistent with Sb located on-top of the uppermost Si atoms of the SiC bulk.

O 18.7 Mon 18:00 P4

Bi(110) islands on epitaxial graphene — •Sergii Sologub[1,2], Julian Koch[1], Chitran Ghosal[1], and Christoph Tegenkamp[1] — [1]Institut für Physik, TU Chemnitz, Reichenhainerstr. 70, 09126 Chemnitz — [2]Institute of Physics, NAS of Ukraine, Nauki avenue 46, 03028 Kyiv

The atomic structure and morphology of ultrathin epitaxial Bi islands grown on the graphene/SiC surface was examined by SPA LEED and STM. Bi films with an average height of a few bilayers were deposited at RT and annealed at 410 K for 30 min afterwards. A Volmer-Weber growth mode with a predominance of the Bi(110) orientation was found. LEED patterns show that the Bi(110) structure has three domains rotated by 60°, with each domain having two subdomains rotated by ±2° with the zig-zag direction of Bi parallel to the armchair direction of graphene and four (minority-)subdomains with the zig-zag direction of Bi parallel to the zig-zag direction of graphene. Moreover, STM investigations revealed an elongation of the islands in the zig-zag direction of Bi as well as preferential ("magic") thicknesses (even numbers of Bi monolayers).

Additionally, magneto transport measurements using a 4 T magnet were performed in order to investigate the influence of the Bi islands on the weak localization effekt in graphene.

O 18.8 Mon 18:00 P4

Quantum well states in Bi(110) islands grown on epitaxial graphene — •Chitran Ghosal and Christoph Tegenkamp — Institut für Physik, TU Chemnitz

The semimetal Bi attracts a lot of interest because of its unique electronic properties such as a low carrier concentrations and large carrier mobilities coming along with mesoscopic Fermi wavelength giving rise to robust quantum confinement effects and spin polarized states in thin films [1]. In this work we studied the growth of Bi on n-type doped monolayer graphene (MLG) on SiC(0001) by means of STS and STM. While for low coverages Bi(110) islands are formed Bi(111) structures were found for higher coverages. This allotropic transition occurs at 10 monolayers (3.3nm) and is significantly larger than the critical coverage reported for Bi on Si(111) [2]. In contrast to Bi/MLG, the deposition of Bi on HOPG results in the formation of islands with an odd number of layers. These differences were attributed to different substrate screening effects. Irrespective of the interface, Bi seems to grow rather in a relaxed bilayers fashion, i.e. supporting the formation of black phosphorous structures [2]. Spectroscopy performed on islands of different heights revealed a large gap opening (750 meV) at the center of 4 ML islands. In addition, we found signatures of edge states, which might refer to a non-trivial topology within these QWS-stabilized nanostructures.

[1] T. Hirahara et al. Phys. Rev. B 75, 035422 (2007); [2] T. Nagao et al. Phys. Rev. Lett. 93, 105501 (2004).

O 18.9 Mon 18:00 P4

Identification of electronic structures and atomic configuration of Nitrogen defects on graphene/Pt(111) — •Jeong Ah Seo[1,2], Hyunmin Kang[3,4], Young Jae Song[3,4], Jungseok Chae[1], and Andreas J. Heinrich[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Korea — [2]Department of Physics, Ewha Womans University, Seoul, Korea — [3]Department of Nano Engineering, Sungkyunkwan University (SKKU), Suwon, Korea — [4]SKKU Advanced Institute of Nano Technology (SAINT), Sungkyunkwan University (SKKU), Suwon, Korea

Nitrogen doped graphene is considered as an effective method for modulating the electronic states and properties of graphene. We have prepared nitrogen doped graphene on Pt(111) surface, grown by deposition of pyridine precursor molecules. We figured out there exist several types of defects including graphitic nitrogen and pyridinic nitrogen structure. The imaging of the defects is performed using low temperature scanning tunneling microscope (STM) and atomic force microscope (AFM). The defects preferred to have strong triangular shape features above certain bias voltage. To figure out the features of nitrogen dopant atom, we measured bias dependent STM images, scanning tunneling spectroscopy and AFM imaging. We have observed prominent difference in spectroscopic feature near the fermi energy between defects and pristine graphene. In addition, we also have observed atomic configuration of defect site using AFM. With the aid of STM simulation based on calculated results from the density functional theory, possible candidates of defect types will be provided.

O 18.10 Mon 18:00 P4

Bell-shaped Electron Diffraction Component in 2D Materials — •Hannah Kohler, Frank Meyer zu Heringdorf, and Michael Horn-von Hoegen — Faculty of Physics and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048 Duisburg, Germany

In low energy electron diffraction a bell shaped component (BSC) is observed for graphene on SiC and Ir(111) and for hBN on Ir(111) [APL118 (2021) 241902]. The distinctiveness of this broad diffuse intensity appears to be indicative of a highly ordered 2D layer. Several attempts were made to explain the origin of the BSC, but the mechanism remains at large still unclear. In particular, a classification in which 2D systems a BSC exists is missing. Low energy electron microscopy (LEEM) combines the possibility to investigate the BSC with micro-diffraction (μ-LEED) and *in-situ* imaging. It was found that the BSC can in fact be detected not only on graphene and hBN, but also on *ex-situ* chemical-vapor-deposition-grown MoS_2. In graphene on Ir(111) the full width at half maximum (FWHM) of the BSC decreases with an increase in rotational angle of the graphene. Measurements after Cs intercalation of the graphene show a smaller FWHM compared to the non-intercalated graphene layer.

O 18.11 Mon 18:00 P4

Buffer Layer Characterization of Epitaxial Graphene on Silicon Carbide with Scanning Tunneling Microscopy — •Benno Harling[1], Anna Sinterhauf[1], Peter Richter[2], Philip Schädlich[2], Thomas Seyller[2], and Martin Wenderoth[1] — [1]IV. Physikalisches Institut, Georg-August-Universität Göttingen, Germany — [2]Institut für Physik, Technische Universität Chemnitz, Germany

In this contribution, we present a scanning tunneling microscopy study on the buffer layer of epitaxial graphene on 6H-silicon carbide (SiC) at 8 K and room temperature in UHV. The local configurations and properties of the buffer layer still leave many open questions concerning the interactions between graphene and substrate. Our goal is to disentangle the contributions of the graphene and the underlying buffer layer to the tunneling current on a local scale and to establish an understanding of the responsible mechanisms. We employ multibias imaging to investigate buffer layer contributions to the tunneling current. This measurement mode takes quasi-simultaneous STM images line per line at given bias voltages resulting in a connected stack of topography maps. This allows to observe the changes of the topography in dependency of the applied bias voltage, with much lower dependency on tip modifications while scanning. We connect local corrugation changes to relative heights and the overall corrugation tendency within the stack. Financial support of the Deutsche Forschungsgemeinschaft (DFG) is given by project We 1889/13-1 and We 1889/14-1.

O 18.12 Mon 18:00 P4

Intercalation of Fe under graphene on Ir(110) — •Jason

Bergelt, Affan Safeer, Stefan Kraus, Jeison Fischer, and Thomas Michely — II. Physikalisches Institut, Universität zu Köln, Zülpicher Str. 77, 50937 Köln, Germany

Graphene on Ir(110) is a single-crystal, where graphene is nearly strain free and the resulting moiré pattern consists of waves with crests and troughs along the [001]-Ir direction. Also it has been shown that the growth of graphene prevents the formation of nano-facets that are present in bare Ir(110) at room temperature. In this work, we intercalated several layers of Fe between Gr and unreconstructed Ir(110). By using scanning tunneling microscopy (STM) and low energy electron diffraction (LEED), we confirm that Fe grows pseudomorphic with respect to unreconstructed Ir(110). Due to strong adhesion of graphene to Ir(110), deposition of Fe on a closed graphene layer results in the formation of clusters on the surface and negligible intercalation at temperatures where alloying of Fe with Ir(110) is absent. Therefore, we implemented a procedure to etch with molecular oxygen small holes of controlled size into graphene. Moreover, we investigated the temperature and evaporation flux dependence of intercalation. Furthermore, we discuss the limit of pseudomorphic growth in view of the formation of a reconstruction at thicker intercalated Fe films.

O 18.13 Mon 18:00 P4

Strongly correlated boundary states in topologically insulating chiral graphene nanoribbons tuned by contact potential — •Leonard Edens[1], Francisco Romero[1], Sofia Sanz[2], Amelia Domínguez-Celorrio[5], Manuel Vilas-Varela[3], Jingcheng Li[4], David Serrate[5], Diego Peña[3], and Nacho Pascual[1,2] — [1]CIC nanoGUNE BRTA, San Sebastián, Spain — [2]DIPC, San Sebastián, Spain — [3]CIQUS and Departamento de Química Orgánica, Univ. de Santiago de Compostela, Spain — [4]School of Physics, Sun Yat-sen University, Guangzhou, China — [5]INMA, and Univ. de Zaragoza, 50009 Spain

Chiral graphene nanoribbons present a special class of nanographenes in that their distinct edge symmetry causes a topologically nontrivial band dispersion. Truncating such a ribbon results in a pair of symmetry-protected topological end states predicted to be spin-polarized. Metal substrates interact with the molecular orbitals, transferring charge and screening electron-electron correlations. We decouple the nanoribbons by dragging them onto an ultrathin large bandgap insulator using an STM, and also synthesize them on a low-work function surface. Scanning tunneling spectroscopy on the surface intermetallic GdAu2 reveals a near-neutral ribbon, showing its end state spin-split. On MgO/Ag(001), the ribbons are highly decoupled, leading a greatly enhanced tunneling lifetime, vibronic sidebands, a negative charge and an enhanced correlation gap of opening far in the quantized conduction band.

O 18.14 Mon 18:00 P4

Structural investigation of Pb intercalated graphene on SiC — •Shaista Andleeb, Julian Koch, Chitran Ghosal, and Christoph Tegenkamp — Institut für Physik, TU Chemnitz, Reichenhainerstr. 70, 09126 Chemnitz

In order to modify the electronic properties of graphene, intercalation experiments have received a lot of attention recently. In this research, we examined the intercalation of Pb on carbon buffer layer on SiC(0001) using SPA-LEED and STM. The samples were prepared in UHV by several cycles of Pb deposition using MBE at RT followed by annealing at temperatures ranging from 500 to 900°.

In LEED the intercalated phase shows six characteristic spots around the (00)-spots, which we correlated with a striped phase in STM. The local structure on the stripes shows two honeycomb Pb layers with approximately twice the lattice constant of graphene and turned by ±7.5° with respect to graphene. The corresponding spots in LEED coincide with two buffer layer spots. Their energy dependent behavior was analyzed.

O 18.15 Mon 18:00 P4

Spectroscopic evidence of BCS-BEC crossover in FeSe monolayer — •wantong huang[1,6], haicheng lin[1], gautam rai[2], yuguo yin[1], lianyi he[1], qi-kun xue[1,5], stephan wolfgang haas[2,3], stefan kettemann[3,4], xi chen[1,5], and shuai-hua ji[1,5] — [1]State Key Laboratory of Low-Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing 100084, China — [2]Department of Physics and Astronomy, University of Southern California, Los Angeles, CA 90089-0484, USA — [3]Jacobs University, Campus Ring 1, 28759 Bremen, Germany — [4]Division of Advanced Materials Science,POSTECH, San 31, Hyoja-dong, Nam-gu, Pohang 790-784, South Korea — [5]Frontier Science Center for Quantum Information, Beijing 100084, China — [6]Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany

It has been difficult to realize the crossover from Bardeen-Cooper-Schrieffer (BCS) to Bose-Einstein condensates (BEC) in solids. Here we report direct evidence of BEC in a FeSe monolayer. The Fermi energy of the FeSe film can be tuned by graphene/SiC substrate to realize a BCS-BEC crossover. As the Fermi energy drops, the local density of states measured by STM evolves continuously from a BCS gap to a step-like asymmetric spectrum of BEC state. The theoretical calculation based on a two-band model reproduces well the measured spectra and, in particular, identifies features in the quasi-particle density of states that indicate a transition from the BCS to the BEC regime. In addition, the Zeeman splitting of the quasi-particle states is found to be consistent with the characteristics of a condensate.

O 18.16 Mon 18:00 P4

Investigating the correlated ground state of metallic monolayer MoS_2 with scanning tunneling spectroscopy — •Camiel van Efferen, Jeison Fischer, Thomas Michely, and Wouter Jolie — II. Physikalisches Institut, Universität zu Köln, Germany

Using contactless chemical doping [1], we grow metallic monolayer 2H-MoS_2 on a graphene on Ir(111) substrate. Since this method leaves the surface accessible, we perform real space investigations of correlated behavior in metallic monolayer MoS_2 via scanning tunneling microscopy (STM) and spectroscopy (STS). In STS, we find a broad depression in the density of states around the Fermi level, of about 100 meV. Upon closer inspection, the depression is revealed to consist of a series of small gaps and peaks, spaced by 26-28 meV. This interval corresponds to a flat band in the phonon dispersion of MoS_2. The appearance of the peaks can be explained by strong coupling between the MoS_2 electrons and phonons at the high-symmetry K-point of the Brillouin zone, and may point to the formation of Holstein polarons. The behavior of the polarons is studied near grain boundaries, edges and defects, where the polaron bands are suprisingly seen to bend under the influence of charge.

[1] C. van Efferen et al., 2D Mater. 9 025026 (2022)

O 18.17 Mon 18:00 P4

Artificial electronic lattices on InAs(111)A — •Rian Ligthart and Ingmar Swart — Condensed Matter and Interfaces, Debye Institute for Nanomaterials Science, Utrecht, The Netherlands

Artificial electronic lattices are a promising tool to elucidate novel effects in the quantum world. The Scanning Tunnelling Microscope (STM) allows to build the lattices by manipulating atoms with nanoscale precision and to probe the electronic properties. The electronic lattices consist of artificial atoms on a metallic/semiconducting crystal with a surface state which acts as a 2D electron gas (2DEG). The 2DEG is patterned by scatterers on the surface creating artificial atoms. The versatile artificial atoms are used in lattices to study the effect of structure on electronic characteristics.

An already well-known system is CO on Cu(111), however, this system has a low energy resolution of 80 mV due to coupling of the surface state with bulk states. Here, the promising InAs(111)A surface with native In adatoms is studied since it has a higher energy resolution of around 5 meV. Vertical manipulation allows to place multiple In-adatoms in vicinity of each other. The In adatoms create a potential well that confines the surface state electrons of InAs. The artificial atom created can be coupled into dimers with different bond strengths by varying the distance or by introducing bridge sites. The artificial electronic lattices created on InAs(111)A can help to probe new characteristics in the lattices due to the vastly improved energy resolution.

O 18.18 Mon 18:00 P4

Resonant photoemission studies at the Fe 3*p* threshold on thin FeTe/Bi_2Te_3 and FeSe/Bi_2Se_3 — •Martin Vondráček[1], Tomáš Skála[2], Karel Carva[2], Gunther Springholz[3], and Jan Honolka[1] — [1]Institute of Physics of the Czech Academy of Sciences, Prague, CZ — [2]Charles University, Faculty of Mathematics and Physics, Prague, CZ — [3]Johannes Kepler University, Institute of Semiconductor and Solid State Physics, Linz, AT

Monolayers (MLs) of tetragonal FeTe and FeSe grow on hexagonal Bi_2Se_3(111) and Bi_2Te_3(111) substrates as three 60°-rotated domains. In scanning tunnelling spectroscopy, a gap appears at the Fermi level of FeTe/Bi_2Te_3 suggesting interface-induced superconductivity below

Tc $\approx$ 6 K [1]. FeSe/Bi$_2$Se$_3$ remains in a gapless state [2]. Unconventional superconductivity in Fe-chalcogenides (FeChs) is believed to be correlated to the texture of the Fermi surface at Γ- and M-points. However, ARPES data of FeCh MLs on Bi$_2$Ch$_3$ is complex due to the superposition of contributions from all three domains. Here we show photon energy dependent UPS data probing k_z dispersions along the Γ-Z direction. Fano-like modulations of valence band intensities are observed in the energy range 30-80 eV, indicative of a resonant photoemission effect at the Fe 3p edge around 55 eV. Our ML data is compared to previous bulk studies [3].

[1] A. Eich et al., Phys. Rev. B **94**, 125437 (2016). [2] S. Manna et al., Nat. Commun. **8**, 14074 (2017). [3] T. Yokoya et al., Sci. Technol. Adv. Mater. **13** (2012) 054403.

O 18.19 Mon 18:00 P4

Electron-stimulated photon emission on TMD defects — •Lysander Huberich[1], Jonas Allerbeck[1], Feifei Xiang[1], Riccardo Torsi[2], Anne Marie Tan[3], Pascal Ruffieux[1], Roman Fasel[1], Oliver Gröning[1], Richard Hennig[3], Yu-Chuan Lin[2], Joshua Robinson[2], and Bruno Schuler[1] — [1]Empa - Swiss Federal Laboratories for Materials Science and Technology, Switzerland — [2]Department of Materials Science and Engineering, The Pennsylvania State University, University Park, USA — [3]Department of Materials Science and Engineering, University of Florida, USA

Due to their exceptionally long electron spin coherence and spin-selective optical readout nitrogen-vacancy centers in diamond are considered a key building block in quantum sensing and quantum-cryptography applications. However, defects in bulk materials suffer from limited tunability and placement control, poor photon extraction efficiency, and coherence degradation close to the surface. 2D materials such as monolayer transitional metal dichalcogenides (TMDs) are expected to overcome these challenges while offering new synthetic strategies for the bottom-up design of solid-state defects. Here we present an NV$^-$ center analog in 2D; the dopant vacancy complex (Re$_{Mo}$ + Vac$_S$)$^-$ in MoS$_2$. We investigate its electronic states using scanning tunneling spectroscopy and present atomically-resolved photon emission maps of single TMD defects by means of STM luminescence.

O 18.20 Mon 18:00 P4

Electronic and structural properties of Fe-doped SnS van der Waals crystals — •Damla Yesilpinar[1,5], Martin Vondráček[1], Čestmír Drašar[2], Patrik Čermak[2], Ondřej Caha[3], Karel Carva[4], Václav Holý[4], Jan Prokleška[4], Harry Mönig[5], and Jan Honolka[1] — [1]Institute of Physics, AV ČR, Na Slovance 1999/2 182 21 Praha 8, CZ — [2]Faculty of Chemical Technology, University of Pardubice, Studentská 573, 532 10 Pardubice, CZ — [3]Department of Condensed Matter Physics, Masaryk University, Žerotínovo nám. 617/9, 601 77 Brno, CZ — [4]Department of Condensed Matter Physics, Charles University, Ke Karlovu 5, 121 16 Prague, CZ — [5]Physikalisches Institute, Wilhelm-Klemm Str. 10, 48149 Münster, DE

We investigate the effect of low concentrations of iron on the physical properties of SnS van der Waals crystals grown from the melt. By means of scanning tunneling microscopy (STM) and photoemission spectroscopy we study Fe-induced defects and observe an electron doping effect in the band structure of the native p-type SnS semiconductor. Atomically resolved and bias dependent STM data of characteristic defects are compared to *ab initio* DFT simulations of vacancy (V$_{\mathrm{S}}$ and V$_{\mathrm{Sn}}$), Fe substitutional (Fe$_{\mathrm{Sn}}$) and Fe interstitial (Fe$_{\mathrm{int}}$) defects. In line with our EXAFS data, we propose the importance of Fe$_{\mathrm{int}}$ and discuss possible pairing defects, e.g. with V$_{\mathrm{S}}$.

O 18.21 Mon 18:00 P4

***In Operando* Soft X-Ray Photoemission Spectroscopy of TaS$_2$ and HfS$_2$ based memristive devices** — •Tammo Zimmermann[1], Alena Nierhauve[1,2], Matthias Kalläne[1,2], Jens Buck[1,2], Zhansong Geng[3], Chao Zhang[3], Frank Schwierz[3], Martin Ziegler[3], and Kai Rossnagel[1,2,4] — [1]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität Kiel, 24098 Kiel, Germany — [2]Ruprecht-Haensel-Labor, DESY and CAU Kiel, 22607 Hamburg and 24098 Kiel, Germany — [3]Department of Electrical Engineeering and Information Technology , TU Ilmenau, 98684 Ilmenau, Germany — [4]Deutsches Elektronen-Synchroton DESY, 22607 Hamburg, Germany

In neuromorphic engineering interface-based memristive devices (IMDs) are promising candidates to mimic synaptic behavior. A refined understanding of bias-induced changes in the band structure and the underlying transport mechanisms of layered transition-metal dichalcogenide-based IMDs will improve the development toward future applications, e.g., in artificial neural networks. Here, we show first *in operando* position- and momentum-resolved soft x-ray photoemission spectroscopy measurements obtained from transistor-like TaS$_2$- and HfS$_2$-based IMDs.

O 18.22 Mon 18:00 P4

Comparison of spin-crossover properties between thin film and bulk sample of a binuclear Fe(II) complex — •Marcel Walter[1], Sangeeta Thakur[1], Clara Trommer[2], Felix Tuczek[2], Sebastien Elie Hadjadj[1], Jorge Torres[1], and Wolfgang Kuch[1] — [1]Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin — [2]Institut für Anorganische Chemie, Christian-Albrechts-Universität zu Kiel, Max-Eyth-Straße 2, 24118 Kiel

Spin-crossover molecules (SCMs) are a promising material in the field of spintronics, due to the reversibility of switching between a high-spin and a low-spin state, which can be triggered by light and temperature [1]. The focus of our research is to deposit large SCMs to explore cooperativity in spin switching on surfaces. A pulsed-valve vapor deposition method is used for thin films. This has the advantage of depositing (sub-) monolayers of SCMs in an UHV environment without applying large amounts of thermal energy, which can decompose the large SCMs. We compare the spin-switching behavior of the dinuclear SCM [$\{$Fe(H$_2$B(pz)$_2$)$_2\}_2$(μ-bipy-ac-bipy)] as thin film on highly oriented pyrolytic graphite and as bulk sample using X-ray absorption spectroscopy. The spin-crossover properties were examined for thermal-induced spin state switching as well as for light-induced excited spin-state trapping. The results show that the thin films are locked in a mixed spin state while the bulk sample switches completely, although the equilibrium temperature of the spin states is comparable. [1] L. Kipgen et al., Advanced Materials 33, 24 (2021)

O 18.23 Mon 18:00 P4

Neutral and charged excitations in two-dimensional MoTe$_2$ from first principles — •Franz Fischer[1,2], Abderrezak Bester[1], and Gabriel Bester[1] — [1]University of Hamburg, Institute of Physical Chemistry, 22761 Hamburg, Germany — [2]Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany

Atomically thin layers of transition metal dichalcogenides attract remarkable interest due to their extraordinary electronic and optical properties. The lack of inversion symmetry in their crystal structure combined with strong spin-orbit interaction caused by heavy metal atoms gives rise to an extra valley degree of freedom as well as large spin splittings in the Brillouin zone.

We present an effective first-principle formalism that is extendable to study any order of neutral or charged excitation [1]. Our formalism relies on configuration interaction and the GW-approximation and reduces in the case of excitons to the standard form of the BSE. We will present results of the excited states in monolayer MoTe$_2$ and their emergence from the particular single-particle configurations. In the future we want to extend our methodology to the temporal domain in order to study dynamics of excited states in 2D materials.

[1] Phys. Rev. B. **100** 201403(R) (2019)

O 18.24 Mon 18:00 P4

Relaxation, the moiré potential and excited states for twisted TMDC bilayers — •Carl Emil Mørch Nielsen, Miguel da Cruz, Abderrezak Torche, and Gabriel Bester — University of Hamburg, Institute of Physical Chemistry, 22761 Hamburg, Germany

In recent years, the research of transition metal dichalcogenides has amassed much attention due to interesting properties such as strong localisation of excited states. The field of twistronics emerged as twisting provides a new degree of freedom in engineering specific properties. However, the theoretical ab-initio approach shows an immediate challenge to overcome; large systems, where the moiré unit cell may hold thousands of atoms.

In this project, the aim is to theoretically study moiré confined optical excitations in twisted TMD vdW-homo and heterostructures. We have successfully integrated a force-field based method of relaxation using LAMMPS as suggested in a paper by Jain et. al. We have re-parameterized the SW and KC potentials seen in this paper and expanded the parameter set to include all possible bilayer TMD combinations. With these we can accurately model the associated local band gap variation, e.g. the moiré potential, taking into effect both lattice corrugation and atomic reconstruction. Our goal is now to investigate the excited state properties with our group-developed code from first-principles.

O 18.25 Mon 18:00 P4

Tip-induced excitonic luminescence of an atomically-resolved van der Waals heterostructure — •Luis Parra Lopez[1,2], Anna Roslawska[2], Fabrice Scheurer[2], Stéphane Berciaud[2], and Guillaume Schull[2] — [1]Department of Physical Chemistry,Fritz Haber Institute of the Max Planck Society, 14195 Berlin,Germany. — [2]Université de Strasbourg,CNRS,IPCMS,UMR 7504,67000 Strasbourg,France.

Van der Waals heterostructures (vdWH) made from stacks of transition metal dichalcogenides and other 2D-materials are appealing systems to investigate light-matter interaction.Their optical response is dominated by tightly bound excitons that are sensitive to the presence of atomic-scale inhomogeneities.These inhomogeneities lie at length-scales below the spatial resolution accessible with standard optical spectroscopies.Here, we present an approach using a scanning tunneling microscope to induce the luminescence of an MoSe$_2$/FL-graphene vdWH [1].We correlate the atomic-scale landscape with the locally induced optical response and observe sizeable variations in the excitonic emission between different nm-sized areas.This study paves the way for novel investigations regarding the local properties of vdWH and highly localized excitons.To gain deeper insight into interlayer coupling mechanisms on the nanoscale, which occur on sub-ps timescales, requires additional high temporal resolution as well.I thus conclude with an outlook of an experimental setup capable of addressing ultrafast dynamics with fs-temporal and nm-spatial resolution[2].[1] Parra et al, arXiv:2204.14022(2022).[2] Müller et al, ACS photonics, 7(8),(2020).

O 18.26 Mon 18:00 P4

Changing structural and electronic properties of h-BN on Ir(111) by potassium intercalation — •Philip Grimm, Friedrich Wanierke, Felix Otto, Maximilian Schaal, Roman Forker, and Torsten Fritz — Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany

2D hexagonal boron nitride (h-BN) is applied in electronics components, e.g., in gate dielectrics for transistors. However, its insulating behaviour is strongly influenced by the growth on metal substrates. One possibility to restore the insulating effect and to decouple h-BN is the intercalation of akali metals due to their low ionization energies.

In this study, we use h-BN grown by CVD on Ir(111) while potassium is deposited by thermal evaporation. The samples are investigated at various temperatures, the lateral structure by means of LEED as well as the electronic properties by means of XPS and (AR)PES. A $(\sqrt{3} \times \sqrt{3})$R30° superstructure of K and a decrease of the moiré pattern originating from h-BN/Ir(111) is observed. In addition, the core levels and the band structure shifts to higher binding energies whereas the work function decreases by the similar value. Another observation is a disappearing band folding of the σ-bands. Due to the metastability of the superstructure at room temperature, we switched to low temperature at 35 K and observed even larger shifts of the core levels as well as of the band-structure ($\approx$ 3 eV). In conclusion, our results indicate an intercalation of K in the 2D-system and consequently h-BN is decoupled from Ir(111).

O 18.27 Mon 18:00 P4

Adsorption and Reaction of Bromine on h-BN/Rh(111) — •Eva Marie Freiberger, Natalie J. Waleska, Felix Hemauer, Valentin Schwaab, Hans-Peter Steinrück, and Christian Papp — Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany

Due to their unique chemical and electronic properties, rendering them promising for many applications, numerous two-dimensional materials (2DM) have been predicted, synthesized and characterized. To tailor the properties of 2DM towards possible applications, their chemical modification is of special interest. Hexagonal boron nitride (h-BN), a graphene analogue, exhibits a so-called nanomesh on Rh(111), which can be used as a template enabling spatially defined modification of the 2DM. Using this template, we aim for selective functionalization of h-BN/Rh(111) with halogens, which lead to strong electronic doping. Here, we present a synchrotron radiation-based high-resolution X-ray photoelectron spectroscopy (XPS) study on the adsorption and thermally induced reaction of bromine on h-BN/Rh(111). The adsorption of different amounts of bromine was followed in situ at 170 K, confirming the template effect of the nanomesh for low coverages. Based on temperature-programmed XPS (TPXPS) experiments, we propose covalent functionalization of the pores and a thermally induced on-surface reaction of bromine. Furthermore, the adsorption is observed to be reversible upon heating to 800 K. By shedding light on their controlled chemical modification on the molecular scale, our work paves the way for purposeful tailoring of the properties of 2DM.

O 18.28 Mon 18:00 P4

Structural investigation of antimony monolayers on Ag(111) — •Stefanie Hilgers[1], Julian A. Hochhaus[1,2], Malte G. H. Schulte[1,2], and Carsten Westphal[1,2] — [1]TU Dortmund University, Department of Physics, Otto-Hahn-Str. 4a, 44227 Dortmund, Germany — [2]DELTA, TU Dortmund, Maria-Goeppert-Mayer-Str. 2, 44227, Dortmund, Germany

Similar to graphene and further elements of the 4th main group, Group-V elements such as antimony are also predicted to have extraordinary electronic properties. Because of the strong spin-orbit coupling and the resulting topological properties, antimony monolayers are of great interest for future electronic applications. Since the structural configuration of 2D-materials has major influence on the electronic properties of the material, structural investigations are highly relevant. In the here presented research we report on the synthesis and structural investigation of antimoy monolayers on Ag(111).

After several cleaning cycles in UHV, antimony is evaporated by a Kudsen cell to deposit monolayers on the Ag(111) surface. The well-known $(\sqrt{3}\times\sqrt{3})$R30° superstructure can be identified by low-energy electron diffraction (LEED). In addition, a $(2\sqrt{3}\times2\sqrt{3})$R30° superstructure can be verified for higher coverages. Furthermore, the real space structure is investigated by scanning tunneling microscopy (STM).

O 19: Poster Monday: Scanning Probe Techniques 1

Time: Monday 18:00–20:00 Location: P4

O 19.1 Mon 18:00 P4

Quantifying Force and Energy in Single-Molecule Metalation — Karl Rothe[1], •Nicolas Néel[1], Marie-Laure Bocquet[2], and Jörg Kröger[1] — [1]Institut für Physik, Technische Universität Ilmenau, D-98693 Ilmenau, Germany — [2]PASTEUR, Département de Chimie, École Normale Supérieure, PSL University, Sorbonne Université, CNRS, 75005 Paris, France

An atomic force microscope is used to determine the attractive interaction at the verge of adding a Ag atom from the probe to a single free-base phthalocyanine molecule adsorbed on Ag(111). The experimentally extracted energy for the spontaneous atom transfer can be compared to the energy profile determined by density functional theory using the nudged-elastic-band method at a defined probe-sample distance.

O 19.2 Mon 18:00 P4

Live demodulation of pseudo-heterodyne SNOM at kHz repetition rates — •Philipp Schwendke[1], Samuel Palato[1], Nicolai Grosse[2], and Julia Stähler[1] — [1]PC dept., FHI of the MPG and HU Berlin — [2]ELAB, FHI of the MPG

Scanning near-field optical microscopy (SNOM) allows the spectroscopic investigation of functional surfaces with spatial resolution beyond the diffraction limit. For the observation of electron dynamics on a femtosecond and nanometre scale, common setups rely on high repetition rate laser systems in order to avoid the Nyquist limit, imposed by signal modulation through the tapping AFM tip. Phase-domain sampling schemes allow sample rates below the Nyquist frequency, permitting the use of kHz-class optical amplifiers for tunable wavelengths down to the UV and femtosecond time resolution. We introduce quadrature demodulation for a robust and fast signal demodulation, while also simplifying the experimental setup.

Here, we illustrate and evaluate different methods for signal modulation and data analysis, with the aim of increasing the signal to noise ratio while maintaining high spatial resolution. The near-field contribution is identified through retraction curves, while noise and overall performance is evaluated on real-space images of a Si test sample, showing a significant signal contrast at high harmonic orders of tapping modulation. The presented methods are straightforward to combine with established optical methods, paving the way towards time-resolved SNOM and nano-spectroscopy.

O 19.3 Mon 18:00 P4

Designing a Scanning Probe Microscope to quantify electron correlations in novel 2D materials — •Nikhil Seeja Sivakumar, Henning von Allwörden, Daniel Wegner, Alexander Ako Khajetoorians, and Nadine Hauptmann — Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

Quantum phases in a single or few layers of van der Waal materials often exhibit novel types of charge and spin orders that are driven by electron correlations. To understand the role of electron-electron interactions, it is required to quantify the interplay between the geometric structure, charges and spins at the atomic scale. Scanning tunneling microscopy (STM) and its accompanying magnetic mode, spin polarized STM, are powerful techniques to study the geometric, electronic and magnetic structure, but their application is limited to metallic or semiconducting quantum phases. Here, we present the design and setup of a home-built scanning probe microscopy setup working at 1K based on a JT-stage with 4He, operating in a 3T out-of-plane magnetic field. The setup will combine STM with non-contact Atomic Force Microscopy, Kelvin Probe Force Microscopy, and Magnetic Exchange Force Microscopy to independently study the geometric, electronic and magnetic structure in insulating quantum phases of 2D materials, as well as at phase transitions to conducting phases. A gating stage will allow to study 2D materials in a device geometry.

O 19.4 Mon 18:00 P4

Nanoscale capacitance and dielectric permittivity measurements — •Pascal Rohrbeck[1], Lukas Drago Cavar[1,2], Peter Reichel[2], and Stefan A. L. Weber[1,2] — [1]Max Planck Institute for polymer research, Department physics at interfaces, Ackermannweg 10, 55128 Mainz, Germany — [2]Johannes Gutenberg University, Department of Physics, Staudingerweg 10, 55128 Mainz, Germany

The knowledge of capacitance and dielectric properties in the nanoscale is important to understand the basic physics of semiconductor materials, such as solar cells or battery materials.[1]

In this work, we demonstrate quantitative capacitance and dielectric constant measurements of individual nanostructures using an Atomic Force Microscope (AFM). The new Heterodyne Scanning Capacitance Microscopy (H-SCM) method is based on frequency mixing of two alternating current (AC)-voltages with frequencies in the MHz range. This new method enables quantitative measurements of tip-sample capacitance and local dielectric permittivity with the lateral resolution of the AFM. We can show that the H-SCM reduces the effect of stray capacitance and thereby yields superior lateral resolution.

References:

[1] Fumagalli, L.; Ferrari, G.; Sampietro, M.; Gomila, G. Applied Physics Letters 2007, 91(24), 236243110. doi:10.1063/1.2821119

O 19.5 Mon 18:00 P4

Sphere Probes for Scanning Thermal Microscopy — •Sophie Rodehutskors, Fridolin Geesmann, Philipp Thurau, and Achim Kittel — Insitut für Physik, Carl-von-Ossietzky Universität Oldenburg

Scanning thermal microscopes based on STM and AFM have been used for years to observe near-field mediated heat transfer. Using custom-built coaxial thermocouple tips in an STM setup, spatially highly resolved heat transfer measurements are possible [1]. The total heat transfer between a spherically approximated tip and a sample is expected to depend on the square of the tip's radius [2]. By attaching a 20 μm borosilicate sphere to such a coaxial thermocouple sensor, heat flux sensitivity is further increased in distance-dependence measurements of the heat transfer between a cooled sample and a tip at room temperature for different materials. These sensors can be used for highly resolved radiative heat transfer measurements as well as for heat conduction measurements through self-assembled monolayers of organic molecules.

[1] K. Kloppstech et al., Nat. Commun., 8, 14475 (2017)

[2] E. Rousseau et al., Nat. Photonics 3.9, 514-517 (2009)

O 19.6 Mon 18:00 P4

A New Sensor Concept for Scanning Thermal Microscopy — •Marvin Glittenberg, Philipp Wiesener, and Achim Kittel — Institut für Physik, Carl von Ossietzky Universität Oldenburg

Near-field mediated heat transfer (NFMHT) between two surfaces separated by a vacuum gap has become a widely investigated topic in the past decades. To measure this phenomenon, we are using a self-developed near-field scanning thermal microscope (NSThM) based on an STM. It is equipped with self-made thermocouple tips consisting of a platinum wire molten into a borosilicate glass capillary which is then coated with gold. Since the gold layer can also be used as a tunneling electrode, we were able to measure the thermoelectric voltage and the tunneling current between the tip and sample simultaneously. However, the voltage drop due to the tunneling current interfered with the thermo voltage, making break junction experiments investigating heat and electrical conduction uninterpretable. In a new design of the tips for the NSThM, the thermocouple is covered first by an insulating silicon dioxide film and finally by a second gold film, which then acts as a tunneling electrode separate from the thermocouple and eliminates crosstalk. These tips are called PASA-tips due to the sequence of materials (platinum-gold-silicon dioxide-gold). Some example measurements of radiative heat transfer and heat conduction with these tips are shown to illustrate their capabilities.

O 19.7 Mon 18:00 P4

Implementation of a SPPX Setup and investigation of the delay-time modulation induced intensity modulation — •Marlo Teichmann, Georg Traeger, and Martin Wenderoth — V. Physical Institute, University of Göttingen, 37077, Göttingen, Germany

Shaken-pulse-pair-excited STM (SPPX-STM) successfully combines optical pump-probe techniques with STM to obtain high temporal and spatial resolution [1]. SPPX-STM achieves this by introducing a modulation of the delay time between pump- and probe pulses. The modulation of the delay time instead of the intensity leads to a constant thermal load and hence minimizes thermal effects. However, recent studies have shown that higher harmonics of this modulation causes additional undesirable signals in time resolved measurements [2].

In this study, we implemented a SPPX setup with two semiconductor lasers. These lasers can be triggered electronically which allows for delay times below 1ns between pump- and probe pulse. We implemented two different pump probe schemes to suppress the additional signals, caused by the SPPX-Method. By choosing an improved modulation scheme, we show that it is possible to suppress the contribution of the higher harmonic signals by more than one order of magnitude.

[1] Kloth et al. Rev. Sci. Instrum. 87 (2016)

[2] Takeuchi et al. 2019 Jpn. J. Appl. Phys. 58 SIIA12

O 19.8 Mon 18:00 P4

Grating Coupled Illumination with Image Recognition for Scanning Tunneling Microcopy under Highly Stable and Truly Localized Optical Excitation — •Georg A. Traeger[1], Marlo H. Teichmann[1], Benjamin Schröder[1,2], and Martin Wenderoth[1] — [1]IV. Physikalisches Institut, Georg-August-Universität Göttingen, Germany — [2]Max Planck Institute of Multidisciplinary Sciences, Göttingen, Germany

We present a versatile approach for a highly stable and localized optical excitation in a scanning tunneling microscopy (STM). Optical gratings on STM tips allow to excite a surface plasmon polariton (SPP), which travels to the tunneling junction and results in a highly confined optical excitation. Unfortunately, this technique comes with the drawback, that a much larger thermal load compared to the conventional direct illumination of the tunnel gap is introduced to the tip. This necessitates a high stability of the laser setup both in terms of long-term power stability and pointing[1]. The latter is limited by the mechanical decoupling of the STM from the environment, leading to independent movement of the microscope and the laser focus. We utilize an auto focus approach based on a CCD camera, which is aligned with the optical pathway of a fs-laser to achieve a active coupling between the laser and the STM. Studying ultrafast photo-currents in tunneling junctions we find that the new setup allows for STM operation under higly localized and background free illumination with a lateral resolution better than 2nm. [1] Kloth et al., Rev. Sci. Instrum. 87, (2016).

O 19.9 Mon 18:00 P4

Shot-noise measurements of single-atom Josephson junctions — •Verena Caspari, Idan Tamir, Christian Lotze, and Katharina J. Franke — Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany

Current passing through small constrictions fluctuates due to the discreteness of charge. Measuring this so-called shot noise in atomic-scale superconducting junctions can provide valuable information, from the quanta of charge transferred in multiple Andreev reflections to the correlations between the transmitted electrons in such processes. Here, we use a local shot-noise measurement apparatus, operating at low temperatures, to investigate shot noise in single-atom Pb-Pb junctions. We observe charge doubling inside the superconducting gap and an unexpected deviation from the thermal limit at zero bias voltage.

O 19.10 Mon 18:00 P4

Surface science with haptic feedback — •Maximilian Koall[1], Denis Heitkamp[2], Jaccomo Lorenz[2], Philipp Lensing[2], and Philipp Rahe[1] — [1]Universität Osnabrück, Barbarastraße 7, 49076 Osnabrück, Germany — [2]Hochschule Osnabrück, Albrechtstraße 30, 49076 Osnabrück, Germany

Modern scanning probe microscopy requires intuitive 3D tip positioning and direct access to 3D physical data for interpretation. Still, it is common practice to rely on time-consuming 2D control protocols with only very few approaches involving 3D virtual reality methods [1].

Here, we employ a haptic device for moving the tip at the microscopic scale by hand. An immediate feedback from the measurement signals that reflect the interaction of the tip with the sample is provided, allowing for a quick and intuitive exploration of conductivity landscapes and force fields at surfaces as well as for atom manipulation and tip preparation procedures.

We present the *offline* implementation for recorded data of the well-studied system PTCDA/Ag(111) in a virtual environment (Unity) for improved intuitive understanding of the physical properties. Furthermore, we discuss the approach to interface this system with the scan controller for *online* experiments. The 3D Systems Touch X is used to provide tip control and haptic feedback. The physical observable (tunneling current, frequency shift, damping, force or others) at the tip position in the recorded data is translated into haptic feedback.

[1] Leinen, P.; Green, M. F. B.; Esat, T.; Wagner, C.; Tautz, F. S.; Temirov, R. Beilenstein J. Nanotechnol. 2015, 6, 2148

O 20: Poster Monday: Solid-Liquid Interfaces

Time: Monday 18:00–20:00 Location: P4

O 20.1 Mon 18:00 P4

Pd nanoparticles supported on ordered Co3O4(111): Particle size effects in electrochemical environment — •Maximilian Kastenmeier[1], Xin Deng[1], Tomáš Skála[2], Nataliya Tsud[2], Lukáš Fusek[1,2], Viktor Johánek[2], Josef Mysliveček[2], Yaroslava Lykhach[1], Olaf Brummel[1], and Jörg Libuda[1] — [1]FAU Erlangen-Nürnberg, Erlangen, Germany — [2]Charles University, Prague, Czech Republic

Pd nanoparticles (NPs) are efficient electrocatalysts for oxidation of ethanol in alkaline direct ethanol fuel cells. We investigated the morphology and the oxidation state of Pd NPs supported on well-ordered Co3O4(111) films as a function of the particle size after treatment in ultrahigh vacuum and in alkaline electrolyte under potential control. We combined synchrotron radiation photoelectron spectroscopy and scanning tunneling microscopy. Electronic metal support interaction associated with the charge transfer at the Pd/Co3O4(111) interface yield partially oxidized ultra-small Pdδ+ aggregates and Pd2+ species at Pd coverages below 0.1 ML followed by the growth of two-dimensional metallic Pd0 NPs at higher coverages. The stabilities of ultra-small and conventionally-sized NPs supported on Co3O4(111) and HOPG were compared following an emersion from alkaline electrolyte at potentials between 0.5 and 1.5 VRHE. We observed different oxidation behavior related to two-dimensional and three-dimensional morphologies of supported Pd NPs on Co3O4(111) and HOPG, respectively. In sharp contrast, the oxidation state of the ultra-small Pd deposits remains unchanged between 0.5 and 1.5 VRHE.

O 20.2 Mon 18:00 P4

Film growth and stabilty of the ionic liquid [C$_1$C$_1$Im][Tf$_2$N] on Cu(111) — •Timo Talwar, Stephen Massicot, Florian Maier, and Hans-Peter Steinrück — Lehrstuhl für Physikalische Chemie 2, Friedrich-Alexander-Universität Erlangen-Nürnberg, Egerlandstr. 3, 91058 Erlangen

Ionic liquids (ILs) are salts with melting points below 100°C and extremly low vapor pressure. These properties made them promising candidates for various new catalytic concepts like solid catalysis with Ionic Liquid Layer (SCILL). In this context, the interplay of ILs with metal surfaces is highly important.

In this study, the growth and thermal behavior of ultrathin films of 1,3-dimethylimidazolium bis[(trifluoromethyl)sulfonyl]imide [C$_1$C$_1$Im][Tf$_2$N] on Cu(111) are investigated under UHV conditions. The films are prepared in vacuum by physical vapor deposition and measured by angle-resolved and temperature-programmed X-ray photoelectron spectroscopy. Different film thicknesses are investigated to identify the underlying growth model, which is 2D growth up to 0.3 ML and 3D growth for higher coverages. Note that 1 ML corresponds to a bilayer of cations and anions irrespective of their arrangement. These films are stable up to 300 K. However above 300 K, the anion partly decomposes, and above 500 K, the remaining IL desorbs leaving a decomposed residual on the surface.

Supported through an ERC Advanced Investigator Grant (ILID 693398) to HPS and by the DFG (SFB 1452 CLINT).

O 20.3 Mon 18:00 P4

Influence of water in the electrolyte on the electrochromic characteristics of WO_3 — •Sophie Göbel, Thi Hai Quyen Nguyen, and Derck Schlettwein — Justus-Liebig-Universität Gießen, Institut für Angewandte Physik

In view of global warming, the reduction of CO_2 emissions in the energy sector, e.g. in private households is urgent. In this case, especially heat flow through windows has a main impact. With the help of so-called smart windows, the irradiation of sunlight can be controlled to decrease the energy consumption by air conditioning or heating systems. A possible approach to such switching is provided by electrochromic layers. One of the best observed materials for such smart windows is tungsten oxide WO_3. In this work, porous WO_3 films were prepared via a sol-gel process and spin-coating on fluorine-doped tin oxide (FTO). In order to achieve the porous structure, polymers ($PEG400$ and $PIB_{50} - b - PEO_{45}$) were added as additives in the precursor solution. To enhance the electrochromic characteristics of the WO_3 films in contact to an electrolyte consisting of $LiClO_4$ in propylene carbonate, addition of water into the electrolyte was studied. The amount of added water was varied while the influence on the electrochemical and spectral properties was monitored by cyclic voltammetry and UV/Vis spectroscopy.

O 20.4 Mon 18:00 P4

In situ surface X-ray diffraction studies of Pt(110) — •Jan Ole Fehrs[1], Timo Fuchs[1], Jakub Drnec[2], Marta Mirolo[2], Serhiy Cherevko[3], Valentin Briega[3], David Harrington[4], Chentian Yuan[4], and Olaf Magnussen[1] — [1]Christian-Albrechts Universität zu Kiel, Germany — [2]European Synchrotron Radiation Facility, Grenoble, France — [3]Helmholtz Institute Erlangen-Nürnberg for Renewable Energy, Germany — [4]University of Victoria, Canada

The surface oxidation of platinum is an important process in the degradation of platinum electrocatalysts in PEM fuel cells. Previously, the oxidation of the (111) and (100) surfaces of platinum was investigated by Fuchs et al. to understand the underlying mechanisms and growth of the oxide structures. In this work the (110) surface was studied by similar in situ high energy surface X-ray diffraction (HESXRD) at the ID31 beamline of the European Synchrotron Radiation Facility. Unreconstructed as well as (1x2)-reconstructed Pt(110) surfaces were prepared and their restructuring upon oxidation was examined. Distinct differences in this restructuring of the unreconstructed and the (1x2)-reconstructed surface were found. The changes in occupancy of the topmost surface layers after one oxidation/reduction cycle are greater for the unreconstructed surface than for the reconstructed surface, suggesting that the latter is more stable upon oxidation. In accordance with this result, it was also found that the missing row reconstruction of the surface was only lifted during the reduction of the surface oxide.

O 20.5 Mon 18:00 P4

Increase of the Spectroelectrochemical Performance of WO_3 Films by Using Additives During Film Growth — •Thi Hai Quyen Nguyen[1], Florian Eberheim[1], Sophie Göbel[1], Pascal Cop[2], Marius Eckert[1], Tim P. Schneider[1], Lukas Gümbel[1], Bernd M. Smarsly[2], and Derck Schlettwein[1] — [1]Justus-Liebig-Universität Gießen, Institut für Angewandte Physik — [2]Justus-Liebig-Universität Gießen, Physikalisch-Chemisches Institut

Tungsten oxide (WO_3) is commonly used as an electrochromic material in smart windows. The electrochromic performance of WO_3 is highly influenced by the mixed electronic and ionic transport in the film and, thus, by the accessibility of the internal film volume. In this work, WO_3 thin films were prepared by spin-coating from a precursor solution based on peroxotungstic acid and different polymer additives such as polyethylene glycol (PEG), a block copolymer ($PIB_{50} - b - PEO_{45}$) and a combination thereof. The influence of the additives on the porosity and composition of WO_3 was studied by, e.g., scanning electron microscopy, X-ray photoelectron spectroscopy and atomic emission spectroscopy. Electrochromic characteristics of the films were measured with $LiClO_4$ in propylene carbonate as electrolyte. The intercalation of Li^+ ions was analyzed by time-of-flight secondary ion mass spectrometry. The use of PEG provided microporous films leading to improved effective diffusion coefficients, transmittance modulations and response times compared to compact WO_3. Further improved characteristics were obtained for films with interconnected mesopores prepared with PEG and $PIB_{50} - b - PEO_{45}$.

O 20.6 Mon 18:00 P4

Visualizing electrochemical interfaces with combined AFM/STM — •Andrea Auer, Bernhard Eder, and Franz Giessibl — Institute of Experimental and Applied Physics, University of Regensburg, Germany

Atomic force microscopy (AFM) that can be simultaneously performed with scanning tunneling microscopy (STM) modes using metallic tips attached to self-sensing quartz cantilevers (qPlus sensors) has advanced the field of surface science by allowing for unprecedented high spatial resolution under ultrahigh vacuum conditions. Applying a qPlus sensor-based AFM/STM setup to electrochemistry could offer new and groundbreaking possibilities to locally image both the 3D layering of the interfacial water and the lateral structure of the electrochemical double layer. In this work, a home-built AFM/STM instrument equipped with a qPlus sensor for operation under precise potential control in an electrochemical liquid cell is presented. Special care is taken in the preparation of etched Pt/Ir tips, which are attached to the qPlus sensor and subsequently coated with insulating wax to allow for both AFM and STM measurements in the electrolyte with minimal leak currents. Ongoing work includes investigations of the potential-dependent structural interface organization of the electrochemical double layer on both highly oriented pyrolytic graphite (HOPG) and Au(111) electrodes in acidic media by means of (simultaneous) AFM/STM imaging and force spectroscopy.

O 20.7 Mon 18:00 P4

Phase-shifting electron holography in an environmental TEM — •Ulrich Ross[1], Jonas Lindner[2], Tobias Meyer[2], Michael Seibt[1], and Christian Jooss[2] — [1]IV. Physik Georg-August Universität Göttingen — [2]IMP Georg-August Universität Göttingen

Off-axis electron holography is a phase reconstruction technique which enables access to the complex exit-wave of thin samples. Information on the phase and amplitude of the exit wave is useful in order to gain insight on nm-scale electromagnetic fields. In the field of catalyst research electrostatic surface fields are of particular interest, since the potential drop over the surface can be assumed to play a major role in the reaction mechanisms.

Conventional off-axis holography reconstruction operates via the Fourier domain. As a consequence, the accessible range of spatial frequencies is band-limited by the carrier frequency of the hologram. A trade-off is always necessary in order to optimize fringe frequency, visibility, phase-contrast transfer and instrument stability for a certain range of spatial frequencies. In contrast, we demonstrate the implementation of phase-shifting holography at sub-nm resolution combined with an advanced drift correction scheme, and successfully match the results to multislice image simulations as well as conventional defocus series. The method is added to the in-situ capabilities of a third-order aberration-corrected environmental TEM in order to investigate surface effects in catalytic platinum samples under low pressures of oxygen and water.

O 20.8 Mon 18:00 P4

Self-organized structures of peri-arylenes on electrode surfaces — •Kristin Gratzfeld[1], Anna Kny[1], Tomasz Kosmala[2], Radosław Wasielewski[2], Marek Nowicki[2], Klaus Wandelt[1,2], and Moritz Sokolowski[2] — [1]Institut für Physikalische und Theoretische Chemie der Universität Bonn — [2]Institute of Experimental Physics, University of Wroclaw, Poland

We investigated self-ordered structures of 3,4,9,10-perylene-tetracarboxylic acid (PTCA) on the Au(111) electrode interface. In recent electrochemical studies, the protonation/ deprotonation process of PTCA was studied, however no structural data information was reported, yet [1]. To investigate the structures of PTCA at the Au(111) electrode interface we used cyclic voltammetry (CV) and electrochemical STM (EC-STM). For negatively charged surfaces, we observed stripe like phases of PTCA with an edge-on orientation of the molecules that transform into a less dense phase at more positive potential with flat-lying molecules. We suppose that this transition is correlated with a partial deprotonation of the PTCA. Acknowledgement: This work was supported by the DFG through the research training group 2591, the DAAD (Deutscher Akademischer Austauschdienst) program Ostpartnerschaften and the NAWA (National Agency for Academic Exchange) program. We thank H. Baltruschat for helpful discussions and experimental support. [1] J. Am. Chem. Soc., 138, 1493, 2016.

O 20.9 Mon 18:00 P4

DFT study of the interaction of Br and S adsorbates on the Ag(100) surface — •Sönke Buttenschön and Eckhard Pehlke — Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

Adsorbate-adsorbate interactions on metal surfaces affect the adsorption and diffusion on surfaces and, in case of self-diffusion, the growth of surfaces. As a model system, the diffusion of S adatoms on halogen-covered Cu and Ag electrodes at electrochemical interfaces has been studied by O. Magnussen and his group [1]. Prerequisite for a theoretical analysis is an accurate quantitative description of the adsorbate-adsorbate interactions of the species involved. To this end we have carried through density-functional total-energy calculations of the Br-Br and S-Br interaction on Ag(100) using the codes PWscf and PWneb from the Quantum ESPRESSO package [2]. We observe that additional contributions to the interaction energy beyond the dipole-dipole interaction [3] need to be included, which are due to electronic and elastic mechanisms [4].

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O 20.10 Mon 18:00 P4

The optical spectroscopy of InP(100) in contact with hydrochloric acid: A first-priciples study — •Jongmin Kim[1,2], Margot Guidat[1,2], Mario Löw[1], and Matthias M. May[1,2] — [1]Institute of Physical and Theoretical Chemistry, Universität Tübingen, D-72076 Tübingen, Germany — [2]Institute of Theoretical Chemistry, Universität Ulm, Ulm, Germany

The III–V semiconductors, such as indium phosphide (InP), show highest solar energy conversion efficiencies, and are commonly used for a variety of applications, particularly high-performance opto-electronic devices. However, these semiconductors face fundamental challenges since they tend to corrode in aqueous electrolytes [1]. A passivisation layer on their surface is the most efficient way to address this drawback. Studies have demonstrated that a system of the InP(100) in contact with hydrochloric acid exhibits high conversion efficiencies and reasonable stability. In this computational work, we investigate the interaction of InP(100) with hydrochloric acid by means of computational optical spectroscopy. Theoretically derived reflection anisotropy spectroscopy (RAS) is employed for the optical optical spectroscopy. According to our calculations, the RA spectra are significantly changed with probable surface geometries. A comparison of our results with experiment spectra reveals that the fully Cl-covered structure is the most reasonable surface. This verified structure can be used as a starting structure for further investigations of the InP(100)-electrolyte interface.

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O 20.11 Mon 18:00 P4

Imaging and Illumination of Self-Assembled Molecules at Solid and Liquid Interfaces — •Bensu Günay, •Shilpa Panchami Raj, Christophe Nacci, and Leonhard Grill — University of Graz

Nanotechnological approaches for photochemical on-surface synthesis of covalently-bonded nanostructures have received recent attention at solid/air and solid/liquid interfaces. At the solid/liquid interface, there is in principle always a dynamic equilibrium between the molecules adsorbed on the surface and those still dissolved in the supernatant solution. Additionally, on-surface photochemical activation of chemical reactions provides new reaction pathways and enables the formation of long-range ordered covalent structures. The scanning tunneling microscope (STM) is a powerful tool for observing such structures at surfaces. In this study we present STM measurements under ambient pressure and at room temperature at the solid/liquid interface. In a first step for investigating photoactivated polymerisation under such conditions, 2,5-didodecyl-1,4-di-1-propenyl benzene was introduced onto a highly oriented pyrolytic graphite (HOPG) surface. We report on the self-assembled structures as well as attempts for photochemical activation using various wavelengths at room temperature.

O 21: Poster Monday: Topology and Symmetry-Protected Materials

Time: Monday 18:00–20:00 Location: P4

O 21.1 Mon 18:00 P4

Local manifestations of thickness dependent topology and axion edge state in topological magnet $MnBi_2Te_4$ — •Felix Lüpke[1,2], Anh Pham[2], Yi-Fan Zhao[3], Ling-Jie Zhou[3], Wenchang Lu[4,5], Emil Briggs[4], Jerzy Bernholc[4,5], Marek Kolmer[2,6], Wonhee Ko[2], Cui-Zu Chang[3], Panchapakesan Ganesh[2], and An-Ping Li[2] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]Center for Nanophase Materials Sciences, Oak Ridge National Lab, USA — [3]Physics, The Pennsylvania State University, USA — [4]Physics, North Carolina State University, USA — [5]Computational Sciences and Engineering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37916, USA — [6]Ames Laboratory, USA

The interplay of non-trivial band topology and magnetism gives rise to a series of exotic quantum phenomena, such as the emergent quantum anomalous Hall (QAH) effect. Many of these phenomena have local manifestations when the global symmetry is broken. Here, we report local signatures of the thickness dependent topology in intrinsic magnetic topological insulator $MnBi_2Te_4$ (MBT), using scanning tunneling microscopy and spectroscopy on molecular beam epitaxy grown MBT thin films. A thickness-dependent band gap with an oscillatory feature is revealed, which we reproduce with theoretical calculations. At step edges, we observe localized electronic features, in agreement with topological phase transitions across the steps.

O 21.2 Mon 18:00 P4

Electronic Structure of the Weak 3D Topological Insulator Candidate $Bi_{12}Rh_3Ag_6I_9$ — •Johannes Hessdörfer[1,2], Eduardo Carillo-Aravena[2,3], Armando Consiglio[2,4], Michael Ruck[2,3], Domenico Di Sante[5], and Friedrich Reinert[1,2] — [1]Experimentelle Physik VII, Universität Würzburg, Würzburg, Germany — [2]Würzburg-Dresden Cluster of Excellence ct.qmat, Würzburg Dresden, Germany — [3]Anorganische Chemie II, Technische Universität Dresden, Germany — [4]Theoretische Physik I, Universität Würzburg, Würzburg, Germany — [5]University of Bologna, Bologna, Italy

The electronic structure of $Bi_{12}Rh_3Ag_6I_9$, a weak topological insulator (TI) candidate, is investigated by means of angle-resolved photoelectron spectroscopy (ARPES) and density functional theory calculations. The compound consists of alternating layers of a 2D TI in a Kagome configuration, separated by insulating spacer layers. The Kagome net is formed by rhodium centered bismuth cubes, while the spacer consists of silver and iodine. The results are compared to the mother material $Bi_{14}Rh_3I_9$ [1], denoting the first experimentally observed weak TI. In particular, the influence of the silver substitution into the spacer layer and the potential modification of the coupling between the 2D TI layers is discussed.

[1]Rasche et al., Nature Mater, 12, 422-425 (2013)

O 21.3 Mon 18:00 P4

Characterisation of Fe adsorbates and their effect on the local density of states on topological insulators $TlBiSe_2$ and Bi_2Se_3 by means of combined STM/STS and AFM — •Adrian Weindl, Christoph Setescak, Alexander Liebig, and Franz J. Giessibl — University of Regensburg, Germany

Can one tailor the properties of the topological surface state (TSS) of topological insulators (TIs) by magnetic doping of the TI material? Here, we study the effect of magnetic adatoms on TI surfaces by means of combined scanning tunneling microscopy (STM) and atomic force microscopy (AFM). Two archetypical TIs, Bi_2Se_3 and $TlBiSe_2$, are studied, which both have relatively large band gaps with their Dirac points well isolated and far from bulk states. While the surface of Bi_2Se_3 is atomically flat, $TlBiSe_2$ exhibits a peculiar surface termination consisting of half a monolayer of thallium atoms sitting on top of a full selenium layer. Magnetic impurities, in this case single Fe adatoms, and their influence on the local density of states (LDOS) of the two TIs are investigated by means of scanning tunneling spectroscopy. We detect resonances in the LDOS for the Fe adatoms that arise due to the scattering of electrons in the TSS at these impurities. The position and shape of these resonances are a function of the exact adsorption position of the adatoms, which can be determined by means of atomically-resolved AFM measurements.

O 21.4 Mon 18:00 P4

Investigation of the V/TI interface by TEM and ARPES — •Xiao Hou[1], Max Vassen-Carl[2], Mohammed Qahosh[1], Xiankui Wei[3], Peter Schüffelgen[2], Claus Michael Schneider[1], and Lukasz Plucinski[1] — [1]PGI-6, Forschungszentrum Jülich, Germany — [2]PGI-9, Forschungszentrum Jülich, Germany — [3]ERC-2, Forschungszentrum Jülich, Germany

Topological insulators (TIs) can host so-called Majorana zero modes (MZM) when proximitized with superconductors (SCs). Such a TI/SC system is a promising platform for realizing fault-tolerant quantum computers by employing braiding of the Majorana zero modes [1], in which a sharp interface between SC and TI is one of the prerequisites to realize the Majorana mode [2-3]. Here, vanadium(V) - $(Bi_{0.08}Sb_{0.92})_2Te_3$ is chosen as the SC/TI system.

We use advanced transmission electron microscopy (TEM) and angle-resolved photoemission spectroscopy (APRES) to study structural and electronic properties of the V/TI interfaces. High-resolution TEM and high-angle annular dark-field imaging provide details on crystallinity and atomic arrangements associated with various types of structural defects, while the energy-dispersive X-ray spectroscopy reveals the elemental distribution and also the interfacial interdiffusion. The band alignments between TI and V are studied using ARPES on ultrathin V films deposited on vacuum-transferred TI surfaces.

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O 21.5 Mon 18:00 P4

Heterostructure engineering with the van der Waals topological insulator Bi_2Te_3 — M. Dittmar[1,2], •E. Mantel[1,2], P. Kagerer[1,2], C. I. Fornari[1,2], H. Bentmann[1,2], and F. Reinert[1,2] — [1]Exp. Physik VII, Uni Würzburg — [2]Würzburg-Dresden Cluster of Excellence ct.qmat

Recently, combining magnetism and topology has emerged as a promising research field spanning from topological insulator (TI)-superconductor interfaces to intrinsic magnetic systems [1]. In these systems exciting new phenomena such as Majorana modes are predicted to emerge. Here we present two approaches to study these

promising structures.
The first focuses on the intrinsically ferromagnetic monolayer of $MnBi_2Te_4$. In order to observe the predicted effects in this compound its Fermi level should be tuned inside the bulk band gap. Our approach to control the Fermi level position is to prepare a single layer of $MnBi_2Te_4$ on top of a p-n-junction of Sb_2Te_3 and Bi_2Te_3 grown by molecular beam epitaxy (MBE). In the second approach we investigate the preparation of thin films of the TI Bi_2Te_3 on superconductor substrates. In order to obtain more information about the crystalline structure and surface orientation, we use different characterization methods such as reflection high-energy electron diffraction, X-ray diffraction and low-energy electron diffraction. To investigate the impact on the electronic structures of these systems, angle resolved and X-ray photoemission specroscopy are employed.
[1] M.M. Otrokov et al., Nature **576** (2019)

O 22: Poster Monday: Surface Structure, Epitaxy, Growth and Tribology

Time: Monday 18:00–20:00 Location: P4

O 22.1 Mon 18:00 P4

Ordering processes and phase transitions in amorphous carbon thin films induced by optical excitation — •Carl Arne Thomann[1], Adrian Wittrock[1], Alexandra Wittig[2], Filipe Lopes Dias[2], Dominic Stangier[2], Wolfgang Tillmann[2], and Jörg Debus[1] — [1]Experimentelle Physik 2, TU Dortmund — [2]Lehrstuhl für Werkstofftechnologie, TU Dortmund

Amorphous carbon is a metastable network of short-range ordered carbon atoms often used as protective coatings owing to their exceptional tribological properties. However, high temperatures in a tribological contact may cause considerable changes in the structural ordering, solid-to-solid phase transitions, and degradation of the film. In this work, we present an optical method to initiate and investigate the structural evolution including ordering processes of differently modified films. A pulsed pump laser with micrometer spot size introduces heat into the film, while a second laser probes the Raman scattering signatures. Increasing with laser power, five different stages of structural evolution are found: The first one is coined by thermally resistive amorphous carbon. It is followed by a continuous reduction in the number of lattice defects and non-sixfold aromatic rings. Further increasing pumping power induces a transition from a-C to defected graphite and eventually leads to graphite-dominant defect relaxation and an enhancement in hexagonal lattice areas. Our optical method provides a versatile tool to analyze temperature-induced structural surface changes in a controlled manner, which will improve the understanding about the conditions in tribological contacts.

O 22.2 Mon 18:00 P4

Interactions between bovine calf serum and metallic surface of hipimplant taper junctions — •Adrian Wittrock[1], Saskia Heermant[1], Christian Beckmann[1], Markus A. Wimmer[2], Alfons Fischer[2,3], and Jörg Debus[1] — [1]Experimental Physics 2, TU Dortmund University, Dortmund, Germany — [2]Department of Orthopedic Surgery, Rush University Medical Center, Chicago, USA — [3]Department Microstructure Physics and Alloy Design, Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf, Germany

Within biomedical taper junctions metal surfaces interact with the human body fluid and sustain structural and chemical changes which are of particular interest from the medical and engineering point of view. To simulate the tribological contact of a hip implant taper junction, fretting tests of low-carbon CoCrMo alloys and high-nitrogen FeCrMnMoN steels are performed in bovine calf serum under different numbers of cycles. We investigate structural and chemical variations within the fretting contact by means of label-free and non-destructive Raman spectroscopy. A different absorption behavior of long-chained molecules is observed and the amide I band is shifted from $1655\,\mathrm{cm}^{-1}$ to about $1665\,\mathrm{cm}^{-1}$, which indicates that once a protein is bound to the surface the conformation changes from α-helix to a random or β-sheet structure. A general denaturation of proteins occurs during the fretting experiment. At the heavily worn sample positions lipids are not detected, but sp^2-hybridized amorphous carbon is sometimes measured. Our results contribute to a deeper understanding about structural and chemical properties of biomedical tribological surfaces.

O 22.3 Mon 18:00 P4

Distance dependence of local work function on Pb/Si(111) island — Thomas Späth[1], •Daniel Rothhardt[2], Manuel Schulze[2], and Regina Hoffmann-Vogle[2] — [1]Karlsruhe Institut Technology, D76131 Karlsruhe, Germany — [2]Universtity of Potsdam, Institute of Physics and Astronomy, Experimentelle Physik kondensierter Materie, D14469 Potsdam, Germany

In order to gain a better understanding of diffusion of Lead (Pb) islands on Si(111)-(7x7), it is extremely important to provide a complete description of the electronic properties and the forces acting on the system. Using a Scanning Force Microscope in non-contact mode with Pt coated Si-cantilever allows us to perform point bias-approach measurements at 115 K on Pb islands. We have investigated how the local work function changes as a function of tip-sample distance and how the electrostatic force changes as a function of the applied bias between tip and sample. The resulting force was calculated from the frequency shift distance curves using Baratoff's force inversion method. A significant change in the work function was found when the tip-sample distance was less than 1 nm, which could arise from the overlap of the tip wave functions and the sample wave functions.

O 22.4 Mon 18:00 P4

Epitaxial growth of gold films on elemental superconductors — •Dongfei Wang[1], Katerina Vaxevani[1], Jon Ortuzar[1], Stefano Trivini[1], Danilo Longo[1], Samuel Kerschbaumer[2], Maxim Ilyn[2], Celia Rogero[2], and José Ignacio Pascual[1] — [1]CIC nanoGUNE-BRTA, 20018 Donostia-San Sebastian, Spain — [2]Materials Physics Center (CSIC-UPV/EHU), San Sebastian 20018, Spain

In recent years, superconductivity proximity effect was employed in the designing of topological superconductors [1]. The basic principle in designing a topological superconductor is to introduce superconductivity, spin-orbital coupling and magnetism at the same time. Unfortunately, most of the available elemental superconductors shows less spin-orbital coupling. Gold (Au) is a material famous for its relatively large spin-orbital coupling strength. By placing Au on elemental superconductors, a superconducting system with strong spin-orbital coupling is expected [2]. In our research, Au films with different thickness were grown on elemental superconductors such as V(100) and Nb(110). The films quality were examined by STM as well as XPS technique. By placing magnetic molecules FeTPP on the surface Au/Nb(110), we demonstrate the tunability of magnetic exchange interaction between the molecule and the substrate with Yu-Shiba bound states. Moreover, great Kondo signal enhancements near the pyrrole sites are observed.

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[2] A. Gupta, et al., Physical Review B 69, 104514 (2004)

O 22.5 Mon 18:00 P4

Preparation of highly pure Cu(110) surfaces — •Manuel Seitz, Andreas Christ, Markus Leisegang, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

Ballistic transport of hot charge carriers on the nanoscale becomes more and more important for the development of microelectronic components [1]. The molecular nanoprobe (MONA) technique [2] is a novel method to investigate ballistic charge transport which has already been applied to detect anisotropic transport on the Pd(110) surface [3]. For a more detailed understanding, a survey of other anisotropic fcc(110) surfaces, e.g. Cu(110), is aspired. To detect characteristics intrinsic to the Cu(110) surface, a highly clean and defect-free area of the surface is needed. However, cleaning (110) surfaces with such high purity has proven to be challenging. In this presentation, we summarize our experiences with the preparation of Cu(110) which are compared to similar experiments on Cu(111). While optimizing the preparation parameters, i.e., sputter and anneal cycles, we found a high mobility of surface atoms at room temperature on the Cu(110) surface. Furthermore, upon sputtering the Cu(111) surface, island formation is observed besides the expected vacancy islands [4].

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[4] T. Michely, et. al., Phys. Rev. B 50.15, 11156 (1994).

O 22.6 Mon 18:00 P4

Nanotribological properties of Nitrogen doping-induced modification graphene in ultrahigh vacuum — •Shuyu Huang[1,2], Antoine Hinaut[1], Yiming Song[1], Sebastian Scherb[1], Gema Gnavarro[1], Thilo Glatzel[1], and Ernst Meyer[1] — [1]Department of Physics, University of Basel, 4056 Basel, Switzerland — [2]Key Laboratory for Design and Manufacture of Micro-Nano Biomedical Instruments, School of Mechanical Engineering, Southeast University, Nanjing 211189, China

Graphene, as typical atomically-thin solid lubricant with potential applications in micro- and nano-electromechanical systems (MEMS/NEMS), has been extensively investigated on its nanotribological properties. In the present work, by using a novel experimental approach, for the first time we directly compare the frictional properties between pristine graphene and modified graphene on a single image, showing that atomic-scale friction can be significantly altered by Nitrogen doping-induced modification. Specifically, C60 nano-flakes are deposited as a mask on graphene/Ir (111) surface by thermal evaporation. The sample is then exposed to a nitrogen radical flux produced by a remote RF plasma source. After thermal annealing, to desorb C60 molecules, both nano-patterned modified graphene and pristine graphene, located below former C60 islands, surface is obtained simultaneously. By the means of high-resolution ultrahigh vacuum atomic force microscopy, the topography of surface with two different regions are characterized and discussed in non-contact mode and friction force variation is measured in contact mode.

O 22.7 Mon 18:00 P4

Investigation of indium fluctuation inside Al0.81In0.19N layers — •Keyan Ji[1], Qianqian Lan[2], Yan Lu[2], Michael Schnedler[1], Philipp Ebert[1], and Rafal E. Dunin-borkowski[2] — [1]Peter Gruenberg Institut, Forschungszentrum Juelich, D-52425 Juelich, Germany — [2]Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, Forschungszentrum Juelich, D-52425 Juelich, Germany

We report on the characterization of Al0.81In0.19N/GaN heterostructures grown by Metalorganic Vapor Phase Epitaxy. The specimen was investigated by Electron Holography, High-angle annular dark-field (HAADF) imaging, Secondary-ion mass spectrometry, and Energy-dispersive X-ray spectroscopy. We reveal with different techniques that the Indium concentration gradually increases from 15% to the nominal value of 19% inside Al0.81In0.19N layers. We conduct quantitative analysis on experimental phase images from off-axis electron holography by comparing them with images calculated from the self-consistent electrostatic simulations. Our results illustrated that to accurately determine the electrostatic potential in semiconductor materials from electron holographic phase images, comprehensive knowledge of surface conditions, chemical compositions, and strains is required.

O 22.8 Mon 18:00 P4

Measuring energy dissipation on Si(111) with Lateral Force Microscopy (LFM) — •Thomas Holzmann, Shinjae Nam, Oliver Gretz, Alfred John Weymouth, and Franz Josef Giessibl — Universität Regensburg, Deutschland

In Lateral Force Microscopy (LFM), a cantilever is oscillated parallel to a sample surface at a set amplitude. The forces acting on such an oscillating cantilever are not necessarily conservative. The energy gain or energy loss can be position dependent, depending on the surface. We observed a strong lateral dependence of dissipation around adatoms of the 7x7-resconstruction of Si(111). We used LFM to measure this energy dissipation and applied different bias voltages between tip and sample, which changed the energy dissipation. Certain mechanisms, that contribute to energy dissipation, such as CO-bending (when using a CO-terminated tip) and chemical bonding should not be sensitive to changes in the bias voltage. Only the electrostatic part of the ineraction should be influenced by a change in the bias voltage. We developed a model to simulate the energy dissipation due to electrostatic forces similar to that proposed in Ondracek et al. Nanotechnology 27, 274005 (2016). In this model, electrons tunnel to a local quantum dot on the tip or sample before diffusing to the bulk. It can predict the area where the dissipation occurs. But surprisingly the dissipation signal changes its sign, depending on which side of the quantum dot the tip is located. Following this observation we looked at possible explanations for this behaviour.

O 23: Poster Monday: Nanostructures 1

Time: Monday 18:00–20:00 Location: P4

O 23.1 Mon 18:00 P4

An Atomic Boltzmann Machine capable of Self-Adaption — Brian Kiraly[1], Elze J. Knol[1], •Werner M.J. van Weerdenburg[1], Hilbert J. Kappen[2], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University Nijmegen, the Netherlands — [2]Donders Institute for Neuroscience, Radboud University Nijmegen, the Netherlands

A grand challenge in creating materials with brain-like functionality is understanding multi-well systems. Such multi-well landscapes are linked to energy-based machine learning models, often based on Ising spins. However, the typical short-ranged exchange coupling of Ising spins in real materials prohibit the connectivity required for multi-well landscapes. Therefore, understanding how to create multi-well systems and link these to attractor network models is vital [1].

We present an atomic Boltzmann machine capable of self-adaption using single Co atoms on Black Phosphorus (BP). Using the concept of orbital memory in Co atoms [2], we design a tunable multi-well energy landscape by patterning atoms with atomic manipulation in a scanning tunneling microscope (STM). By electrically gating the structure with the STM tip, we allow the dynamical exploration of its configurations. Due to the anisotropic BP, we find two different timescales that emulate a fast "neural" and a slow "synaptic" timescale. We demonstrate the self-adaption of the synaptic weights to electrical stimuli and explore frequency-based input signals in new types of orbital memory.

[1] Kolmus et al., New J. Phys. 22 (2020);
[2] Kiraly et al, Nat. Comm. 9 (2018)

O 23.2 Mon 18:00 P4

Pulling individual polar molecular wires off of a surface — •Christophe Nacci and Leonhard Grill — Department of Physical Chemistry, University of Graz, 8010 Graz, Austria

The frictional properties of individual nanostructures are strongly influenced by the reduced size and low dimensionality. Accordingly, challenging experiments are required to measure them. Probing the mechanical response of individual nanostructures on chemically different surfaces opens the possibility to explore how the static and dynamic friction depends on the interplay between structural commensurability and chemical composition. Here, we report the vertical pulling of DAD molecular wires [1] off metal surfaces by non-contact atomic force microscopy (AFM), performed under ultrahigh vacuum and at low temperatures. These are polar wires with alternating donor (D) and acceptor (A) groups incorporated in their chemical structure. The aim is to probe the mechanical response of individual molecular wires by force spectroscopy. To furthermore explore the role of structural commensurability between polymers and surface, the polar wires are grown on a variety of different surfaces, from stepped and corrugated surfaces to ultrathin insulating NaCl films on metal surfaces.

[1] C. Nacci et al., Nature Comm. 6, 7397 (2015)

O 23.3 Mon 18:00 P4

Efficient Sieving Performance with Carbon Nanomembranes (CNMs) — •Yubo Qi, Petr Dementyev, and Armin Gölzhäuser — Bielefeld University, Bielefeld, Germany

Nanoporous membranes are promising candidates in ion transport and molecular separation; however, it is still a great challenge to achieve high permeability and selectivity. Carbon nanomembranes (CNMs) emerge as attractive materials for water purification, energy storage, and gas separation. In this work, we present facile CNMs fabrication from polycyclic aromatic hydrocarbons that are drop-cast onto arbitrary supports, including foils and metalized films. The electron-induced polymerizetion is shown to result in continuous sheets of various thickness, and the material is characterized by some spectroscopic and microscopic techniques. The permeation measurements with freestanding membranes reveal a high degree of porosity, and the water permeance (˜10^{-4} mol m^{-2} s^{-1} Pa^{-1}) is four orders of magnitude higher than helium (˜10^{-8} mol m^{-2} s^{-1} Pa^{-1}) with a membrane thickness of 3.0 nm. The ion transport properties were investigated via bias voltage applied across through CNMs. The nanomembrane showed the ability to sieve monovalent and divalent cations, selectivity of all cations follow the ordering $K^+ > Na^+ > Li^+ > Ca^{2+} > Mg^{2+}$. Functional CNMs fabricated from inexpensive precursors pave the way towards the rational design of 2D membranes for high efficient separation.

O 23.4 Mon 18:00 P4

Structure of NbO Nanocrystals on the Nb(110) Surface — •Samuel Berman[1], Kuanysh Zhussupbekov[1], Ainur Zhussupbekova[1], Brian Walls[1], Killian Walshe[1], Sergey I. Bozhko[1,2], Andrey Ionov[2], David D. O'Regan[1,3], and Igor V. Shvets[1] — [1]School of Physics and Centre for Research on Adaptive Nanostructures and Nanodevices (CRANN), Trinity College Dublin, Dublin 2, Ireland — [2]Chernogolovka — [3]AMBER, the SFI Research Centre for Advanced Materials and BioEngineering Research, Dublin 2, Ireland

The properties of Niobium based devices are greatly influenced by the presence of surface oxygen. The highly stable NbO(111) overlayer on the Nb(110) surface is known to host a distinctive nanocrystal structure, as observed by scanning tunnelling microscopy. However the exact structure of this surface has remained a mystery. In order to understand this surface structure, we carry out density functional theory calculations, along with scanning tunnelling microscopy/spectroscopy and X-ray/ultraviolet photoelectron spectroscopy experiments. We propose a new model, contrary to those previously proposed in the literature. The nanocrystal pattern is ascribed to a subtle reconstruction in the surface layer which locally breaks the hexagonal NbO(111) symmetry, giving rise to modulations in the local density of states. Excellent agreement is found between our model and the observed experimental data. We also investigate the underlying reasons that the surface adopts this unusual structure.

O 23.5 Mon 18:00 P4

Helium Ion Microscopy of insulating Materials using Charge Compensation — •Michael Westphal[1], Natalie Frese[1], Yubo Qi[1], Hiroyuki Takei[2], Petr Dementyev[1], André Beyer[1], and Armin Gölzhäuser[1] — [1]Bielefeld University, Germany — [2]Toyo University, Japan

Surface-sensitive imaging capabilities in nanotechnology have become increasingly important in recent years. While scanning electron microscopes (SEM) have become more powerful, they reach their limits when it comes to electrically insulating samples. The accumulation of charge carriers on the sample surface can lead to severe imaging artifacts which necessitates the application of conductive coatings. Helium ion microscopes (HIM) have the possibility to stabilize electric charges by using an electron flood gun to reveal nanoscopic sample features that would otherwise be covered by a conductive coating. In this contribution, we show the benefits of charge-compensated HIM imaging over conventional SEM imaging using the examples of SARS-CoV-2 virus particles [1], carbon nanomembranes from Aromatic Precursors without Headgroups, metal coated SiO_2- and carbon micro-spheres [2]. [1] N. Frese et al. , Beilstein Journal of Nanotechnology 12 172-179 (2021). [2] M. Wortmann et al., Journal of Analytical and Applied Pyrolysis Volume 161, January 2022, 105404.

O 23.6 Mon 18:00 P4

2D covalent organic frameworks from diboronic acids: the influence of the solvent — •Wenbo Lu, Ethan Malone, Mihaela Enache, and Meike Stöhr — Zernike Institute for Advanced Materials, University of Groningen, Netherlands

Over the past years, two-dimensional (2D) covalent organic frameworks (COFs) have gained substantial interest for their use in nanopatterning, organic electronics, and as a basis for nanoreactors or molecular sieves. In particular, 2D COFs formed by polycondensation of boronic acids on surfaces have already yielded promising results [1, 2].

In this study, 2D COFs were synthesized by 1,4-benzenediboronic acid (BDBA) polycondensation on highly oriented pyrolytic graphite (HOPG) which resulted in the formation of a long-range ordered hexagonal molecular network. For the formation of the 2D COFs, a drop of BDBA solution was deposited onto HOPG, then placed in an autoclave and annealed at certain temperature. The structure and coverage of the 2D COFs was studied by scanning tunneling microscopy and atomic force microscopy under ambient conditions. In order to obtain both high quality as well as extended COFs, the influence of four solvents on the COF formation was studied. Our results provide guidance for obtaining high quality 2D COFs formed by boronic acid derivatives.

[1] S. Spitzer et al., Chemical Communications 53 (2017) 5147.

[2] J. F. Dienstmaier et al., ACS Nano 6 (2012) 7234.

O 23.7 Mon 18:00 P4

On-surface synthesis of narrow porous graphene nanoribbons — •Christoph Dobner[1], Mamun Sarker[2], Adrian Ebert[1], Alexander Sinitskii[2], and Axel Enders[1] — [1]Physikalisches Institut, Universität Bayreuth, Universitätsstraße 30, 95447 Bayreuth, Germany — [2]Department of Chemistry, University of Nebraska-Lincoln, Lincoln, NE68588, USA

Fueled by the technological necessity to induce semiconducting properties in graphenic nanostructures, the search for strategies to manipulate their electronic properties has become an active field of research. Common control parameters that determine the band gap of graphene nanoribbons (GNR) are structural width, edge design and heteroatom doping. It has also been theoretically predicted that periodic vacancies can open up a band gap in to otherwise semimetallic 2D-graphene sheets. It is thus reasonable to expect that porosity could also contribute to the band structure of already semiconducting GNRs. In this work we present a strategy for the on-surface synthesis of porous GNRs using precursor. We will show that modified halogenation sites on the precursor molecules result in GNRs containing a periodic arrangement of vacancies along the ribbons backbones. Tunneling spectroscopy was used to determine the band edge locations and band gap width of such structures, revealing a increased band gap of 1.96 eV. Additional dI/dV mapping revealed that periodic electronic states are located at the edges and inside the backbone, induced by the porosity of the structures. Based on these findings a route towards considerably wider porous graphenic flakes will be discussed.

O 23.8 Mon 18:00 P4

Construction of a regular tessellation via bromine bond on a Ag(100) surface — •Wenchao Zhao, Nan Cao, Biao Yang, and Johannes V Barth — Physics Department E20, Technical University of Munich, D-85748 Garching, (Germany)

Engineering 2D surface tessellations at the molecular level attracted major interest with the development of nanoscience and technology. To this end supramolecular self-assembly exploiting tailored molecular species and programmed intermolecular interaction are widely employed. Halogen bonds are promising for potential application due to the σ-hole anisotropic charge distribution of halogen atoms1,2,3. Herein, we address the behavior of ditopic bromine-terminated linear monomers on a Ag(100) surface at the molecular level, affording a tessellation guided by the surface symmetry. In the temperature range of 120 to 200 K, the building blocks form a porous nested grid structure stabilized by bromine bonds and molecule-substrate interactions. Two kinds of quad nodes are distinguished therein, involving a complex non-covalent bonding scheme. Interestingly, both nodes feature supramolecular chirality, entailing domains with both rectangular and trapezoid cavities. Our work introduces a new and complex surface tessellation scheme based on halogenated hydrocarbons on metal surfaces. References 1. G. Cavallo et al., Chem. Rev. 116, 2478-2601 (2016). 2. Z. Han et al., Science 358, 206-210 (2017) 3. Mallada et al., Science 374, 863-867 (2021)

O 23.9 Mon 18:00 P4

Graphitic nitrogen substitution in graphene nanoribbons — •Nicolò Bassi[1], Feifei Xiang[1], Pascal Ruffieux[1], Akimitsu Narita[2], Klaus Müllen[2], and Roman Fasel[1] — [1]Empa-Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland — [2]Max Planck Institute for Polymer Research, 55124 Mainz, Germany

Graphene nanoribbons (GNRs), nanometer-wide strips of graphene, have attracted significant attention thanks to their tunable electronic properties originating from quantum confinement. A possible way to control these properties is through carbon substitution with heteroatoms, such as nitrogen. Despite a large number of theoretical studies, there are few experiments where controlled N substitution has been achieved. We report the on-surface synthesis on Au of two different types of nanoribbons containing N atoms from specifically designed heteroaromatic precursors. The chemical structures of the resulting ribbons were characterized using scanning tunneling microscopy (STM) and non-contact atomic force microscopy (nc-AFM). We demonstrate the high quality ribbons growth, which involves a facile C-N bond formation at temperatures below typical C-C reactions. The electronic properties were further investigated by scanning tunneling spectroscopy (STS), which revealed unoccupied states localized on N atoms that are absent in the undoped sections. These results open new perspectives for growing and studying novel types of GNRs with the possibility to fine-tune electronic properties by controlled heteroatom substitution.

O 24: Overview Talk Yang Shao-Horn

Time: Tuesday 9:30–10:15 Location: S054

Invited Talk O 24.1 Tue 9:30 S054

Oxygen Evolution on Rutile Ruthenium and Iridium Dioxides — •Yang Shao-Horn — MIT, Cambridge, MA, USA

Rutile oxides have been studied intensively for water oxidation in acidic solutions. Unfortunately, atomic details of processes occurring at the electrified interface and active sites are not well understood. In this work, we combine in situ surface sensitive techniques, electrochemical mass spectrometry and density functional theory calculations to elucidate oxygen evolution reaction (OER) on ruthenium and iridium dioxides. Using synchrotron-based ambient pressure X-ray photoelectron spectroscopy and in situ surface diffraction on single crystal surfaces coupled with density functional theory calculations, we show that with increasing potential from 0.5 VRHE, adsorbed water on the coordinatively unsaturated sites (CUS) is successively deprotonated. Surface diffraction measurements and computation reveal what steps can be rate-limiting for OER. Such analyses are applied also to CUS sites on other surfaces as well as different IrO2 surfaces, from which fingerprints of surface oxygen atoms are correlated with OER activity. In order to connect learnings from single-crystals with practical catalysts, electrochemical activity measurements were combined with highly sensitive electrochemical mass spectrometry to quantify the amount of evolved oxygen and reveal three Tafel regimes at different overpotentials, which was rationalized by the coverage of the reaction intermediates. In summary, by employing different experimental and theoretical techniques to model surfaces and practical catalysts, we discuss the nature of the active sites catalyzing OER on ruthenium and iridium dioxides.

O 25: Focus Session: Atomic-Scale Characterization of Correlated Ground States in Epitaxial 2D Materials

The rapid expansion of the family of two-dimensional (2D) materials led to the observation of various quantum phases with correlated ground states in the 2D limit, such as unconventional superconductivity, charge density waves, and novel magnetic phases. On the fundamental level, there are various open questions regarding the mechanisms that underlie these correlated ground states, as well as the understanding of the role of the interface and dimensionality. The epitaxial growth of 2D materials on inert substrates under ultrahigh vacuum conditions and respective in situ investigation allows direct and unambiguous comparison between experimental findings and first-principles calculations. Experimentally, scanning tunneling microscopy methods are ideal to explore the electronic structure and magnetic properties of emerging 2D quantum phases with ultimate real-space and energy resolution at ultralow temperatures. Theoretically, the use of density functional theory calculations requires atomically well-defined unit cells to precisely predict the electronic and magnetic properties. Combining these complementary approaches helps to elucidate the role of the substrate, defects, and the coupling between quasiparticles in stacked heterostructures.

Organizers: Nadine Hauptmann (Radboud University, Nijmegen), Jeison Fischer (University of Cologne), and Wouter Jolie (University of Cologne)

Time: Tuesday 10:30–13:00 Location: H2

Topical Talk O 25.1 Tue 10:30 H2

Designer electronic states in van der Waals heterostructures — •Peter Liljeroth — Department of Applied Physics, Aalto University, PO Box 15100, 00076 Aalto, Finland

Van der Waals (vdW) heterostructures provide unique opportunities for engineering exotic quantum states not found in naturally occurring materials. I will highlight this approach by describing our recent results on realizing topological superconductivity [1,2] and artificial heavy fermion systems in vdW heterostructures [3]. We use molecular-beam epitaxy (MBE) and low-temperature scanning tunneling microscopy (STM) for the sample growth and characterization. Topological superconductivity can be realized by combining ferromagnetic $CrBr_3$ on a superconducting $NbSe_2$ substrate [1,2], which brings together the necessary ingredients for topological superconductivity (out of plane ferromagnetism, Rashba-type spin-orbit interactions and s-wave superconductivity). On the other hand, the building blocks of heavy fermion systems - Kondo coupling between a lattice of localized magnetic moments and mobile conduction electrons - can be mimicked in a 1T-TaS_2 / 1H-TaS_2 heterostructure. These examples highlight the versatility of vdW heterostructures in realizing quantum states that are difficult to find and control in naturally occurring materials.

References: [1] S. Kezilebieke et al. Nature 588, 424 (2020). [2] S. Kezilebieke et al. Nano Lett. 22, 328 (2022). [3] V. Vaňo et al. Nature 599, 582 (2021).

Topical Talk O 25.2 Tue 11:00 H2

Magnetic order in a coherent Kondo lattice in the 1T/1H TaSe2 heterostructure — Wen Wan[1], Rishav Harsh[1], Paul Dreher[1], Sandra Sajan[1], Antonella Meninno[2], Ion Errea[2], Fernando de Juan[1], and •Miguel Ugeda[1] — [1]Donostia International Physics Center (DIPC), Paseo Manuel de Lardizábal 4, 20018 San Sebastián, Spain — [2]Centro de Física de Materiales (CSIC-UPV-EHU), Paseo Manuel de Lardizábal 5, 20018 San Sebastián, Spain

Kondo lattice systems are of fundamental importance for our understanding of quantum criticality and unconventional superconductivity. At the heart of their complexity lies the competition between the opposing forces of Kondo screening and magnetic interactions, which is revealed at very low temperatures as the moments start behaving coherently and eventually determines the fate of the ground state. While our understanding of Kondo lattices has traditionally relied on technically challenging strongly correlated bulk f-electron systems, new light is being shed on the problem thanks to heterostructures of 2D transition metal dichalcogenides, which realize a tunable Kondo lattice platform in a simple material. Here, we study the 1T/1H-TaSe2 heterostructure with high-resolution Scanning Tunneling Spectroscopy at 300 mK, and show a well resolved splitting of the Kondo peak, which increases monotonically in a non-linear fashion in the presence of an out-of-plane magnetic field. This behavior is unexpected for a fully screened Kondo lattice, and it originates instead from a ground state with residual magnetic order, consistent with a Kondo coupling much below the critical point in the Doniach phase diagram.

O 25.3 Tue 11:30 H2

Topological Surface State in epitaxial zz-GNRs — •M. Gruschwitz[1], T.T.N. Nguyen[1], N. de Vries[2], H. Karakachian[3], J. Aprojanz[1], A.A. Zakharov[4], C. Polley[4], T. Balasubramanian[4], U. Starke[3], C.F.J. Flipse[2], and C. Tegenkamp[1] — [1]Technische Universität Chemnitz, Chemnitz, Germany — [2]Eindhoven University of Technology, Eindhoven, The Netherlands — [3]Max Planck Institute for Solid State Research, Stuttgart, Germany — [4]MAX IV Laboratory and Lund University, Lund, Sweden

Protected and spin-polarized transport channels are the hallmark of topological insulators, coming along with an intrinsic strong spin-orbit coupling. Here we identified such corresponding chiral states in epitaxially grown zigzag graphene nanoribbons (zz-GNRs) on mesa-SiC(0001) templates, albeit with an extremely weak spin-orbit interaction. While the bulk of the monolayer zz-GNR is fully suspended across a SiC facet, the lower edge merges into the SiC(0001) substrate and reveals a surface state at the Fermi energy, which is extended along the edge and splits in energy toward the bulk. All of the spectroscopic details are precisely described within a tight binding model incorporating a Haldane term and strain effects. The concomitant breaking of time-reversal symmetry without the application of external magnetic fields is supported by ballistic transport revealing a conduction of $G = e^2/h$ [1,2]. [1] J. Baringhaus et al., Nature 506, 349 (2014); [2] T.T.N. Nguyen et al. Nano Lett. 21, 2876 (2021)

Topical Talk O 25.4 Tue 11:45 H2

Electron-lattice correlations and charge order in two-dimensional materials — •Tim Wehling — I. Institute of Theoretical Physics, Universität Hamburg, Germany

Electronic correlation phenomena in two-dimensional (2D) materials are often tightly linked to lattice degrees of freedom. Examples include superconductivity, periodic lattice distortions and charge density waves (CDWs), metal-insulator transitions, magnetic, *stripe,* or excitonic order across vastly different systems ranging from transition metal dichalcogenides to cuprate high-temperature superconductors. Here, we will address how to disentangle and eventually control the interplay of lattice and electronic degrees of freedom in 2D materials. We will discuss the concept of electron-phonon fluctuation diagnostics to identify the electronic processes behind phonon anomalies in materials like TaS2 [1] and VS2 [2]. We show how the coupling of low energy electrons and lattice degrees of freedom gives rise to anharmonicities and reveal an ultrastrong non-linear mode-mode coupling in VS2.

[1] J. Berges et al., Phys. Rev. B 101, 155107 (2020).

[2] C. van Efferen et al., Nature Communications 12, 6837 (2021).

O 25.5 Tue 12:15 H2

Superconductivity of ultrathin crystalline Al films — •Werner M.J. van Weerdenburg[1], Anand Kamlapure[1], Niels P.E. van Mullekom[1], Xiaochun Huang[1], Manuel Steinbrecher[1], Peter Krogstrup[2], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University Nijmegen, the Netherlands — [2]Center for Quantum Devices, Niels Bohr Institute, University of Copenhagen, 2100 Copenhagen, Denmark

While bulk Al is one of the best known BCS superconductors (SC), it exhibits strange SC behavior in the thin film limit, e.g. a strong enhancement of the critical temperature T_c [1]. The transition from bulk to thin film can modulate the electronic structure and quasiparticle excitations, and the interface with the substrate becomes relevant [2]. Understanding the role of reduced dimensionality, screening and interface effects is important for SC quantum technology.

Here, we study epitaxially grown crystalline Al films on Si(111) as a function of film thickness. Using scanning tunneling microscopy / spectroscopy with temperatures down to 30 mK, we show a strong increase of both the SC gap size Δ and T_c for coverages between 4 and 35 monolayers. Remarkably, we find that Δ is threefold enhanced compared to the bulk value. We characterize the SC state of the thin films in (vector) magnetic field and find a new vortex structure.

[1] P.W Adams et al., Phys. Rev. B 95, 094520 (2017)

[2] A. Kamlapure et al., arXiv:2109.08498 (2021)

O 25.6 Tue 12:30 H2

Realization of Pb honeycomb stuctures by intercalation of buffer layers on SiC(0001) — •Chitran Ghosal, Markus Gruschwitz, and Christoph Tegenkamp — Institut für Physik, TU Chemnitz

The growth of 2D honeycomb lattices of group IV-elements has attracted a lot of interest during the last years. In contrast to graphene, the 2D analogues of Si and Ge can be stabilized on metals, while stanene and plumbene were recently prepared on compound crystals and magnetic substrates, respectively. Here we report on the intercalation of Pb using buffer layers (BL) on SiC(0001), resulting in the formation of freestanding and charge neutral graphene [1]. Depending on the bias voltage, scanning tunneling microscopy reveals complex Moiré structures, which are consistently explained in consideration of two plumbene lattices. These are rotated each by $\pm 7.5^\circ$ with respect to graphene and, thus, are commensurate with the former $6\sqrt{3} \times 6\sqrt{3}$ symmetry of the BL on SiC(0001). Locally, a (2×2) pattern becomes visible in STM and is expected since the lattice constant of plumbene is twice as large as for graphene. Local spectroscopy (STS) done at 4 K reveals an electronic gap of 30 meV and is in qualitative agreement with the band structure of low-buckeled and charge-neutral plumbene.

[1] M. Gruschwitz et al., Materials 14, 7706 (2021)

O 25.7 Tue 12:45 H2

Sn intercalation of the buffer layer/SiC(0001) interface studied by SPA-LEED and STM — •Zamin Mamiyev, Chitran Ghosal, and Christoph Tegenkamp — Institut für Physik, Technische Universität Chemnitz, Chemnitz, Germany

Growth of graphene (Gr) with tailored properties and the simultaneous formation of an exotic 2D interface can be achieved by intercalation of different species below the buffer layer (BL) on SiC(0001). In this regard, Sn is interesting because of a potential Mott ground state imposed by the triangular lattice of Sn atoms at the interface and protected by the Gr layer. Therefore, we studied Sn-intercalation on BL/SiC(0001) by means of spot profile analysis low energy electron diffraction (SPA-LEED) and STM. Starting with a $6\sqrt{3} \times 6\sqrt{3}$ BL surface, we deposited monolayers of Sn at RT and annealed subsequently to higher temperatures. The formation of quasi-free standing monolayer graphene (QFMLG) is confirmed by an apparent 1×1 periodicity. Further insights for a successful decoupling of QFMLG were gained from inspecting the bell shape broadening of the diffraction spots, which is characteristic of weakly coupled 2D systems on surfaces. By further optimization of the post-intercalation treatment, a Sn-induced $\sqrt{3}\times\sqrt{3}$ reconstruction at the interface was achieved. Moreover, detailed studies showed a simultaneous transformation of $6\sqrt{3} \times 6\sqrt{3}$ periodicity into a $(\sqrt{39} \times \sqrt{39})$R16.1° reconstruction with corresponding domains. Interestingly, the new periodicities formed by the intercalant atoms are triggered by the former symmetry of the BL structure.

O 26: Surface Magnetism

Time: Tuesday 10:30–12:45 Location: H4

O 26.1 Tue 10:30 H4

Investigation of bubble domains in Fe_3GeTe_2 by spin-polarized scanning tunneling microscopy — •Namrata Bansal[1], Hung-Hsiang Yang[1], Philipp Ruessmann[2], Markus Hoffmann[2], Lichuan Zhang[2], Dongwook Go[2], Amir-Abbas Haghighirad[3], Kaushik Sen[3], Matthieu Le Tacon[3], Yuriy Mokrousov[2], and Wulf Wulfhekel[1] — [1]Physikalisches Institut, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany — [2]Peter Gruenberg Institut (PGI-1) and Institute for Advanced Simulation (IAS-1) Forschungszentrum Juelich GmbH, D-52425 Juelich — [3]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany

We investigated the magnetic structure of the van der Waal material, Fe_3GeTe_2 (FGT), using a home-built low temperature (0.7 K) spin-polarized scanning tunneling microscope (SP-STM). Out-of-plane magnetic imaging with chromium-coated tungsten tip reveals bubble domains in FGT. When applying out-of-plane magnetic fields, the domain shape transitioned from elliptical to circular and collapsed at around 0.32 T. We observed an inverse relation between the size of the bubble domains and the magnetic fields. Benefiting from the spatial resolution of the SP-STM, the domain wall widths were determined, which contains information about the exchange stiffness and the anisotropy constant of FGT.

O 26.2 Tue 10:45 H4

Magnetization switching on self-assembled structure of alpha-helix-polyalanine molecules observed by ambient STM — •Nguyen T. N. Ha[1], L. Rasabathina[2], O. Helwig[2], A. Sharma[3], G. Salvan[3], S. Yochelis[4], Y. Paltiel[4], and C. Tegenkamp[1] — [1]Solid Surface Analysis, Technische Universität Chemnitz, Germany — [2]Functional Magnetic Materials, Technische Universität Chemnitz, Germany — [3]Semiconductor Physics, Technische Universität Chemnitz, Germany — [4]Department of Applied Physics, Hebrew University of Jerusalem, Israel

Polyalanine (PA) with an alpha-helix conformation has gathered recently a lot of interest as the propagation of electrons along the helical backbone structure comes along with spin polarization of the transmitted electron. The magnetization switching on self-assembly of PA molecules on Au(111)/Co/Au/Pt/Al2O3 substrate by an external magnetic field resulting the preferentially transmitted electrons depending on molecule's specific handedness and the direction of magnetization. The transmission of the electrons through this structure comes along with an quite high efficient spin polarization. The tip-sample distance variation due to magnetization switching on PA films has shown the jumping states between up and down magnetic direction, while on the bare substrate without PA film the stable tip-sample distance was observed. This observation is further confirmed the spin filter behavior of PA film on a magnetic substrate due to CISS effect at nanoscale.[1]N.T.N.Ha et al, J. Phys. Chem. C,124,10,5734-5739,2020.

O 26.3 Tue 11:00 H4

Inelastic Electron Tunneling through Nanomagnetic Structures — •Daria Medvedeva and Jindrich Kolorenč — Institute of Physics (FZU), Czech Academy of Sciences

Inelastic electron tunneling spectroscopy (IETS) is a well-established technique used for investigation of vibrational spectra and, more recently, also for characterization of spin excitations in nanosystems probed in scanning tunneling microscopes (STM). If a magnetic molecule is attached to the STM tip, the possibilities of the probe are expanded [1], for instance, one can observe how excitations in the molecule are modified by exchange interactions with the magnetic nanosystem adsorbed on a surface. We use an in-house implementation of cotunneling theory [2] to model IETS spectra of an STM tip decorated with nickelocene molecule (having spin 1) probing magnetic atoms with spins 3/2 and 2 subject to easy-axis anisotropy of varied direction. Experimentally, these situations were recently realized in an Fe adatom on Cu(100) [1] and in metal-organic chains, incorporating Co and Cr atoms, placed on Au(111) [3]. We discuss how accurate the theory is in reproducing the measured inelastic spectra, how the spectra depend on the direction of the magnetic anisotropy, and how important the appropriate alignment of the anisotropies of the probing molecule and the other spin is for efficient spin sensing.

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[1] B. Verlhac *et al.*, Science **366**, 623 (2019); [2] F. Delgado and J. Fernández-Rossier, Phys. Rev. B **84**, 045439 (2011); [3] Ch. Wäckerlin *et al.*, to be submitted (2022).

O 26.4 Tue 11:15 H4

Magnetocrystalline anisotropy in two-dimensional $EuAu_2$ and $GdAu_2$: the role of band structure — •Maria Blanco-Rey[1,2], Rodrigo Castrillo-Bodero[3], Khadiza Ali[2,3], Polina Sheverdyaeva[4], Enrique Ortega[1,3,2], Laura Fernandez[3], and Frederik Schiller[3,2] — [1]Universidad del País Vasco UPV/EHU, Spain — [2]Donostia International Physics Center, Spain — [3]Centro de Física de Materiales CSIC-UPV/EHU-MPC, Donostia-San Sebastián, Spain — [4]Instituto di Struttura della Materia ISM-CNR, Trieste, Italy

In rare earth (RE) intermetallic crystals, the magnetocrystalline anisotropy has a single-ion contribution from the f shell, often associated with a large orbital quantum number L, and an itinerant one, due to spin-orbit coupling effects in the band structure. As Eu and Gd are $4f^7$ RE atoms, L in 2D atom-thick $EuAu_2$ and $GdAu_2$ is essentially quenched. Therefore, these systems allow us to isolate the itinerant electron contribution in this 2D compound family. X-ray magnetic circular dichroism shows out-of-plane anisotropy in $EuAu_2$, in contrast to in-plane in $GdAu_2$. By means of angle-resolved photoemission and density-functional theory, we explain these behaviours in terms of the occupation of the spin-orbit-split dispersive RE(d)-Au(s) hybrid bands, which is ultimately dictated by the RE valence state (Eu^{2+} and Gd^{3+}). In terms of energy, the itinerant electron contribution is $\approx 1\,\mathrm{meV}$ per unit cell, which may eventually compete with the single-ion contribution.

O 26.5 Tue 11:30 H4

Magnetic Domain Structures of Gd(0001)/W(110) Films — •Patrick Haertl, Markus Leisegang, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

Rare earth metal films are known to exhibit an extremely rich magnetic behavior. Depending on the sign of the RKKY coupling and details of the film preparation various domain structures have been observed, for example on Dy(0001)/W(110) films [1]. Here we report on experiments on Gadolinium (Gd) films epitaxially grown on W(110). Gd is a ferromagnetic metal with a Curie temperature of 293 K. Its half-filled 4f shell results in an almost spherical charge distribution which –in comparison to other rare earth metals– results in a very low magnetic anisotropy [2]. Our investigation on Gd(0001) films grown on W(110) indeed show a rather rich magnetic structure in spin-polarized STM studies. In agreement with earlier Kerr measurements [2], we find a thickness-dependent spin reorientation transition from in-plane at thin films to out-of-plane for films thicker than around 40 nm. The latter form up and down magnetized stripe domains which are tilted by ±30° with respect to the W[001] direction. With increasing coverage their periodicity increases from (50 ± 10) nm up to (120 ± 40) nm and domain branching is observed. We will discuss the energetics of transition of the magnetic structure.

[1] L. Berbil-Bautista et. al., Phys. Rev. B **76**, 064411 (2007).
[2] A. Berger et. al., Phys. Rev. B **52**, 1078 (1995).
[3] P. Härtl et. al., Phys. Rev. B **105**, 174431 (2022).

O 26.6 Tue 11:45 H4

Tuning the electron spin-polarization via tunneling through image states — Maciej Bazarnik[1,2] and •Anika Schlenhoff[2] — [1]Institute of Physics, Poznan University of Technology, Poland — [2]Department of Physics, University of Hamburg, Germany

Image-potential states (IS) are unoccupied electronic states in front of polarizable surfaces. Towards step edges, IS energy bands bend [1], leading to laterally localized IS at the rim of nanoislands [2]. Our spin-resolved scanning tunneling microscopy (SP-STM) and spectroscopy experiments on nanomagnets reveal a spin-polarization of the IS rim state, causing a spatial modulation of the electron spin-polarization above uniformly magnetized nanoislands. An inversion of the electron spin-polarization is found at specific energies. When the electrons relax from the IS into the surface, a spin-transfer torque (STT) is excerted on the sample [3]. We show that according to the IS-induced inversion

of the electron spin-polarization, the STT also changes its sign. Our findings indicate that the IS in front of the surface serves as a spin-filter that can tailor the spin-polarization of the resonant tunneling current. The existence of the rim state and its impact on the electron spin-polarization is expected at the step edge of any magnetic adlayer-substrate system. Hence, nanostructuring a magnetic surface enables tuning the local spin-polarization and thus tailoring the STT for current-induced magnetization switching.
[1] J.-F. Ge *et al.*, Phys. Rev. B **101**, 035152 (2020)
[2] S. Stepanow *et al.*, Phys. Rev. B **83**, 115101 (2011).
[3] A. Schlenhoff *et al.*, Phys. Rev. Lett. **109**, 097602 (2012).

O 26.7 Tue 12:00 H4

Circular dichroic UV-PEEM measurements of magnetic surfaces: From Magnetic Domain Imaging to Magnetoplasmonics — •Maximilian Paleschke and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg, Halle (Saale), Germany

Over the last decades, the rapid progress in the ultrafast optical manipulation of magnetic materials has opened the development of several new methods for the investigation and control of spin and magnetization dynamics. Here, we investigate magnetic thin films and nanostructures on nanometer-femtosecond scales with a newly-designed experimental setup. We combine state-of-the-art time-resolved photoemission electron microscopy (PEEM) with circular dichroism imaging and normal incidence excitation via a tunable femtosecond fiber laser system.

In this talk, we report on two dichroism imaging techniques, namely magnetic circular dichroism (MCD) and the recently discovered plasmonic dichroism. The first is used to image magnetic in-plane and out-of-plane domains of ferromagnetic surfaces. The second was successfully used to image propagating surface plasmon polaritons (SPPs) on a ferromagnetic material in threshold photoemission for the first time [1]. With this, we show clear edge-induced SPPs with sub-micrometer wavelength and propagation length of about $3.5\,\mu$m on polycrystalline $Ni_{80}Fe_{20}$ microstructures. This finding extends experimental investigation of SPPs to materials with high plasma frequency and large damping.
[1] M. Paleschke *et al.*, New J. Phys. **23**, 093006 (2021)

O 26.8 Tue 12:15 H4

Chirality-induced electron spin polarization in chiral CuO and CoO_x catalyst surfaces — •Paul Valerian Möllers[1], Jimeng Wei[2], Supriya Ghosh[2], Soma Salamon[3], Manfred Bartsch[1], Heiko Wende[3], David Waldeck[2], and Helmut Zacharias[1] — [1]Center for Soft Nanoscience, WWU Münster, Germany — [2]Department of Chemistry, University of Pittsburgh, Pittsburgh, USA — [3]Faculty of Physics and Center for Nanointegration Duisburg-Essen, Universität Duisburg-Essen, Germany

Spin-polarized catalytic surfaces can greatly enhance the selectivity of chemical reactions, e.g., in a photoinduced water splitting process. Here, we confirm that spin-polarized (photo)currents can be obtained from chiral cupric oxide[1] (CuO) and cobalt oxide[2] (CoO_x), and explore the underlying mechanism. Chiral oxide thin films were deposited using a method pioneered by Switzer et al.[3] Photoelectrons were excited with deep-UV laser pulses and their average spin polarization (SP) was measured. For CuO thin films, correlating the SP values with electron energy spectra reveals that the measured SP values can be rationalized assuming an intrinsic SP in the chiral oxide layer and a chirality-induced spin selectivity (CISS)-related spin filtering of the electrons. On chiral CoO_x layers, the SP was found to depend on the Co oxidation state, which allows for reversible switching of the preferred spin orientation. The results support efforts towards a rational design of further spin-selective catalytic oxide materials.
[1] J. Phys. Chem. C **123**, 3024 (2019) [2] J. Phys. Chem. C **124**, 22610 (2020) [3] Chem. Mater. **16**, 4232 (2004)

O 26.9 Tue 12:30 H4

A spin-polarized STM investigation of 3 AL Mn films on W(001) — •Paula M. Weber, Jing Qi, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Julius-Maximilians-Universität Würzburg, Germany

Spin spirals and dead magnetic layers in the antiferromagnetic transition metal Mn on the heavy bcc(001) surface of W have recently attracted considerable interest [1,2]. In this talk, we present a spin-polarized STM investigation of 2-4 atomic layer (AL) thick Mn films on W(001). For 3 AL Mn on W(001) it has been theoretically proposed that this system grows pseudomorphically while exhibiting an antiferromagnetic state [3]. Our topographic STM data confirm that pseudomorphic growth even prevails up to a Mn film thickness of 4 AL. Spin-resolved data were acquired with W tips which had been magnetized on Mn layers by *in-situ* treatment on Mn/W(001). This allowed us to collect topographic and spin-resolved data on the same scanning area. Applying this method, we identify a magnetic zig-zag $2\sqrt{2} \times \sqrt{2}$ structure on 3 AL Mn and a strongly bias-dependent labyrinth overlay structure on 4 AL. Potential spin structures will be discussed.
[1] Ferriani *et al.*, Phys. Rev. Letters **101**, 027201 (2008).
[2] Meyer *et al.*, Phys. Rev. Research **2**, 012075 (2020).
[3] Dennler *et al.*, Phys. Rev. B **72**, 214413 (2005).

O 27: Electron-Driven Processes

Time: Tuesday 10:30–11:45 Location: H6

O 27.1 Tue 10:30 H6

Surprisingly fast adsorbate excitation: CO/Ru(0001) probed with X-ray absorption spectroscopy — •Elias Diesen[1], Hsin-Yi Wang[2], Johannes Voss[1], Alan C. Luntz[1], and Anders Nilsson[2] — [1]SUNCAT Center for Interface Science and Catalysis, SLAC National Accelerator Laboratory, Menlo Park, California, USA — [2]Stockholm University, Sweden

Energy transfer between substrate excitations and adsorbate motion plays an essential role in determining chemical reactivity and selectivity on surfaces. Femtosecond pump-probe experiments, using an X-ray probe pulse from a free electron laser, give unique insights into such processes due to the short pulse duration and species-selective probing. We measure the time evolution of the C K-edge X-ray absorption spectrum from CO/Ru(0001) after excitation by a femtosecond high-intensity optical laser pulse, and use detailed spectrum simulations to distinguish excitations of different modes [1]. We find high excitation of the CO internal stretch and frustrated rotation modes within 200 fs of laser excitation - one order of magnitude faster than theoretical predictions. Consequences for our understanding of ultrafast adsorbate excitation will be discussed.
[1] Diesen et al., Phys. Rev. Lett. 127, 016802 (2021)

O 27.2 Tue 10:45 H6

Are vacancies in field ion microscopy artefacts? A DFT study — •Shyam Katnagallu, Jörg Nuegebauer, and Christoph Freysoldt — Department of computational materials design, Max Planck Institut für Eisenforschung GmbH, Max-Planck- Str. 1, 40237, Düsseldorf, Germany.

Resolving the atomic structure of engineering materials in 3D continues to be an extensive research field. Field ion microscopy under evaporating conditions (3D-FIM) is one of the few techniques capable of delivering such atomic-scale information, allowing to even image vacancies and their interactions with solute atoms in alloys. However, the quantification of the observed vacancies and their origins are still a matter of debate. It was suggested that high electric fields (1-10 V/Å) used in FIM could introduce artefact vacancies. To investigate the possibility of this mechanism, we used density functional theory (DFT) simulations. Stepped Ni surfaces with kinks were modelled in the repeated slab approach with a (971) surface orientation. A field of up to 4 V/Å was introduced on one side of the slab using the generalized dipole correction. Contrary to conventional wisdom, we show that the reaction barrier to form vacancies on the electrified metal surface increases compared to the field-free case. We also find that the electric field can introduce kinetic barriers to a potential "vacancy-killing" mechanism. We compare these findings with field evaporation models proposed in the literature.

O 27.3 Tue 11:00 H6

Modeling electron beam damage in Gold nanoparticles and MoS2 — •Cuauhtemoc Nuñez Valencia[1], Matthew Helmi Leth

Larsen[1], William B. Lomholdt[2], Pei Liu[2], Daniel Kelly[2], Thomas W. Hansen[2], and Jakob Schiøtz[1] — [1]DTU Physics, Technical University of Denmark, Kgs. Lyngby, Denmark — [2]DTU Nanolab, Technical University of Denmark, Kgs. Lyngby, Denmark

Beam damage in High-Resolution Transmission Electron Microscopy (HRTEM) is poorly understood theoretically, and not yet well described phenomenologically. The interaction between the specimen and the electron beam changes significantly depending on the material: semiconductors (like MoS2) and metals (like Gold) have different lifetime excitations and beam damage can be triggered in different mechanism.

In this study, we investigate the beam damage in MoS2 and Gold nanoparticles simulating the macroscopic timescale with a Kinetic Monte Carlo like algorithm, using different approaches: long Molecular Dynamics (MD) simulations for Gold and statistical model using different threshold energy (T_D) values for MoS2 considering the thermal vibrations.

The second approach for modeling the beam damage in MoS2 was through ab Initio Molecular dynamics the calculation of the threshold energies for the different scenarios. Where a Kinetic Monte Carlo simulation was implemented for study the dynamics of the defect production and the beam damage for MoS2.Also, we studied the effect in the T_D when we remove or add electrons to the MoS2.

O 27.4 Tue 11:15 H6

Electron-driven mobility of the hydrogen-bonded ammonia clusters — •prashant srivastava[1], Daniel Miller[2], and Karina Morgenstern[3] — [1]Chair of physical chemsitry-I, Ruhr University Bochum, Germany — [2]Department of Chemistry, Hofstra University, 106 Berliner Hall, Hempstead, New York 11549, United States — [3]Chair of physical chemsitry-I, Ruhr University Bochum, Germany

Electron-driven processes in polar solvents are of great interest to study, e.g., electron solvation in ammonia plays a vital role in ozone layer depletion. We study the impact of electrons on the ammonia clusters adsorbed on a copper surface using a combination of low-temperature scanning tunneling microscopy, femtosecond laser pulses, and ab-initio calculations. Photo-injected electrons from the copper surface lead to diffusion or desorption of the second layer of the clusters. Upward mass transport (UMT) and downward mass transport (DMT) also play an important role in modifying the hydrogen-bonded network of these clusters. Theoretical calculations confirm electron solvation into the second layer. We present a molecular-scale insight into the interactions of photo-injected electrons with the ammonia clusters in the second layer. Our results show that this interaction can modify an ammonia cluster and enhance its mobility.

O 27.5 Tue 11:30 H6

Distribution of Charge and Lattice Defects via Machine Learning. — Viktor Birschitzky[1], Michael Prezzi[1], Marco Corrias[1], Lorenzo Papa[1], Igor Sokolovic[2], Alexander Gorfer[1], Martin Setvin[2,3], Michael Schmid[2], Ulrike Diebold[2], Cesare Franchini[1,4], and •Michele Reticcioli[1] — [1]University of Vienna (Austria) — [2]Institute of Applied Physics, TU Wien (Austria) — [3]Charles University, Prague (Czech Republic) — [4]University of Bologna (Italy)

Lattice defects and localized charge on oxide surfaces impact the properties of the material to a different degree depending on their spatial distribution. However, the high number of possible defect configurations poses practical challenges to first-principles studies. Here, we propose a machine-learning-accelerated approach to explore in the framework of density functional theory the spatial configurations of charge and lattice point defects. We apply this approach to analyze the distribution of surface oxygen vacancies on rutile $TiO_2(110)$. The attractive interaction with small polarons (electrons localized on the Ti atoms) are revealed to weaken the repulsion between oxygen vacancies, favoring particular arrangements of the vacancies. The resulting distribution can be compared with the patterns identified by computer vision algorithms on scanning-probe microscopy images.

O 28: Organic Molecules at Surfaces 3: Theory

Time: Tuesday 10:30–12:45 Location: S051

O 28.1 Tue 10:30 S051

Switchable interfaces based on bistable molecules: tetrachloropyrazine on Pt(111) — •Lukas Hörmann, Andreas Jeindl, and Oliver T. Hofmann — Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria

Organic/inorganic interfaces govern the properties of many organic electronic devices. To imbue devices with additional functionality, it would be useful to make these interface properties reversibly switchable by means of easily accessible external parameters, such as the temperature.

In this work, we realize such a switchable interface with tetrachloropyrazine (TCP) on Pt(111). TCP can either chemisorb or physisorb on the Pt(111) surface, forming a double-well potential with strongly differing adsorption geometries. These allow forming diverse interface structures with notably different work functions and coherent fractions (obtained by X-ray standing wave measurements).

We model this switchable interface using a machine learning algorithm, based on Gaussian process regression. This facilitates structure search for commensurate as well as higher-order commensurate adlayers. We find three different classes of interface structures with varying work functions and coherent fractions and demonstrate that external stimuli, such as temperature and pressure, allow to reversibly shift between these different classes. Based on our insights, we discuss how systems need to be constructed so that the switch between different states leads to an even larger change in their properties.

O 28.2 Tue 10:45 S051

Substrate enhanced Jahn-Teller effect in single molecule junctions — •Moritz Frankerl[1], Laerte Patera[2,3], Thomas Frederiksen[4], Jascha Repp[3], and Andrea Donarini[1] — [1]Institute for Theoretical Physics, University of Regensburg — [2]Catalysis Research Center, Technical University of Munich — [3]Institute of Experimental and Applied Physics, University of Regensburg — [4]Donostia International Physics Center (DIPC), Spain

The stabilization of several charge states of single molecules deposited on non-conductive NaCl films [1] allows to map out the electronic transition between different charge states by means of single-electron alternate-charging scanning tunneling microscopy [2].Copper-phthalocyanine (CuPc) revealed a Jahn-Teller splitting (JTS) of its doubly degenerate LUMOs upon charging and, more surprisingly, the occupation of the same LUMO for both electrons upon double charging. We show by DFT calculations complemented by a group-theoretical analysis how the JTS for CuPc in gas phase is not sufficient to explain this behavior. We propose, instead, a cooperation between molecule and substrate deformation which enhances the strength of the JT coupling. The result is based on a microscopic model of the electron-phonon coupling between the molecule and the underlying thick layer of NaCl. The magnitude of the obtained substrate-induced JTS highlights the impact of the substrate on the electronic configuration of charged molecules, far beyond the mere charge stabilization. [1] Fatayer S. et al., Nature Nanotechnology 13, 376-380(2018) [2] Patera L.L. et al., Nature 566, 245-248(2019)

O 28.3 Tue 11:00 S051

Impact of electron-phonon interaction on metal-organic interface states — •Lukas Eschmann, Jan Neuendorf, and Michael Rohlfing — Institut für Festkörpertheorie, Westfälische Wilhelms-Universität Münster, 48149 Münster, Germany

We discuss electron phonon interaction (EPI) in the context of metal-organic adsorbate systems and the occurring nearly-free electron-like interface states (IS) whose energy E is strongly coupled to the vertical binding distance d with a coupling strength of $\lambda = \Delta E/\Delta d$ that is in the order of $1\,\mathrm{eV/Å}$ for several adsorbate systems.

We present an EPI model that is based on ab-initio data and uses the nearly-free electron-like character of the IS to obtain an analytic expression for the temperature induced renormalization of the latter[1,2]. We find that the energy shift is dominated by the coupling to the out-of-plane phonon modes that change the average binding distance and scales quadratically with the coupling strength, i.e., $\sim \lambda^2$. Applied to the system of a NTCDA layer adsorbed on Ag(111), our model predicts a renormalization of the IS onset energy through quantum mechanical coupling with the adsorbate phonons by -10 meV per 100 K. With this, we are able to explain the discrepancy between a classical lifting-effect and the experiment, thus revealing the important impact of EPI on spectroscopic states at metal-organic internal interfaces.

[1] Eschmann, et al., Phys.Rev.B 104, L241102 (2021).

[2] Eschmann, et al., Phys.Rev.B 104, 245403 (2021).

O 28.4 Tue 11:15 S051

X-ray spectroscopic fingerprints of chemical bonding at molecule-metal interfaces revealed by first-principles core-level simulation — •Samuel J. Hall[1], Benedikt P. Klein[1,2], and Reinhard J. Maurer[1] — [1]Department of Chemistry, University of Warwick, Coventry, UK — [2]Diamond Light Source, Harwell Science and Innovation Campus, Didcot, UK

Characterisation of the chemical environment and electronic structure of organic materials and metal-organic interfaces can be carried out with x-ray photoelectron spectroscopy (XPS) and near-edge x-ray absorption fine structure (NEXAFS) spectroscopy. The adsorption of molecules onto surfaces has been shown to change various spectral features which have previously been connected to changes in chemical bonding and charge distribution. However, these can be difficult to interpret due to overlapping features of multiple species and widely broadened spectra. Through the use of density functional theory (DFT) calculations we study two molecular isomers, azulene and naphthalene, adsorbed onto three metal (111) surfaces of silver, copper, and platinum. Categorised into three regimes of molecule-metal chemical bonding, we connect the changes seen in the spectra to the chemical bonding behaviour and decompose the signatures into initial core-state and final valence-state contributions. We analyse the effect of charge transfer, electronic hybridisation and dispersion effects, and aim to provide guidance for experimental spectral analysis.

O 28.5 Tue 11:30 S051

The sensitivity of NMR chemical shifts to organic/inorganic interfaces — •Viviana Piccinni[1], Emmanouil Veroutis[2], Karsten Reuter[1], Josef Granwehr[2], and Christoph Scheurer[1] — [1]Fritz Haber Institute of the Max Planck Society, Berlin — [2]Forschungszentrum Jülich, IEK-9, Jülich

Composite polymer/ceramic solid electrolytes are promising materials for Li-ion batteries due to their high stability and safety. Yet, their organic/inorganic interface is believed to hinder the ionic conduction. NMR experiments on a poly(ethylene oxide)/$Li_{1.5}Al_{0.5}Ti_{1.5}(PO_4)_3$ electrolyte have shown that Li ions cross the interface on a timescale of seconds. To remedy the problem, mechanistic insights are required, but even the polymer/ceramic interface structure is unknown.

We therefore investigate representative potential binding situations between the two materials by computing the NMR shielding tensors, in order to verify whether there is a significant change in the chemical shifts depending on the interface structure. Due to the high computational cost, a simplified model exhibiting the same local chemical interactions is constructed, composed by an $AlPO_4$ slab and ethane-1,2-diol. NMR measurements on an $AlPO_4$/methanol system have also been carried out.

While the computed NMR properties are sensitive to changes in the interface structure, experiments reveal no chemical shifts variations in the ^1H NMR methanol/$AlPO_4$ spectra relative to neat methanol. This can be ascribed to the presence of surface water in $AlPO_4$, which would leave no available binding sites for methanol/$AlPO_4$ interactions.

O 28.6 Tue 11:45 S051

Classifying Chiral Structure by a Convolutional Neural Network — •Peer Kasten[1], Mandy Stritzke[2], Johannes Tim Seifert[1], Björn Möller[3], Timo de Wolff[2], Tim Fingscheidt[3], and Uta Schlickum[1] — [1]Institut für Angewandte Physik, Technische Universität Braunschweig — [2]Institut für Analysis und Algebra, Technische Universität Braunschweig — [3]Institut für Nachrichtentechnik, Technische Universität Braunschweig

Scanning tunneling microscopy (STM) is an important tool to image surfaces at atomic scale. To examine structures of molecules in STM images can be a difficult and time-consuming task. We present a method to recognize chirality within experimentally observed self-assembled molecular structures using the convolutional neural network (CNN) based object detection framework Faster R-CNN. Thereby we can classify unit cells in the image towards one of both chiral structures.

To train the neural network, a sufficient amount of correctly labeled images is necessary. To obtain such data and labels, we utilize a method to create realistic-looking, synthetic STM images in varying zoom-sizes containing lifelike properties such as noise and step edges along with corresponding labels.

Using this synthetic data, we trained a model capable of classifying synthetic images at sizes ranging from 8 nm to 100 nm with high performance. Evaluations of the CNN's predictions for real images show that this network trained on synthetic data can generalize towards inference on real images.

O 28.7 Tue 12:00 S051

Energy landscaping with external electric fields: Selective stabilization of interface polymorphs — Johannes Cartus, Andreas Jeindl, •Anna Werkovits, and Oliver Hofmann — Graz University of Technology, Graz, Austria

The polymorphs that inorganic/organic interfaces assume can have drastic consequences for the interface properties (such as, e.g., the work function). It is, therefore, of great relevance for applications in organic electronics to achieve precise control of the interface polymorphism. The stability of the respective polymorphs is determined by deposition conditions (e.g., temperature and pressure). In this contribution we investigate electric fields as an additional handle for better control over the polymorphism, thus selectively stabilizing polymorphs at conditions where they were previously inaccessible.

Using density functional theory calculations augmented by a smart-data machine learning approach (SAMPLE) we demonstrate how electric fields can be used to foster desired or even previously unseen polymorphs. Tetracyanoethylene (TCNE) on Cu(111) is an ideal test system because its work function can change by more than 3 eV for different polymorphs.

O 28.8 Tue 12:15 S051

Interlayer orbital overlap governing thin-film geometry: the role of interfacial charge transfer — •Fabio Calcinelli, Andreas Jeindl, Lukas Hörmann, and Oliver Hofmann — Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria

Organic thin films exhibit a large structural variability, and the substrate on which they grow has a relevant influence on their polymorphism. Predicting which structure a thin film will assume on a substrate is impossible through traditional first-principle modeling alone, because of the combinatorial explosion in the number of possible polymorphs. However, recent developments in machine-learning assisted structure search have made structure-to-property investigations accessible. Employing smart-data machine learning, we demonstrate the impact that different substrates can have on the geometry of the first two layers of a thin film. We identify the energetically most favourable geometries for benzoquinone on silver and on graphene, and compare their electronic properties. While the polymorphs formed in the first layer of benzoquinone are very similar, for the second layer we find two significantly different structures. We explain this difference as an effect of the interplay between different charge transfer on the two substrates, and different interlayer orbital overlap for the two structures. Furthermore, we investigate the systematic impact of interlayer orbital overlap in defining the most stable polymorphs for different charge transfers.

O 28.9 Tue 12:30 S051

Polymorph trapping by optimized deposition conditions: A first-principles prediction for TCNE/Cu(111) — •Anna Werkovits, Andreas Jeindl, Lukas Hörmann, Johannes J. Cartus, and Oliver T. Hofmann — Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria

Physical properties of small-molecule organic semiconductors are strongly determined by their polymorphs. Depending on the orientation in which molecules adsorb on the substrate, properties can significantly vary, as is the case, e.g. for tetracyanoethylene (TCNE) on Cu(111).

We propose conditions under which TCNE remains in the energetically less favorable lying position also for higher coverages. This requires fulfilling two prerequisites: To enable ordered growth, the temperature must be high enough to allow for sufficient diffusion of lying molecules, whereas the temperature must be low enough to inhibit the reorientation to the upright-standing position.

In this work, we utilize the nudged elastic band method and density functional theory to compute energy barriers. By means of the harmonic transition state theory temperature-dependent diffusion and reorientation rates are obtained for predicting a temperature range where the kinetic stabilization is attained.

O 29: Metal substrates 1

Time: Tuesday 10:30–12:15 Location: S052

O 29.1 Tue 10:30 S052

Controlling the translation of a single molecule — •Grant J. Simpson[1], Mats Persson[2], and Leonhard Grill[1] — [1]University of Graz, Graz, Austria — [2]University of Liverpool, Liverpool, UK

Controlling both of the orientation and the direction of translation of a single molecule is crucial to the understanding of molecular machines [1,2]. In addition, overcoming microscopic reversibility and realising unidirectional motion is a major criterion if a molecular machine is to do any meaningful work. Here, we report how single molecules can be translated in a directional manner over a metallic surface using voltage pulses from the tip of a scanning tunnelling microscope. Directionality of motion is possible due to translations occurring via an asymmetrically accessible intermediate state. The reaction pathway is discussed as well as how this molecular motion can be coupled to the motion of surface adsorbates.

[1] G. J. Simpson, V. García-López, A. D. Boese, J. Tour, L. Grill, Nat. Commun., 10, 4631 (2019) [2] G. J. Simpson, V. García-López, P. Petermeier, L. Grill, J. Tour, Nat. Nanotechnol., 12, 604 (2017)

O 29.2 Tue 10:45 S052

On-Surface Formation of Cyano-Vinylene Linked Chains by Knoevenagel Condensation — •Kwan Ho Au-Yeung[1], Tim Kühne[1], Dimitry A. Ryndyk[3,4], Gianaurelio Cuniberti[3], Thomas Heine[4], Xinliang Feng[2], and Francesca Moresco[1] — [1]Center for Advancing Electronics Dresden (cfaed), TU Dresden, 01062 Dresden (Germany) — [2]Institute of Molecular Functional Materials, Faculty of Chemistry and Food Chemistry, TU Dresden, 01062 Dresden (Germany) — [3]Institute for Materials Science, TU Dresden, 01062 Dresden (Germany) — [4]Theoretical Chemistry, TU Dresden, 01062 Dresden (Germany)

The incorporation of C=C bonds into conjugated nanostructures with additional functional groups (e.g. nitrogen substitution) has attracted extensive attention in recent years due to their intriguing electronic properties. However, the activation modes inside the toolbox of on-surface synthesis are still limited. We present the novel on-surface formation of CN-substituted phenylene vinylene chains on the Au(111) surface, thermally induced by annealing the substrate stepwise at temperatures between 220°C and 240°C. The reaction is investigated by scanning tunneling microscopy (STM) and density functional theory (DFT). Supported by the calculated reaction pathway, we assign the observed chain formation to a Knoevenagel condensation between an aldehyde and a methylene nitrile substituent.

O 29.3 Tue 11:00 S052

Self-assembly and on-surface Ullmann-type polymerisation of a DPP-based molecular wire on Au(111). — •Michael Clarke[1], Abigail Bellamy-Carter[2], Ferdinando Malagreca[3], David B. Amabilino[4], and Alexander Saywell[1] — [1]School of Physics & Astronomy, University of Nottingham, NG7 2RD — [2]School of Liberal Arts and Natural Sciences, University of Birmingham, Edgbaston, Birmingham, B15 2TT — [3]School of Chemistry, University of Nottingham, NG7 2RD — [4]ICMAB-CSIC, Campus de la UAB 08193 Bellaterra Barcelona, Spain

Diketopyrrolopyrrole (DPP) is a chemical moiety which may act as an electron acceptor within organic electronic-devices and exhibits a high charge carrier mobility. [1] Charge transport within such systems is affected by the local arrangement, and orientation, of molecules/domains; hence controlled formation of ordered structures is of interest. Self-assembly of functionalised DPP species is a route towards ordered structures [2], and on-surface protocols allow monomer units to be covalently coupled under appropriate reaction conditions (e.g. Ullmann-type reactions [3]). Here we study an alkyl chain functionalised DPP unit possessing aryl-hide groups to facilitate on-surface covalent coupling. The self-assembled structure of the monomer units is characterised and ordered polymers, formed via thermal treatment, are investigated using Scanning Tunnelling Microscopy (STM). [1] Y. Li et.al, Energy Environ. Sci., 2013, 6, 1684. Y. Zang et.al, J. Am. Chem. Soc., 2018, 140, 13167. [2] A. Honda et.al , Bull. Chem. Soc. Jpn., 2015, 88, 969.[3] L. Grill & S. Hecht, Nat. Chem., 2020, 12, 115.

O 29.4 Tue 11:15 S052

Quantifying the diffusion of porphyrins on Au(111): A temperature-dependant STM study — •Matthew Edmondson and Alex Saywell — School of Physics & Astronomy, University of Nottingham, Nottingham, UK

The diffusion of surface-confined molecules is a fundamental step within the formation of self-assembled structures and on-surface reactions. Scanning probe microscopies provide a route to characterising the diffusion pathways of these molecules, and allow a quantitative analysis of energetic barriers via Arrhenius-type rate analysis.(1) In particular, SPM allows the relationship between atomic-scale surface structures and molecular diffusion to be explored; a potential method for influencing on-surface reactivity.(2)

The Au(111) surface, frequently used as a substrate for on-surface coupling reactions (3), exhibits the well-known 'herringbone' reconstruction which may influence the diffusion of molecule species. In this work, we report on the diffusion of individual 2H-TPP on the Au(111) surface; characterising the diffusion rate via variable-temperature scanning tunnelling microscopy within specific regions of the reconstructed surface. The energy barrier to diffusion (obtained via Arrhenius analysis) was found to differ between the FCC and HCP regions of the herringbone reconstruction, indicating that local geometric/electronic surface-features play a role in on-surface diffusion.

1. H. Marbach, H.-P. Steinrück, Chem. Commun. 50, 9034 (2014).
2. S. Clair, D. G. de Oteyza, Chem. Rev. 119, 4717 (2019).
3. L. Grill et al., Nat. Nanotechnol. 2, 687 (2007).

O 29.5 Tue 11:30 S052

LT-STM induced reversible chiral switching of thiophene-based molecule on Au(111) — •Suchetana Sarkar[1,2], Kwan Ho Au-Yeung[1,2], Tim Kuehne[1,2], Dmitry A. Ryndyk[2,3], Albrecht Waentig[1,4], Xinliang Feng[1,4], and Francesca Moresco[1,2] — [1]Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany — [2]Institute for Materials Science, TU Dresden, 01062 Dresden, Germany — [3]Theoretical Chemistry, TU Dresden, 01062 Dresden, Germany — [4]Chair of Molecular Functional Materials and Faculty of Chemistry & Food Chemistry, TU Dresden, 01062 Dresden, Germany

In the ongoing quest for miniaturization of machines, single molecule machinery holds a plethora of possibilities. Advancements in nanoscale imaging techniques, such as Scanning Tunneling Microscopy and Spectroscopy(STM, STS), allows not only the addressing of single molecules individually but through tunneling electrons/electric field stimulation with an STM tip, one can experimentally study fundamental properties of molecules such as pi-conjugation and charge transfer to a surface, and the effect it has on the switching behavior. We present the design and synthesis of a nanoswitch which exhibits a reversible switching from achiral to chiral mode on Au(111). The electronic states of both conformations have been measured with a high degree of spatial resolution, thereby showing the pathways of the electron-induced isomerization. Furthermore, we demonstrate this effect being suppressed on Ag(111).

O 29.6 Tue 11:45 S052

Enravelling effects of dispersion interactions in enantioselective adsorption — •Raymond Amador[1,2], Samuel Stolz[3], Nestor Merino-Diez[1], Oliver Groening[1], Roland Widmer[1], and Daniele Passerone[1,2] — [1]Empa - Dübendorf, Dübendorf, Switzerland — [2]ETH Zürich, Zürich, Switzerland — [3]Department of Physics, UC Berkeley, United States

van der Waals (vdW) interactions play a central role in a wide variety of systems. They are responsible for many natural processes, and thus, an accurate description of vdW forces is essential for improving our understanding physical phenomena. In this talk, we present our findings on the role of non-local vdW interactions in the enantioselective adsorption and debromination of the 10,10´-dibromo-9,9´-bianthracene (DBBA) and 9-Phenanthracenylboronic acid (9PBA) molecules, as catalyzed by chiral surfaces of the palladium-gallium (PdGa) intermetallic compound. After a brief recapitulation on the framework of density-functional theory (DFT), we discuss its principle shortcomings within the context of many-body perturbation theory, and how our current calculations address and rectify these faults via higher-order treatments of the exchange integrals. We then present several figures of interest: adsorption configurations before and after geometry optimizations and charge localization plots of adsorption, and explore effects of both vdW interactions and the nonlocal exchange term via inclusion of hybrid functionals. In doing so, we provide the foundations for further study of non-local electronic correlation in chirality.

O 29.7 Tue 12:00 S052

The role of Adatoms for the Adsorption of F4TCNQ on Au(111) — •Richard Berger, Andreas Jeindl, Lukas Hörmann, and Oliver Hofmann — TU Graz Institut für Festkörperphysik, Graz, Österreich

Molecular adsorption on inorganic substrates often includes the incorporation of native adatoms within the adsorbate layer. The presence of the adatom in the adlayer causes significant changes in the electronic structure of the interface affecting properties such as the adsorption geometry, the bonding type, and the work function. Here we investigate the adsorption of F4TCNQ on Au (111), which is a prototypical system for the adsorption of an acceptor type molecule on a metallic substrate. Using density functional theory, we show that incorporating adatoms significantly changes the interface charge transfer and modifies the Fermi-level pinning mechanism for the adsorbed species. Furthermore, we find that the 5d orbitals of the Au adatom hybridize with the F4TCNQ molecular orbitals, introducing covalent coupling within the adlayer. The combination of this effect explains why the incorporation of adatoms, despite the high cost of extracting them from the bulk, is energetically favorable.

O 30: Semiconductor Surfaces

Time: Tuesday 10:30–12:45 Location: S053

Topical Talk O 30.1 Tue 10:30 S053

Surface Phase Transitions in Atomistic Detail and with Femtosecond Resolution — •Wolf Gero Schmidt — Universität Paderborn

Ab initio molecular dynamics on ground and excited-state potential energy surfaces may be used to gain deep insight in the driving forces and mechanisms of surface phase transitions and can greatly assist the interpretation of experimental data. This is illustrated in my talk using two prominent examples: (i) Photoholes localized at the Brillouin zone boundary of the In/Si(111)(8x2) surface are shown to drive an ultrafast (8x2) ⟶ (4x1) phase change that is accompanied by an insulator-metal transition [1,2]. (ii) Thermal excitation of the Au/Si(553)(1x6) surface leads to soft Au chain vibrations that reduce transiently the Au electron affinity, which lowers the barrier for a $sp^2 + p \longrightarrow sp^3$ hybridization change of Si step edge atoms. This leads eventually to an order-disorder phase transition and the formation of a two-dimensional spin liquid [3].

[1] T Frigge et al., Nature 544, 207 (2017).
[2] CW Nicholson et al., Science 362, 821 (2018).
[3] C Braun et al., PRL 124, 146802 (2020).

O 30.2 Tue 11:00 S053

Dimer coupling energies of the Si(001) surface examined by SPA-LEED — •Christian Brand, Giriraj Jnawali, Jonas Fortmann, Mohammad Tajik, Alfred Hucht, Peter Kratzer, Hamid Mehdipour, Björn Sothmann, and Michael Horn-von Hoegen — Department of Physics and Center for Nanointegration CENIDE, University of Duisburg-Essen, Lotharstraße 1, 47057 Duisburg

Though the surface of Si(001) belongs to the most famous in the world still some of its properties and phenomena are unrevealed. Si(001) exhibits buckled dimers in the topmost layer, arranged in dimer rows, and thus forming a (2×1) reconstruction at room temperature. Upon cooling the structure undergoes an disorder-order transition to the $c(4\times2)$ reconstructed ground state. High-resolution SPA-LEED (Spot Profile Analyzing - Low Energy Electron Diffraction) was used to quantify the structural change along the transition upon heating from low to high temperatures. Rapid cooling in the regime of critical slowing down formed a so-called domain structure with typical size of ~ 14 nm. The data is analyzed in the framework of the anisotropic 2D Ising model and complemented by density functional theory calculations. We determined a phase transition temperature of $T_c = 190.6$ K, critical exponents β, γ, and ν of the Ising model and the coupling constants $J_\parallel = (-24.9\pm1.3)$ meV and $J_\perp = (-0.8\pm0.1)$ meV of the Si dimers by solving Onsager's equation and evaluating the correlation length ratio of the two directions.

O 30.3 Tue 11:15 S053

Reduced contact resistance to gallium nitride by plasma-assisted atomic layer deposition — •Maximilian Christis — Walter Schottky Institut, Technische Universität München

Gallium nitride is an industrially relevant III-V semiconductor that draws significant attention for a range of both established and emerging applications, including for light emitting diodes, power electronics, photocatalysis, and sensing. Established contacting schemes for GaN rely on wet-chemical surface preparations and post-metallization high-temperature (≥ 600°C) annealing processes, which complicate fabrication and may adversely affect device performance. Here, we present a low-temperature gas-phase process (200 °C) that reduces the Schottky barrier height and the contact resistivity at the GaN/metal interface. In particular, we employ H2 plasma-enhanced atomic layer deposition (ALD) that creates an ultimately thin, homogeneous AlOx monolayer using oxygen from the native gallium oxide as oxidant. This AlOx coating reduces the surface band bending and results in Ohmic current-voltage characteristics for n-doped GaN contacted by Ti metal. In ongoing work, we are also exploring how the monolayer AlOx ALD approach can be applied to improve contacts to p-doped GaN in combination with high work function metals. Among the various applications that could benefit from this low thermal budget contact fabrication strategy, we are investigating how such metal/semiconductor structures impact the electrocatalytic performance of n-GaN/Pt cathodes for water splitting.

O 30.4 Tue 11:30 S053

Distance dependance and lateral change of electrostatic forces between Pb-islands and wetting layer on Pb/Si(111)-(7x7) — Ben Lottenburger, •Paul Philip Schmidt, Daniel Rothhardt, Manuel Schulze, and Regina Hoffmann-Vogel — Universität Potsdam, Institut für Physik und Astronomie, Experimentelle Physik kondensierter Materie

Pb islands on silicon show a wide range of interesting properties, such as explosive island growth [1,2]. Previous work has already been able to explain some features, such as the unusual height distribution of the islands[3,4]. To understand this system in more detail, we have investigated the differences in the electrostatic interaction between the tip and the Pb islands on one side and the Pb-containing wetting layer on the other side. We have used scanning force microscopy in the non-contact frequency modulation mode and bias distance measurements in ultrahigh vacuum at ~120K. The Si has been cleaned by direct current heating. Subsequently, Pb has been vapor deposited. The Frequency shift as a function of tip-sample distance has been measured on both Pb islands and the wetting layer. The differences in force and work function between have been investigated both. [1] Hershberger et al, PRL 113, 236101 (2014). [2] Huang et al, PRL 108, 026101 (2012). [3] Hupalo et al, PRB 65, 115406 (2001). [4] Späth et al, PRL 124, 016101 (2020)

O 30.5 Tue 11:45 S053

Growth of well-ordered K3C60 thin films on Bi2Se3 — •Michael Herb and Isabella Gierz — Department of Physics, University of Regensburg, 93040 Regensburg, Germany

The molecular solid K3C60 is a BCS-type superconductor with a critical temperature of 20K [1]. More intriguingly, K3C60 powder exposed to strong mid-infrared driving fields exhibits the optical properties of a transient superconductor [2] possibly up to room temperature [3] and with nanosecond lifetimes [4]. The microscopic mechanism behind these observations remains poorly understood. We want to shed light onto this issue by measuring the transient band structure of driven K3C60 using time- and angle-resolved photoemission spectroscopy (tr-ARPES). These experiments cannot be performed on the originally used K3C60 powder as they require large, well-ordered K3C60 single crystals. Therefore, we investigate the growth of K3C60 thin films on different substrates using low-energy electron diffraction (LEED) and ultraviolet photoemission spectroscopy (UPS). We provide evidence for well-ordered growth on Bi2Se3, enabling future tr-ARPES studies on the material.

O 30.6 Tue 12:00 S053

Structure and origin of antiphase domains and related defects in thin GaP epilayers on As-modified Si(100) — •Franz Niklas Knoop[1], Agnieszka Paszuk[2], Benjamin Borkenhagen[1,3], Oliver Supplie[2,4], Manali Nandy[2], Gerhard Lilienkamp[1], Peter Kleinschmidt[2], Thomas Hannappel[2], and Winfried Daum[1] — [1]IEPT, TU Clausthal — [2]Institute of Physics, TU Ilmenau — [3]Fallstein Gymnasium Osterwieck — [4]Physics Department, HU Berlin

The deposition of low-defect III-V-layers on Si(100) is impaired by the formation of antiphase domains (APDs) in the epilayer. We study the origin and formation of APDs and related defects in thin GaP buffer layers on nearly-single-domain, As-modified Si(100) substrates. By comparing results obtained by low energy electron microscopy (LEEM), AFM, STM and scanning Auger electron microscopy, we identify two different types of APD-related defects in the GaP layer and trace these defects back to residual minority (2x1) domains of the Si substrate. GaP growth on minority domain terraces with widths in the range 40-100 nm gives rise to APDs of comparable lateral dimensions. The observation of trench-like defects in the epilayer extending down to the surface of the substrate indicates that homogeneous layer-by-layer growth of GaP is impeded on narrow terraces (<20 nm) of the (2x1)-reconstructed minority domain of the substrate. We propose that insufficient nucleation of GaP on these terraces leads to the formation of trenches, while on wider minority terraces APDs are formed by 3D-like growth.

O 30.7 Tue 12:15 S053

Adsorption of CO and CO$_2$ on the Y-stabilized ZrO$_2$(100) surface — •shuang chen, xiaojuan yu, eric sauter, alexei nefedov, stefan heissler, christof wöll, and yuemin wang — Institute of Functional Interfaces (IFG), Karlsruhe Institute of Technology (KIT), 76344 Eggenstein-Leopoldshafen, Germany

Understanding the stabilization mechanism of the Y- stabilized zirconia (YSZ) (100) surface has triggered a great debate over the past 30 years. In this work, we focused on a fundamental study on the surface structure of YSZ(100) by polarization-resolved infrared reflection absorption spectroscopy (IRRAS), in combination with grazing-emission X-ray photoelectron spectroscopy (XPS). The combined IR and XPS results allowed to gain detailed insight into the adsorption of carbon monoxide and carbon dioxide on YSZ(100) over a large coverage range from submonolayer to multilayers. The experimental data were further analyzed based on the density functional theory (DFT) calculations.

O 30.8 Tue 12:30 S053

Starting from a Fixed Geometry: Real-Time XPS Investigation of a Surface Reaction with Controlled Molecular Configurations — •Timo Glaser[1], Christian Länger[1], Julian Heep[1], Jannick Meinecke[2], Mathieu Silly[3], Ulrich Koert[2], and Michael Dürr[1] — [1]Institut für Angewandte Physik und Zentrum für Materialforschung, Justus-Liebig-Universität Giessen, Germany — [2]Fachbereich Chemie, Philipps-Universität Marburg, Germany — [3]Synchrotron SOLEIL, 91192 Gif sur Yvette, France

The relative orientation between two reactants can have a major influence on the reactivity and on the products of a chemical reaction. In gas-surface chemistry with well-defined single-crystal surfaces, the orientation and configuration of one reactant is fixed by the surface, but still the reacting gas molecules can impact on the surface in all possible orientations. Here we show how to constrain the relative orientation of both reactants by attaching a reactive group (ether) via a linker (cyclooctyne) on a single crystal surface. This keeps the reacting group in a highly-constrained configuration close to the surface. We demonstrate this concept for ether cleavage on silicon (001), the surface analogue of an S$_N$2 reaction. The kinetics of the further reaction of the ether group with the silicon surface is studied by means of real-time XPS using synchrotron radiation. We find both a low-energy barrier and a low prefactor, which we discuss in terms of the constrained starting configuration of the ether group. As this configuration represents the starting point of a low-energy pathway, it gives direct experimental access to the underlying reaction mechanism.

O 31: Solid-Liquid Interfaces 3: Reactions and Electrochemistry

Time: Tuesday 10:30–12:30 Location: S054

Topical Talk O 31.1 Tue 10:30 S054

Towards a realistic description of electrified solid-liquid interfaces — •Nicolas G. Hörmann — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Atomistic modelling of electrified solid-liquid interfaces in the context of electrocatalytic and electrochemical transformations is a challenging task. This is in particular so as the description of many relevant processes involves the consideration of charge transfer, solvent reorganization and the long-range screening of the electrolyte. Many of these processes have been studied in the past using effective model hamiltonians and from a physical chemistry perspective, e.g. based on mean-field kinetic models. However, recent advances in the first-principles-based description have opened up pathways to study the involved phenomena with atomistic resolution and to gain new, fundamental insights.

In this talk, I will give a brief overview of such recent developments, concentrating mainly on the use of and results from DFT calculations in continuum solvation environments[1]. Without doubt, such calculations have had a tremendous impact on the community, as they paved the way to describe the interfacial energetics in a grand-canonical framework, aka at applied potential conditions[2]. In addition to discussing a range of results obtained on model systems, I will as well clarify the evident limitations and inaccuracies of such an approach and discuss briefly possible future pathways to improve upon those.

[1] S. Ringe *et al.*, Chem. Rev. (2021). [2] N.G. Hörmann *et al.*, JCP, **150**, 041730 (2019).

O 31.2 Tue 11:00 S054

Electrostatic potentials in molecular dynamics — •Ludwig Ahrens-Iwers[1], Gregor Vonbun-Feldbauer[1], and Robert Meissner[2] — [1]Institute of Advanced Ceramics, Hamburg University of Technology, Hamburg, Germany — [2]Institute of Polymers and Composites, Hamburg University of Technology, Hamburg, Germany

Molecular dynamics (MD) simulations in a constant potential ensemble are an increasingly important tool to investigate charging mechanisms in next-generation energy storage devices. The constant potential method (CPM) can be used in classical MD to model metallic electrodes at an electrostatic potential. In this method, charges of individual electrode atoms are set to meet the applied potential. Unfortunately, existing implementations are either highly specialized or not very performant.

As a new implementation of the CPM, the ELECTRODE package for the MD code LAMMPS is presented. This package integrates a particle-mesh solver to greatly reduce computation times of the long-range Coulomb interactions. Further, a dipole correction that is required for systems with a slab geometry is included. In addition to the CPM, the code features a constant charge method which distributes the charges within each electrode as well as a thermopotentiostat that utilizes the CPM algorithm. Moreover, the integration in LAMMPS allows the use of many tools from the base code and other packages.

O 31.3 Tue 11:15 S054

First-principles molecular dynamics simulations of electrified Pt(111)/H2O interfaces — •Lang Li, Nicolas G. Hörmann, and Karsten Reuter — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Metal-water interfaces play a fundamental role in electrochemistry. An accurate understanding of their properties is required in any attempt to describe electrochemical phenomena such as electrocatalytic reactions or charge transfer processes.

In this work, we benchmark the description of electrified Pt(111)/water interfaces based on first-principles molecular dynamics simulations at applied potential conditions using density functional theory. We apply the potential by introducing excess electrons that are counterbalanced by partially charged hydrogen atoms. This method is tested with a variety of slab setups and cell sizes. Additionally, we investigate different methods to determine the reference potential.

We analyze in detail the structure of the interface as well as the obtained capacitance vs. potential curves and compare these with published theoretical and experimental results [1]. Our results highlight the response of interfacial water to an applied potential and its importance for understanding the hump in the capacitance, observed at high electrolyte concentrations [2].

[1] L. Li, J. -B. Le, J. Cheng, Cell Rep. Phys. Sci., 3, 100759 (2022). [2] J.B. Le, Q.Y. Fan, J.Q. Li, J. Cheng, Sci. Adv., 6, eabb1219 (2020).

O 31.4 Tue 11:30 S054

First step of the oxygen reduction reaction on Au(111): An ab initio molecular dynamics study of the electrified metal/water interface — •Alexandra M. Dudzinski, Elias Diesen, Hendrik H. Heenen, Vanessa J. Bukas, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin, Germany

The oxygen reduction reaction (ORR) is a key electrocatalytic process for developing sustainable energy technologies. And yet, many aspects of the underlying reaction mechanism are still poorly understood at the molecular level. Especially at weak-binding electrode surfaces such as Au, even the ability to bind aqueous O2 species as a first mechanistic step remain unclear. Resolving these questions requires going beyond the simplified thermodynamic models of charge-neutral reaction intermediates that have been commonly used in computational electrocatalysis so far. Here, we perform molecular dynamics simulations based on periodic density-functional theory (DFT) to investigate O2 adsorption at an electrified Au(111)/water interface. We elucidate structural interfacial properties as a function of surface charge, and show that the latter can significantly alter the O2 binding energy. Adsorption is specifically enhanced under electric fields that are realistic for ORR operation, suggesting this as a very possible first electrochemical, rather than purely chemical step of the mechanism and showing that field effects in corresponding DFT models cannot be neglected. The resulting dependence on (absolute) electrostatic potential may further explain the superior activity measured experimentally for this catalyst in alkaline vs. acidic media.

O 31.5 Tue 11:45 S054

Modeling varying potential conditions in electrochemical simulations: The case of O_2/Au(111) — •Elias Diesen, Alexandra M. Dudzinski, Hendrik H. Heenen, Vanessa J. Bukas, and Karsten Reuter — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin

While significant insight has been gained in recent years by DFT-based simulations of electrochemical processes, crucial methodological challenges remain, especially for accurate determination of reaction energies under operando conditions. One open question is how to represent the constant electrode potential in simulations of an electrified water/metal interface, where the simulation setup requires a constant charge in the simulation cell throughout the reaction. Here we compare different levels of treatment of the electrochemical interface: explicit ab initio molecular dynamics, an implicit solvent model, and a sawtooth-potential electric field in vacuum, for the case of O_2 adsorption on Au(111). We characterize the dynamics near the surface and identify ways an applied electrode potential influences the adsorption. We find, in all methods, significantly enhanced O_2 adsorption at more reducing conditions. However, we also show that certain aspects of the process can only be captured using a fully explicit treatment of the solvent.

O 31.6 Tue 12:00 S054

Understanding the Interfacial Capacitance of 2D Materials in an Implicit Water Environment — •Hedda Oschinski[1,2], Nicolas G. Hörmann[1], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Technical University of Munich, Germany

The interfacial capacitance (C) is a central quantity in electrochemistry. For metal electrodes, C is dominated by the double layer capacitance that derives from the potential drop in the solvent. However, the finite density of states (DOS) in semiconducting 2D electrodes alters the picture and leads to a vanishing C around the point of zero charge. This entails a challenge in describing the energy-potential relation and the connected field effects when considering adsorbates.

To explore this challenge, we study the interfacial capacitance for 2D metal halides MX_2, using density-functional theory in a continuum solvent environment. We break down C into a DOS-filling-related quantum capacitance and the double layer capacitance. Our analysis demonstrates that such a separation into individual components is not straightforward. Nevertheless, the qualitative behavior of C can be rationalized, making this study a first step towards better understanding of 2D, in particular semiconducting, electrodes.

O 31.7 Tue 12:15 S054

Two-Dimensional 2D Materials interfacing liquid water : the new frontier from ab initio simulations — Benoit Grosjean[1], Felix Mouhat[1], Rodolphe Vuilleumier[1], Francois-Xavier Coudert[2], and •Marie-Laure Bocquet[1] — [1]Ecole Normale Supérieure, PSL university and CNRS, Paris, France — [2]Chimie Paris Tech, PSL university and CNRS, Paris, France

In spite of their computational cost, quantum dynamic insights open unprecedented avenues for the use of 2D materials for nanofluidics. In this talk, I will report on our recent theoretical findings on 2D materials like G (graphene), BN (boron nitride) and GO (graphene oxide) by means of Ab Initio Molecular Dynamics (AIMD) and their implications to rationalize their peculiar fluid transport and filtration properties. We demonstrate that the charging of pristine ideal 2D materials in aqueous conditions originate from the hydroxide anion and is crucially dependent of the electronic structure of the 2D layers ranging from chemisorption to physisorption. [1] We could pioneer the concept of static versus mobile extrinsic charges on prototypical 2D materials that have a huge impact on electro-kinetic transport.[2] We also predict that the water reactivity is further enhanced on hydrid sheets like planar G-BN heterostructure [3]. Finally, we explore the properties of various GO models in neutral water and we unveil several chemical processes [4].

[1] Nat. Comm. 10, 1656 (2019). [2] J. Chem. Phys. 156, 044703 (2022). [3] Phys. Chem. Chem. Phys. 22, 10710 (2020). [4] Nat. Comm. 11, 1566 (2020).

O 32: Poster Tuesday: Adsorption and Catalysis 1

Time: Tuesday 11:00–13:00 Location: P3

O 32.1 Tue 11:00 P3

Analysis of Electron-Transfer in Water-Based Dye-Sensitized Solar Cells — •Daniel Holzhacker[1], Andreas Ringleb[1], Raffael Ruess[2], and Derck Schlettwein[1] — [1]Justus-Liebig-Universität Gießen, Institut für Angewandte Physik — [2]Justus-Liebig-Universität Gießen, Physikalisch-Chemisches Institut

Dye-sensitized solar cells (DSSCs) present a possible low-cost technology for the conversion of sunlight into electrical energy. Most DSSCs are based on electrolytes with organic solvents. Due to environmental reasons it is attractive to replace organic solvents by water. In view of the damage typically caused to DSSCs by water contaminations, the use of water as contact solution represents a big challenge. Complete aqueous DSSCs were assembled with a combination of different organic dyes (D51, D35 and Y123) and organic redox mediators (TEMPO, AZADO and TEMPOL). The combination of Y123 and TEMPO yielded remarkable efficiencies of up to 4.4%. Although heavily limited in the fill factor, TEMPOL proved to be an interesting alternative, as its solubility in water is dramatically increased in comparison to TEMPO. Similar open circuit voltages and short circuit currents were reached but large overpotentials of electron transfer at the electrodes still limited TEMPOL-based cells. Subsequently, the electrolyte and electrode surfaces were modified in order to reduce the respective overpotentials.

O 32.2 Tue 11:00 P3

Novel corrugated geometric moiré pattern of a semi-periodically buckled, zebra-like topography of Xe on Ag(110) — •Inga Christina Langguth and Karina Morgenstern — Chair of Physical Chemistry I, Ruhr-Universität Bochum, Bochum, Germany

Solid rare gas films on metal single crystals are currently gaining interest due to their beneficial properties. As non-ionic, low interacting insulator surfaces they offer a potential application for unperturbed surface science studies. STM investigations of a thin xenon (Xe) film of several monolayers on Ag(110) reveal a novel corrugated structure of a semi-periodically buckled, zebra-like topography. The outstanding feature of this surface is a non-uniform distribution of buckled row orientations and distances. Instead of domain boundaries separating different phases, a gradual transition of row orientations is observed. The corrugated row pattern can be attributed to a geometric moiré pattern at the interface of the incommensurable Ag(110) and Xe(111) crystal lattices. A gradual change of the row orientation can further be explained by a gradually changing Xe-lattice orientation with respect to Ag(110) within a narrow angle regime. The stability of this semi-periodic structure against annealing reveals a remarkably flat potential energy surface for the turning of the Xe layer relative to the Ag(110) surface. The corrugated surface may serve as an interesting substrate for further studies of geometric confinement effects in an anisotropic environment.

O 32.3 Tue 11:00 P3

On-surface Synthesis of Naphthalocyanines with Extended π-Systems — •Lukas J. Heuplick[1], Qitang Fan[1], Dmitriy A. Astvatsaturov[2], Dennis Körmer[1], Tatiana V. Dubinina[2], and J. Michael Gottfried[1] — [1]Fachbereich Chemie, Philipps-Universität Marburg, Marburg, Germany — [2]Moscow

The distinct optical and electronic properties of phthalocyanines (Pc) make them interesting in a wide field of applications. Here we report the on-surface synthesis of new naphthalocyanines with differently extended π-systems. This reaction in form of a cyclization is studied for different dicarbonitriles on the Ag(111) and Au(111) surfaces by scanning tunneling microscopy (STM) and X-ray photoelectron spectroscopy (XPS). 6,7-Diphenyl-2,3-naphthalenedicarbonitrile (DPNDN) forms naphthalocyanines only with co-adsorbed Fe atoms on Ag(111) and Au(111). The larger 6,7-di(2-naphthyl)-2,3-naphthalenedicarbonitrile (DNNDN) can undergo this reaction directly on Ag(111) without co-adsorbed metal, resulting in the corresponding Ag-NPc.

O 32.4 Tue 11:00 P3

Adaptive training of a machine-learned model for nonadiabatic hydrogen chemistry on multiple facets of Copper. — •Wojciech G. Stark, Julia Westermayr, Oscar A. Douglas-Gallardo, James Gardner, and Reinhard J. Maurer — University of Warwick, Coventry, United Kingdom

Traditionally, molecular dynamics methods utilise the Born-Oppenheimer approximation and dynamics are governed by a single potential energy surface. However, on metallic surfaces often the energy exchange between adsorbate and electronic excitations in the metal is significant and causes the breakdown of the Born-Oppenheimer approximation. There are multiple methods to include such nonadiabatic effects, with one of the most efficient being molecular dynamics with electronic friction (MDEF). MDEF introduces nonadiabatic effects via additional electronic friction forces, which can be calculated with time-dependent perturbation theory based on Density Functional Theory. However, a meaningful comparison between computational simulations and experiments demands the capability to run tens of thousands of MDEF trajectories. We present high-dimensional machine-learning based interatomic potential and electronic friction models that enable the simulation of nonadiabatic molecular dynamics of hydrogen scattering and associative desorption at different copper surfaces. We construct deep neural network representations via iterative adaptive sampling based on the target dynamical observables, namely the scattering and reaction probabilities.

O 32.5 Tue 11:00 P3

Adsorption and Diffusion of NH3 on Rutile TiO2(110): An STM Study — •Hanna Bühlmeyer[1], Kraen Christoffer Adamsen[2], Tao Xu[1], Lutz Lammich[2], Jörg Libuda[1], Stefan Wendt[2], and Jeppe Vang Lauritsen[2] — [1]Interface Research and Catalysis, ECRC, Friedrich-Alexander-Universität Erlangen-Nürnberg, Egerlandstraße 3, 91058 Erlangen, Germany — [2]Interdisciplinary Nanoscience Center, Aarhus University, Gustav Wieds Vej 14, 8000 Aarhus, Denmark

The adsorption of NH3 on TiO2 is an important step for many reactions in environmental catalysis such as the selective catalytic reduction (SCR) of NOX over V2O5/TiO2 and the removal of NH3 from air and water over TiO2. To obtain a detailed understanding of the surface chemistry of NH3 on TiO2, we investigated the adsorption of sub-monolayers of NH3 on clean, hydroxylated, and O-precovered rutile TiO2(110) surfaces by means of scanning tunneling microscopy (STM) under ultrahigh vacuum (UHV) conditions. We found that on the clean TiO2(110) surface at 120 K, NH3 adsorbs exclusively as monomers. On the hydroxylated TiO2(110) surface at 160 K, we observe 2(NH3) pairs and (NH3)2 dimers in addition to the majority of monomeric NH3. At 270 K, monomeric NH3 and surface hydroxyls diffuse together along the [001] direction. Additionally, NH3 can also diffuse along the surface hydroxyl bridge in the [1-10] direction. On the surface precovered by O adatoms (Oot) at 160 K, we observe the formation of NH2OH and 2(NH2OH) species formed by the interaction of NH3 monomers with Oot and Oot pairs.

O 32.6 Tue 11:00 P3

Real-space study of carbenes using a diazofluorene precursor adsorbed on a Cu(111) surface — •Hussain Mazhar[1], Julien Rowen[2], Wolfram Sander[2], and Karina Morgenstern[1] — [1]Lehrstuhl für Physikalische Chemie I, Ruhr-Universität Bochum, 44801 Bochum, Germany — [2]Lehrstuhl für Organische Chemie II, Ruhr-Universität Bochum, 44780 Bochum, Germany

Carbene is a reactive intermediate playing a vital role for many synthesis in the pharmaceutical industry [1]. Here, we study their properties on a metal substrate in real space using scanning tunnelling microscopy (STM) at 5 K. As carbenes are highly reactive species, they cannot be deposited directly on metal surfaces. The arrangement of their precursors on the surface is crucial for the carbene reactivity upon carbene formation. Diazofluorene molecules, already studied on the Ag(111) surface [2], are used as a precursor of the carbene to compare the influence of the surface on carbene reactivity. The STM study reveals dimer and cluster formation of the carbene precursor molecules on Cu(111) when deposited at 50 K and 100 K. While at 50 K more dimer formation occurs, at 100 K cluster formation is noticeable. An electrostatic surface potential map is used to determine charge distribution of the dimers. For carbene formation, a particular energy is required to cleave the diazo part from the molecule. In this study, we used STM manipulation to estimate the energy for the dissociation of the molecule. [1] Patil, Siddappa A, Future Med. Chem. 7. (2015) 1305-1333 [2] Mieres-Perez, Joel, J. Am. Chem. Soc. 143. (2021) 4653-4660

O 32.7 Tue 11:00 P3

Thickness-dependent energy level alignment of terrylene molecules on WS2 monolayer — •Qiang Wang[1], Sifan You[3], Björn Kobin[2], Patrick Amsalem[1], Lifeng Chi[3], Stefan Hecht[2], and Norbert Koch[1] — [1]Institut für Physik & IRIS Adlershof Humboldt-Universität zu Berlin, Berlin, Germany — [2]Department of Chemistry & IRIS Adlershof Humboldt-Universität zu Berlin, Berlin, Germany — [3]Institute of Functional Nano and Soft Materials (FUNSOM) , Soochow University, Suzhou, China

The exceptional large surface ratio makes 2D TMDC notably sensitive to extrinsic modification. In particular, we study the Van der Waals structure of WS2 monolayers modified by the conjugated terrylene molecules. We show the terrylene/WS2 heterostructures exist in different stacking configurations dependent on the layered coverage, revealed by photoemission spectroscopy and scanning tunnelling microscopy (STM). Electronically, the hybrid heterostructures exhibit type-II energy level alignment. It is further determined that the ionization potentials of terrylene, are reduced by 0.54 eV as the molecule switching from flat-lying to standing orientation. The adsorption behaviour is spatially resolved by STM, indicating two different atomic assembles. As determined by photophysical characterization, low energy electron-hole pairs are also populated upon molecule deposition. Meanwhile the external dielectric screening lowers the Rydberg states of WS2. Our findings reveal the vertical 2D heterostructures enable effective tailoring of both electronic and photophysical properties, which can be applied for various optoelectronic devices.

O 32.8 Tue 11:00 P3

Nanocar Race II: How fast and how long can we drive nano-vehicles? — •Tim Kühne[1], Kwan Ho Au Yeung[1], Suchetana Sarkar[1], Oumaima Aiboudi[2], Soyoung Park[2], Franziska Lissel[2], and Francesca Moresco[1] — [1]Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany — [2]Leibniz Institute of Polymer Research, 01062 Dresden, Germany

Scanning tunneling microscopy has progressed far beyond just being a probing technique. We can now address and manipulate single molecules and atoms with a great degree of precision and repeatability. The 2nd Nanocar Race saw surface science groups from across the globe design and synthesize molecules of at least 70 atoms with the intent of driving it on Au(111). The conversion of electric impulse from the tip to controllable translation of the nanocar along the FCC sites of Au(111) was demonstrated by each team, although the design philosophy and mediating physics theories varied across the board. Here, we present the TU Dresden nanocar. It was successfully manipulated via inelastic tunneling electrons for 290nm during the 24-hour race.

O 32.9 Tue 11:00 P3

First-Principles Study of Methanol and Benzene Adsorption on $In_2O_3(111)$ — •Andreas Ziegler[1], Margareta Wagner[2], Ulrike Diebold[2], and Bernd Meyer[1] — [1]Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany — [2]Institute of Applied Physics, TU Wien, Austria

Indium oxide is widely used as transparent conductive oxide for electrodes in semiconductor devices, but it is also a promising new catalyst for hydrogenation and dehydrogenation reactions, e.g. methanol synthesis. To probe the reactivity of the most stable (111) termination of In_2O_3, we studied the adsorption of methanol and benzene, two prototypical polar and nonpolar molecules, by DFT geometry optimizations and Car-Parrinello molecular dynamics (CPMD) simulations. We find that the unit cell of the $In_2O_3(111)$ surface is chemically quite heterogeneous: by searching for the most favorable configurations of methanol with increasing coverage from one to nine molecules per unit cell we find that the first three methanol molecules dissociate in one specific region of the unit cell, followed by molecular adsorption on neighboring sites, confirmed by TPD, XPS and ncAFM experiments. Also benzene prefers to adsorb at one specific site in the unit cell. Due to the large size of the unit cell, the benzene molecules are well separated. CPMD simulations show that the molecules can freely rotate and are well trapped at their adsorption site. However, rotation is suppressed for benzene derivatives with additional side groups.

O 33: Poster Tuesday: Ultrafast Processes 2

Time: Tuesday 11:00–13:00 Location: P3

O 33.1 Tue 11:00 P3

Anisotropic carrier dynamics in single crystalline graphite — •Hauke Beyer, Petra Hein, Kai Rossnagel, and Michael Bauer — Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany

Time- and angle-resolved photoelectron spectroscopy at 35 fs time-resolution is employed to study the carrier dynamics in the Dirac cone of graphite upon excitation with linearly polarized light. Due to the pseudo spin degree of freedom, the nascent photogenerated carrier distribution exhibits a strong anisotropy in momentum space. We observe the formation of a quasi-thermalized distribution on ultrafast timescales (~ 10 fs) through e-e interactions exhibiting a clear azimuthal anisotropy similar to findings for graphene [1]. For the azimuthal thermalization our data reveal a characteristic time scale of 40 ± 10 fs. The residual non thermal part of the electron distribution shows a time-dependent shift in energy by ~ 150 meV, which we assign to the formation of a steplike distribution due to e-ph interactions. The results are in good qualitative agreement with calculations based on a model introduced in Ref. [2].

[1] S. Aeschlimann *et al.*, Phys. Rev. B **96**, 020301(R) (2017).
[2] E. Malic *et al.*, Phys. Rev. B **84**, 205406 (2011).

O 33.2 Tue 11:00 P3

High-harmonic generation from the surface state of Bi_2Se_3 with THz driving fields — •Tim Bergmeier, Suguru Ito, Jens Güdde, and Ulrich Höfer — Fachbereich Physik, Philipps-Universität Marburg, Germany

The acceleration of charge carriers through the Dirac-point in the topologically protected surface state (TSS) of topological insulators (TIs) by strong electric fields with frequencies in the THz range gives rise to a nontrivial type of unusually efficient high-harmonic generation (HHG) as demonstrated for Bi_2Te_3 [1]. The long scattering times in the TSS resulting from spin-momentum locking are imprinted in the observation that the high-harmonic orders can be continuously shifted in frequency by varying the carrier-envelope phase of the driving field. This makes it possible to investigate the unusual transport properties in the TSS by this all optical method even at buried interfaces.

Here, we present first results for Bi_2Se_3 by using a newly developed setup that enables THz-HHG with a stable carrier-envelope phase at a repetition rate of 200 kHz reaching field strengths of up to 10 MV/cm in the frequency range of 12-90 THz. We show how the contribution of electrons in the TSS to the HHG spectra can be determined by measurements with varying field strengths and THz-frequencies above and below the bulk band gap, while analyzing the polarization of the high harmonic signal for different directions of the excitation in momentum space.

[1] C. P. Schmid *et al.*, Nature 593, 385 (2021).

O 33.3 Tue 11:00 P3

Towards time-resolved photoemission orbital tomography on van-der-Waals heterostructures — •Wiebke Bennecke, David Schmitt, Jan Phillip Bange, Mattis Langendorf, Katharina D. Feeser, Daniel Steil, Sabine Steil, Marcel Reutzel, G. S. Matthijs Jansen, and Stefan Mathias — I. Physikalisches Institut, Georg-August-Universität Göttingen

Heterostructures of two-dimensional van-der-Waals materials with molecular thin films provide an exceptional platform to tailor electronic energy level alignments that govern the optoelectronic response of such materials. In addition to the energy-level alignment, a detailed knowledge of the electronic wavefunctions in these systems would help to fundamentally understand optical excitations, exciton generation, charge-transfer and relaxation processes. Here, time-resolved orbital tomography is a promising method that potentially provides such information. On this poster, we show first results of femtosecond orbital tomography of PTCDA monolayers adsorbed on bulk as well as monolayer WSe_2.

O 33.4 Tue 11:00 P3

Transient optical properties in non-equilibrium laser excited noble metals — •Marius Wenk[1], Pascal D. Ndione[1], Sebastian T. Weber[1], Dirk O. Gericke[2], and Baerbel Rethfeld[1] — [1]Department of Physics and OPTIMAS Research Center, Technische Universität Kaiserslautern — [2]CFSA, Department of Physics, University of Warwick

Ultrashort laser pulses can induce strong modifications of material properties of solids such as creating highly transient optical parameters. After excitation with lasers of high power, the conduction electrons thermalize quickly to a hot Fermi distribution. Yet, the band occupation numbers can still be far from equilibrium.

We study excitation of noble metals such as copper and gold with visible photons. We use a two-temperature model and construct electron density-resolved rate equations to simulate the non-equilibrium band occupation [1]. Applying the results of the simulation, particularly the occupation and temperature data, we compute the time-resolved dielectric function based on the Drude-Lorentz formalism. In order to compare the results with experimental data, we calculate the transient optical properties such as probe reflectivity and transmissivity. Our predictions are compared with time-resolved measurements of optically excited metals, thus providing insights to electron dynamics on femto- and picosecond timescales.

[1] Pascal D. Ndione, Sebastian T. Weber, Dirk O. Gericke, and Baerbel Rethfeld. Scientific Reports, 12(1) 4693 (2022)

O 33.5 Tue 11:00 P3

LabVIEW based software solution for tr-ARPES experiments — •Johannes Gradl[1], Niklas Hofmann[1], Leonard Weigl[1], Yu Zhang[2], Cephise Cacho[3], Neeraj Mishra[4,5], Stiven Forti[4], Camilla Coletti[4,5], and Isabella Gierz[1] — [1]Department of Physics, University of Regensburg, 93040 Regensburg, Germany — [2]Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot OX11 0DE, United Kingdom — [3]Diamond Light Source, Harwell Campus, Didcot OX11 0DE, United Kingdom — [4]Center for Nanotechnology Innovation @NEST, Istituto Italiano di Tecnologia, 56127 Pisa, Italy — [5]Graphene Labs, Istituto Italiano di Tecnologia, 16163 Genova, Italy

Time- and angle-resolved photoemission spectroscopy (tr-ARPES) is an experimental technique used for visualizing non-equilibrium carrier dynamics as well as transient band structures of photoexcited samples as a function of energy, momentum, and time. A tr-ARPES setup ideally includes femtosecond pump pulses with tunable photon energy, femtosecond extreme ultraviolet probe pulses an ultrahigh vacuum chamber equipped with a cryo-cooled sample manipulator and a photoelectron analyzer as well as additional features for in situ sample preparation and characterization. In addition to this hardware, the execution of tr-ARPES experiments requires a user-friendly software for hardware control, data acquisition, and real time data visualization. We will present our LabVIEW based software solution and demonstrate its successful implementation with a series of data sets from a WS2/graphene van der Waals heterostructure.

O 33.6 Tue 11:00 P3

Ultrabroadband THz-STM and its application to study hot electron dynamics in metals — Natalia Martín Sabanés[1,2], Faruk Krecinic[1], •Vivien Sleziona[1], Takashi Kumagai[1,3], Fabian Schulz[1,4], Luis Enrique Parra Lopez[1], Alkisti Vaitsi[1], Martin Wolf[1], and Melanie Müller[1] — [1]Fritz-Haber Institute of the Max-Planck Society, Berlin, Germany — [2]IMDEA Nanoscience, Madrid, Spain — [3]Institute of Molecular Science, 444-8585 Okazaki, Japan — [4]CIC NanoGUNE BRTA, San Sebastian, Spain

Localized ultrafast currents across the junction of a scanning tunneling microscope (STM) can be generated by photo-assisted hot electron tunneling or *cold* lightwave-induced tunneling. In addition, but so far not considered, fs laser excitation can induce a transient thermalized electron distribution that can give rise to an ultrafast current component. Here we investigate the role of ultrafast thermionic tunneling for photoinduced hot electron tunneling from a photoexcited STM tip [1]. We access the dynamics of hot electron tunneling by phase-resolved sampling of ultrabroadband Terahertz (THz) waveforms inside the STM junction. Our results reveal the strong nonthermal character of photoinduced hot electron tunneling, and provide a new route to probe hot electron dynamics in metals using THz-STM. Furthermore, we report on the development of an ultrabroadband THz-STM with tunable optical excitation for the spatiotemporal investigation of photocarrier dynamics at metal-semiconducting interfaces. [1] N. Martín Sabanés et al., 10.48550/arXiv.2205.08248

O 33.7 Tue 11:00 P3

Dark Exciton Formation Dynamics in TMDC Monolayers — •Sarah Zajusch[1], Lasse Münster[1], Raul Perea-Causin[1], Samuel Brem[1], Katsumi Tanimura[1], Jens Güdde[1], Yaroslav Gerasimenko[2], Rupert Huber[2], Ermin Malic[1], Ulrich Höfer[1], and Robert Wallauer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Fachbereich Physik, Universität Regensburg, Germany

Charge transfer processes in two-dimensional TMDCs are governed by the formation of excitons. The excitonic landscape comprises optically accessible bright excitons as well as momentum- and spin-forbidden dark states. Our experimental setup combines time-resolved momentum microscopy with probe energies in the XUV regime which provides direct access to k-resolved exciton dynamics within the whole Brillouin zone on an ultrafast time scale.

Although structural features are very similar for different semiconductor TMDC materials, slight variations in both electronic band ordering and excitonic binding energy can drastically modify the excited population dynamics. We present a comparison of exciton formation in the two extreme cases of WS_2 and $MoSe_2$ monolayers after valley-sensitive excitation with circular polarized light. On the one hand, in $MoSe_2$ the bright KK-exciton is the energetically most favorable state. On the contrary, in WS_2, we additionally observe the ultrafast formation of energetically lower dark KK'- and KΣ-excitons. The microscopic understanding of these processes is crucial with regard to interlayer exciton formation in TMDC heterostructures.

O 33.8 Tue 11:00 P3

Time- and Angle-Resolved Photoelectron Spectroscopy with Nano-Focused Surface Plasmon Polaritons — •Alexander Neuhaus, Pascal Dreher, David Janoschka, Michael Horn-von Hoegen, and Frank-J. Meyer zu Heringdorf — Faculty of Physics and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048 Duisburg, Germany.

Non-perturbative interactions of intense light fields with the electronic band structure in a solid can result in transient electronic properties. The experimental conditions required to realize the necessary field strength can be realized in nano-optical systems, as these can be designed to provide tremendous enhancements of the local field amplitude. Ultimately, observing the non-equilibrium electron dynamics in such systems requires a combination of precise control over the local driving field, state resolution, and spatial selectivity.

Here, we explore electron emission from nano-focused femtosecond surface plasmon polariton (SPP) pulses, providing us with deep-subwavelength spatial selectivity. Time- and angle-resolved photoelectron spectroscopy with attosecond precision provides us access to the coherent and incoherent dynamics of the electron emission process. The technique is applied to the system Cs/Au(111), where we find a resonant enhancement of the electron emission by an image potential state.

O 33.9 Tue 11:00 P3

Ultra Fast Dynamics in Modified Thiophene based Conjugated Donor-Acaptor Organcic Polymers — •Tobias Reiker[1,2], Carsten Winter[1], Deb Kumar Bhowmick[1,2], Nils Fabian Kleimeier[1,2], Zitong Liu[3], Deqing Zhang[3], and Helmut Zacharias[1,2] — [1]Center for Soft Nanoscience, University of Münster, Germany — [2]Physikalisches Institut, University of Münster, Germany — [3]Institute of Chemistry, Chinese Academy of Science, Beijing, China

Thiophene-based polymers are promising candidates for solar cell, OLED or transistor applications. An internal donor - acceptor system is formed by coupling thiophene polymers with pyrrole chains. The charge transport behavior can be tuned by different alkyl side chains since they influence the electronic structure. A direct assessment of the intramolecular and intermolecular dynamics may guide synthesis routes. With pDPP4T, pDPP4T and pDPTTT we investigated the electronic dynamics of verified high hole-mobility organic semiconductors. Either the backbone or the side chains were modified. In contrast, another polymer pF8T2 with bi-thiophene in the backbone was used, but with fluorene instead of pyrrole as acceptor. These different molecular configurations are intended to provide insights into the change in electron configuration due to both backbone modification and intermolecular packing. We report results of temporally resolved photoemission studies on thiophene polymers on silicon substrates.

O 33.10 Tue 11:00 P3

Density-dependent electron-phonon coupling in multiband systems. — •Tobias Held, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany

If a solid is irradiated with a short-pulsed laser in the visible spectrum, the energy is almost entirely absorbed by the electrons while the lattice remains cold. The subsequent energy flow between electrons and phonons is usually described by the electron-phonon coupling parameter, which plays a central role in the Two-Temperature Model and most other temperature-based models. This coupling parameter depends on a multitude of observables. Most frequently a dependence on the electron temperature is considered.

In this work, we aim to investigate how a varying density distribution between different electron subsystems affects the coupling parameter. In gold, we distinguish between sp- and d-electrons and in magnetic nickel between majority and minority spins. Our results show that for gold, the total coupling strongly depends on the density distribution, while for nickel it is largely independent on the spin densities. In the latter case, the individual coupling contributions of the bands change significantly with density but mostly compensate each other in terms of the total coupling.

O 33.11 Tue 11:00 P3

Electron Dynamics of an intercalated Graphene Layer on Nickel — •Katharina Hilgert[1], Christina Schott[1], Eva Walther[1], KaMan Yu[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, 67663 Kaiserslautern Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, 55128 Mainz, Germany

One of the great challenges in information technology is to develop novel concepts for the realization of active functional units on eversmaller length scales. The simplest approach to reduce the size of any device structure is to employ atomically thin materials as graphene with their unique electronic and optical properties. However, these are often altered on surfaces due to strong chemical interaction with the substrate material. On this poster we present our approach to restore and design the electronic properties of graphene on a Ni(111) surface by intercalation of lead atoms. The changes in the electronic band structure of graphene on Ni(111) are monitored by angle resolved photoelectron spectroscopy with extreme ultra violet radiation. On the highly reactive Ni(111) surface, the linear dispersion of the Dirac cone of the free-standing graphene sheet is completely suppressed by the strong graphene surface interaction. This changes significantly after the intercalation of Pb. The band structure of the graphene/Pb/Ni(111) system reveals again the linear dispersion resembling the behavior in free-standing graphene. This has clear consequences on the carrier dynamics near the K-point as we will discuss in this contribution.

O 34: Poster Tuesday: Scanning Probe Techniques 2

Time: Tuesday 11:00–13:00 Location: P3

O 34.1 Tue 11:00 P3

Implementation of a Scanning Tunneling Microscope for Measurements in Electrochemical Environment — •Fabian Schröfel, Matthias Greve, Karsten Tarhouni, and Olaf Magnussen — Institute of Experimental and Applied Physics, Kiel University, Kiel, Germany

The atomic-scale understanding of processes at the interface between solid electrodes and liquid electrolytes is of high importance for electrochemical energy storage and conversion. Electrochemical scanning tunneling microscopy (ECSTM) is a key technique for the investigation of such interfaces. Operating an STM in an electrochemical environment requires special measures, as the potentials of both STM tip and sample need to be controlled and electrochemical currents at the tip need to be kept way below the tunneling current.

Here, we report details on a new ECSTM built in our group. It consists of a newly developed STM head that is optimized for studies in electrochemical environment, high scanning frequencies, and low thermal drift. The instrument is based on a commercial SPECS Nanonis STM controller that we equipped with a custom-build bipotentiostat and coarse-approach. A suitable control software for electrochemical studies, which we integrated into the Nanonis software, allows to perform cyclic voltammetry parallel and separate from the STM measurements. Characterization of the mechanical stability of the STM and first STM images and electrochemical data will be presented.

O 34.2 Tue 11:00 P3

Determining the phase transfer function of an STM for coherent spin operations — •Evert Stolte — TU Delft

Coherent control of single spin transitions in atoms on a surface has been achieved with electron spin resonance scanning tunnelling microscopy (ESR-STM).[1] Extending that control to a series of sequential gates on different transitions and developing more complex gates requires radio frequency (RF) pulses at different frequencies with controlled relative amplitudes and phases, which are both affected by the transfer function of the power line to the tunnel junction. While the amplitude transfer function can be determined through a well-described rectification procedure [2], characterizing the phase transfer function remains challenging. Straightforward transmission or reflection measurements are excluded as it is not possible to separate phase rotations incurred on the way into the STM from those happening on the outward journey.

Here we report on the development of an in-situ method to determine the phase transfer function at radio frequencies that can be readily implemented to standard ESR-STM setups. The method is based on the envelope detection of the beat signal that is generated by adding two continuous wave RF signals separated by an audio frequency. The effectivity of the procedure is tested through pump-probe autocorrelation measurements with square pulses, which should show a reduced minimum width if the phase correction is successful.

[1] Yang, K. (2019). Science, 366(6464), 509-512.

[2] Paul, W. (2016). Review of Scientific Instruments, 87(7).

O 34.3 Tue 11:00 P3

Performance of an electrically driven q-plus sensor in a commercial Joule Thomson STM — •Hester Vennema, Laëtitia Farinacci, and Sander Otte — Delft University of Technology, Delft, The Netherlands

The q-plus sensor is a wide-spread tool for performing combined STM-AFM measurements in ultra-high vacuum at cryogenic temperatures. Here, we implement the use of q-plus sensors in a JT-STM. Contrary to other set-ups in which q-plus sensors are used, we do not drive our sensor mechanically, via an excitation of the Z-piezo, but electrically, with the excitation signal directly sent to the tuning fork.

In order to characterize the performance of our homemade q-plus sensors we develop a set-up to test their response to an electrical drive in ambient conditions. Following M. Lee et al. [1], we can disentangle the mechanical and electrical response of the sensors to the driving signal. After transfer into the JT-STM, we demonstrate the possibility to use electrically driven q-plus sensors for combined STM-AFM measurements: with a Q factor around 20000 we can control the amplitude of the oscillation to be as low as 70 pm. Our first principle measurements are performed on a CuCl2/Cu(100) surface [2]. We investigate with local contact potential difference measurements the local change of work function that leads to the confinement of field emission resonances above such vacancy patches.

[1] M. Lee et al., Appl. Phys. Lett. 91, 023117 (2007)

[2] R. Rejali et al., arXiv:2204.10559 (2022)

O 34.4 Tue 11:00 P3

Development of a compact millikelvin STM for single spin resonance — •Daria Sostina[1], David Collomb[2], Wantong Huang[2], Mate Stark[2], Christoph Sürgers[2], Philip Willke[2], and Wolfgang Wernsdorfer[2] — [1]Institute for Quantum Materials and Technologies (IQMT), Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany

In the last decade detection and manipulation of spins at the atomic scale has been achieved by combining techniques like electron spin resonance (ESR) with scanning tunneling microscopy (STM) [S. Baumann et al., Science 350 (6259), 417-420 (2015)]. However crucial properties of potential quantum spins such as the spin relaxation time T1 and the phase coherence time T2 remain short. Both T1 and T2 are affected by the proximity to the tip and substrate, which provide thermally excited electrons [P. Willke et al., Science Advances 4(2), 1543 (2018)]. A potential solution lies in lowering the temperatures utilizing dilution refrigerators (DR). Here, we present the design and implementation of a unique DR-STM optimized for ESR reaching millikelvin temperatures. Since the ground state population scales with the resonance frequency, better RF transmission at high frequencies is also desired for ESR-STM. In the compact dilution fridge, the RF line is optimized by a short total length of cables as well as using high-frequency cabling up until the tip. As a result, we believe that our compact DR-STM will help to improve ESR-STM paving the way for quantum information processing using single spin centers on surfaces.

O 34.5 Tue 11:00 P3

Design of a high-stability miniaturized scanning tunneling microscope for small-bore cryostats — •Felix Huber, Stephan Spieker, and Sebastian Loth — University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany

Low-temperature scanning tunneling microscopy (STM) setups are typically housed in large cryostats and require proportionally large vacuum chambers, as well as extensively shielded custom-built laboratories to reach the signal-to-noise ratios (SNR) desired for cutting-edge reasearch. Yet, by miniaturizing the STM-head, the SNR can be significantly improved, due to the favorable scaling of resonance frequencies [1], thermal characteristics and the cryostat hold time. The STM design presented here, is optimized to work in noisy environments, provides optical access to the tunnel junction, and can be used in a standard-bore cryogenic dewar due to its small volume and dimensions [2]. This design could be used for experiments requiring long averaging times, and may serve as an easy upgrade to existing room-temperature setups.

[1] C.R. Ast, et al. Rev. Sci. Instrum. 79, 093704 (2008).

[2] R. Schlegel, et al. Rev. Sci. Instrum. 85, 013706 (2014).

O 34.6 Tue 11:00 P3

Functionalized Tips in low-temperature AFM/STM - Fabrication and Application — •Lorenz Brill, Marco Grünewald, Roman Forker und Torsten Fritz — Friedrich-Schiller-Universität, Jena, Germany

AFM with functionalized tips has been an emergent and fast growing field in the last couple of years. The chemical passivation of the metal tip achieved via functionalization allows for images with atomic or intramolecular resolution to be recorded.

In our contribution, we detail the fabrication of CO-functionalized tips with a commercial low-temperature STM/AFM by Specs Surface Nano Analysis GmbH and demonstrate atomic resolution on a Cu(111)-surface. Furthermore, we show structure elucidation of organic molecular islands on the Cu(111)-substrate via atomic resolution images.

This technique shows great promise for further investigations, especially in areas not accessible via conventional STM, e.g. detailed structure analysis of 1,4-benzoquinone on metal substrates.

O 34.7 Tue 11:00 P3

Inspecting non-linearities in scanning force microscopy — •Lukas Böttcher[1], Anna Dittus[2], Jens Starke[2], Ingo Barke[1], and Sylvia Speller[1] — [1]Institute of Physics, University of Rostock — [2]Institute of Mathematics, University of Rostock

In dynamic force microscopy bistable states are often encountered. This includes two stable states at small and large amplitude flanking one unstable state at intermediate amplitude, separating two attracting regions. Our aim is to investigate the behavior of oscillating cantilevers in dynamic force microscopy [1,2]. We acquired distance dependent frequency sweeps of amplitude and phase. From this we determined saddle-node branches with cusps and compare the behavior with simulations.

[1] A.C. Boccara et al., Applied Physics Letters 58 (1991).

[2] Robert W. Stark, Materials Today 13 (2010)

O 34.8 Tue 11:00 P3

Setup for laser-based time-resolved momentum microscopy — •Felix Passlack, Stefano Ponzoni, Giovanni Zamborlini, and Mirko Cinchetti — Department of Physics, TU Dortmund University, Otto-Hahn-Straße 4, Dortmund, Germany

In this work, we characterize the performance of a recently installed time-resolved angular-resolved photoemission spectroscopy (trARPES) system. trARPES is performed by coupling an optical beamline for pump-probe spectroscopy[1] to a state-of-the-art momentum-microscopy photoemission spectrometer (KREIOS MM, SPECS GmbH). This allows for an almost continuous tunability of the pump photon energy between $0.5eV$ and $3.8eV$ while capturing the full photoemission horizon of the $6eV$ probe. The performance of the system is characterized through a set of measurements on the topological insulator Bi_2Se_3, for which both the static electronic structure and the out of equilibrium electron dynamics are already extensively studied in literature following the evolution of the electronic structure with an energy resolution in the order of $50meV$, a momentum resolution of $0.005A^{-1}$and a time resolution below $500fs$ throughout the whole photoemission horizon. In particular, thanks to the ultimate angular acceptance of the photoemission microscope, we are able to track the dispersion of the photoexcited states in Bi_2Se_3 up to $\pm0.48A^{-1}$ and $1eV$ above the Dirac point, beyond the practical limitations of conventional high-resolution ARPES spectroscopy with $6eV$ photons.

[1] F. Mertens et al., Review of Scientific Instruments **91** (2020)

O 34.9 Tue 11:00 P3

Development and characterization of a Herriott-type multipass cell compression-setup for fs-pulses and variable repetition rates — •Lasse Sternemann[1], Karl Schiller[1], Alan Omar[2], Matija Stupar[1], Mirko Cinchetti[1], and Clara Saraceno[2] — [1]Department of Physics, TU Dortmund University, Otto-Hahn-Straße 4, 44227 Dortmund, Germany — [2]Ruhr-Universitaet Bochum, Germany

We present developement and characterization of a Herriott-style multipass cell aimed at achieving ultrashort pulses with high peak power. The setup is used as a driving laser for a high repetition rate HHG (High Harmonic Generation) source for time-resolved momentum microscopy.

The setup compresses the pulses generated by a commercial Ytterbium-based laser (Carbide, Light Conversion) with an average power of 50 W and a variable repetition rate between 100 kHz and 1 MHz. The pulses are first spectrally broadened by self-phase-modulation by passing through a non-linear $\chi^{(3)}$ medium multiple times. Then, a pair of negative group-dispersion-delay coated mirrors compress the pulse by dispersion correction. By moving the non-linear medium within the mulitpass cell we were able to compress the pulses from 242 fs to around 45 fs for different high repetition rates while keeping a transmission of over 90 % and peak powers between 1 GW and 2 GW [1].

[1] A. Omar, et al. *Advanced Solid State Lasers*, OSA Technical Digest (Optical Society of America, 2021), paper JM3A.55

O 34.10 Tue 11:00 P3

Multispectral time-resolved energy-momentum microscopy using high-harmonic extreme ultraviolet radiation — •Nils Wind[1,2], Michael Heber[3], Dmytro Kutnyakhov[3], Federico Pressacco[3], and Kai Rossnagel[2,4] — [1]Institut für Experimentalphysik, Universität Hamburg, 22761 Hamburg, Germany — [2]Ruprecht-Haensel-Labor, Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany — [3]Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany — [4]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

A 790-nm-driven high-harmonic generation source with a repetition rate of 6 kHz is combined with a toroidal-grating monochromator and a high-detection-efficiency photoelectron time-of-flight momentum microscope to enable time- and momentum-resolved photoemission spectroscopy over a spectral range of 23.6–45.5 eV with sub-100-fs time resolution. Three-dimensional (3D) Fermi surface mapping is demonstrated on graphene-covered Ir(111) with energy and momentum resolutions of $\lesssim$100 meV and $\lesssim$0.1 Å^{-1}, respectively. The table-top experiment sets the stage for measuring the k_z-dependent ultrafast dynamics of 3D electronic structure, including band structure, Fermi surface, and carrier dynamics in 3D materials as well as 3D orbital dynamics in molecular layers.

O 35: Poster Tuesday: Plasmonics and Nanooptics 1

Time: Tuesday 11:00–13:00 Location: P3

O 35.1 Tue 11:00 P3

Positioning of DNA origami based nanostructures on surfaces by lithographic patterning — Zhe Liu[1], Zunhao Wang[2], Birka Lalkens[3], Daesung Park[2], Jannik Guckel[2], Juliane Breitfelder[3], and •Markus Etzkorn[1] — [1]Institute of Applied Physics, Technische Universität Braunschweig, 38106 Braunschweig, Germany — [2]Physikalisch Technische Bundesanstalt, 38116 Braunschweig, Germany — [3]Institute of Semiconductor Technology, Technische Universität Braunschweig, 38106 Braunschweig, Germany

Self-assembly protocols of functionalized DNA-origami structures can be used to create large amounts of identical hybrid nanostructures. We use this approach to create structures with tunable plasmonic properties. In our study, dimers of 15 nm gold nanoparticles with an average gap of 7 nm were self-assembled in solution. In order to control the positioning of such DNA-origami structures on surfaces, we created surface areas with hydrophobic/hydrophilic contrast by electron beam lithography and dry oxidative etching. This protocol offers positional control on the sub 10 nm scale. Here we will present results for hexamethyldisilazane (HDMS) covered silicon, but the approach can be utilized for various substrates and nanostructures. Our origami/dimer structure shows highly selective adsorption on different lithographically patterned structures. We achieve an efficiency in positioning, that is the ratio of origami in wanted to those in unwanted positions, of above 95%. Important parameters controlling the efficiency are discussed.

O 35.2 Tue 11:00 P3

Investigation of the plasmonic Phase-Change Materials $AgSnTe_2$ and In_3SbTe_2 for tuning nanoantenna resonances — •Kilian Wilden, Lukas Conrads, Andreas Hessler, Matthias Wuttig, and Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

Providing a high optical contrast between their amorphous and crystalline phases, phase-change materials (PCMs) can serve for many applications in nanophotonics. [1] Optical pulses can be used for switching between those two states. This enables the tunability of antenna resonances by changing the surrounding medium. Currently, a novel material class of switchable infrared plasmonic PCMs is rising, which is characterized by a negative permittivity and therefore a Drude-like behaviour in the crystalline. This plasmonic PCM In_3SbTe_2 (IST) offers resonance tuning by reconfiguring the antenna shapes of rod antennas and even more complex ones with magnetic resonances.[2] $AgSnTe_2$ (AST) is another plasmonic PCM with a smaller plasma frequency and higher plasmonic losses than IST. Here, we investigate directly optically written antenna structures of crystalline AST. The quality of the plasmonic resonances is compared to IST antennas. Furthermore, the well-established concepts of reconfiguring antenna geometries are applied to inverse antenna structures, in which amorphous holes are created in a crystalline plasmonic surrounding. Their resonance behavior can be described by Babinet's principle. [1] M. Wuttig, H. Bhaskaran and T. Taubner, Nature Photonics 11, 465-476 (2017). [2] A. Heßler et al., Nature Communications 12, 924 (2021).

O 35.3 Tue 11:00 P3

Large area writing of reconfigurable metasurfaces with the plasmonic PCM In_3SbTe_2 — •Natalie Honné[1], Lukas Conrads[1], Andreas Ulm[2], Andreas Hessler[1], Matthias Wuttig[1], Robert Schmitt[2], and Thomas Taubner[1] — [1]I. Institute of Physics (IA), RWTH Aachen University — [2]Fraunhoferinstitut für Produktionstechnologie

Phase-change materials (PCMs) are optimal candidates for optical components being non-volatile and reversibly switchable. Their amorphous and crystalline phases differ tremendously in their properties. Conventional PCMs exhibit a large contrast in their refractive index and can be used to tune antenna resonances by influencing their surrounding.[1] Recently, plasmonic PCMs, like In_3SbTe_2, has been introduced. They switch between a dielectric (amorphous) and a metallic (crystalline) phase in the complete IR and enable optical writing of metallic nanoantennas directly in a dielectric surrounding.[2] Large area writing of reconfigurable metasurfaces is a key feature for tuneable nanophotonic devices. Here, the Nanoscribe Photonic Pofessional GT is used, a femtosecond laser writer with an infrared wavelength. Large area direct laser crystallization is investigated to fabricate antenna arrays of several hundred micrometers. Finally, a beam steering device is designed and fabricated. This work paves the way towards large area fabrication of functional metasurfaces in the IR with the ability of rapid prototyping for IR nanophotonic devices.

1. Wuttig et al., *Nature Photonics* **11**,465 (2017)
2. Heßler et al., *Nature Communications* **12**, 924 (2021)

O 35.4 Tue 11:00 P3

Direct Writing of Chiral and Nonlinear Plasmonic Devices — •Aleksei Tsarapkin[1], Victor Deinhart[1], Thorsten Feichtner[2], and Katja Höflich[1] — [1]Ferdinand-Braun-Institut gGmbH, 12489 Berlin, Germany — [2]University of Würzburg, 97074 Würzburg, Germany

The miniaturization of electrical and optical components allowed many technological and economic advancements over the last decades. Devices that permit control over the polarization of light are crucial in telecommunication and quantum optics but are usually realized as bulky optical systems and thus require further miniaturization. Here we aim at designing a uniquely compact converter and detector based on nanostructures. The device consists of a vertically oriented gold double helix coupled to a planar two-wire transmission line. The helix acts as a sensitive antenna for circularly polarized light, while the plasmonic transmission line guides plasmons on-chip. With numeric analysis we show that antisymmetric modes can be excited in both double helix and two-wire waveguide allowing spatial matching. Furthermore, one can match impedances of both components to maximize power transfer by adjusting their sizes. Finally, we developed fabrication protocols: while the helix can be directly written with an electron-induced deposition, the plasmonic waveguide can be cut from single-crystalline gold flake utilizing focused gallium-ion beam milling. We achieved high structuring resolution with both methods, allowing for efficient coupling to transform linear to circular polarization while retaining a device size of just a few microns.

O 35.5 Tue 11:00 P3

Transverse magnetic routing of light emission in hybrid plasmonic-semiconductor nanostructures: Grating period dependence — •Carolin Harkort[1], Lars Klompmaker[1], Alexander N. Poddubny[2], Leonid V. Litvin[3], Ralf Jede[3], Grzegorz Karczewski[4], Sergij Chusnutdinow[4], Tomasz Wojtowicz[5], Dmitri R Yakovlev[1,2], Manfred Bayer[1,2], and Ilya A. Akimov[1,2] — [1]Experimentelle Physik 2, Technische Universität Dortmund, 44221 Dortmund, Germany — [2]St. Petersburg — [3]Raith GmbH, Konrad-Adenauer-Allee 8, 44263 Dortmund, Germany — [4]Institute of Physics, Polish Academy of Sciences, PL-02668 Warsaw, Poland — [5]International Research Centre MagTop, PL-02668 Warsaw, Poland

We use plasmonic gratings to achieve transverse magnetic routing of the light emission of a nearby quantum well exciton via an external magnetic field. In a hybrid plasmonic-semiconductor (Cd,Mn)Te/(Cd,Mg)Te quantum well (QW) structure the effect of the plasmonic grating period on the directional emission spectra is measured using a Fourier imaging setup to obtain angular and energy resolved photoluminescence spectra for opposite transverse magnetic fields. We achieve a strong directionality of the QW emission of up to 15% at low temperature of about 5 K and magnetic fields of 500 mT with a grating period of 240 nm and demonstrate the effect of the changing SPP dispersion on the directional emission characteristics.

O 35.6 Tue 11:00 P3

Mapping Lamb, Stark and Purcell effects at a chromophore-picocavity junction with hyper-resolved fluorescence microscopy — •Anna Rosławska[1], Tomáš Neuman[1,2,3], Benjamin Doppagne[1], Andrei G. Borisov[3], Michelangelo Romeo[1], Fabrice Scheurer[1], Javier Aizpurua[2], and Guillaume Schull[1] — [1]Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France — [2]Center for Materials Physics (CSIC-UPV/EHU) and DIPC, Paseo Manuel de Lardizabal 5, Donostia - San Sebastián 20018, Spain — [3]Institut des Sciences Moléculaires d'Orsay (ISMO), UMR 8214, CNRS, Université Paris-Saclay, 91405 Orsay Cedex, France

Light-matter interaction plays a crucial role in the properties of light emission from single molecules. Here, we show that it can be probed with sub-molecular precision thanks to the atomically-confined electromagnetic field at the scanning tunneling microscope tip apex, which acts as a picocavity for localized plasmons. Such strong fields interact with the molecular exciton via Purcell, Lamb and Stark effects, which enable tuning the emission energy and line width. Hyper-resolved fluorescence maps of these two parameters can be understood as images of the static charge redistribution upon electronic excitation of the molecule, and the distribution of the dynamical charge oscillation associated with the molecular exciton, respectively [1].

[1] A. Rosławska *et al.*, Phys. Rev. X, 12, 011012, 2022.

O 35.7 Tue 11:00 P3

Steps towards fluorescence detected two-dimensional electronic spectroscopy of a single molecule — •Sanchayeeta Jana and Markus Lippitz — Chair for Experimental Physics III, University of Bayreuth, Bayreuth, Germany

Two-dimensional electronic spectroscopy (2DES) is an ultrafast spectroscopic technique which gives information about the coupling between molecular energy levels. As spectroscopy of a single molecule by fluorescence detection is a well established technique, we want to explore whether fluorescence-detected 2DES is possible at very low concentrations, ideally even on a single molecule. We use a sequence of four phase-modulated pulses to excite the molecules and collect the fluorescence by a high NA objective. The signal amplitude is demodulated at several mixing frequencies to extract the rephasing and non-rephasing contributions.

In this work, we study the 2D spectra measured from an ensemble of molecules in solution and discuss the challenges to experimentally measure 2D spectra from a single molecule.

O 35.8 Tue 11:00 P3

Coupling single epitaxial quantum dots to plasmonic waveguides — •Michael Seidel[1], Yuhui Yang[2], Saimon Covre da Silva[3], Thorsten Schumacher[1], Armando Rastelli[3], Stephan Reitzenstein[2], and Markus Lippitz[1] — [1]Experimental Physics III, University of Bayreuth, Germany — [2]Institute of Solid State Physics, TU Berlin, Germany — [3]Institute of Semiconductor and Solid State Physics, Johannes Kepler University Linz, Austria

Integrated plasmonic nanocircuits are highly promising building blocks for future quantum optical applications. In combination with self-assembled epitaxially grown GaAs quantum dots as stable, bright and narrow-band single-photon sources, ultra-compact nanocircuits operating below the diffraction limit can be designed [1]. A crucial aspect is the coupling of the quantum dot emission into plasmonic waveguide modes. In this work, we demonstrate the coupling of a single near-surface GaAs quantum dot to a silver nanowire by introducing a 100nm thick dielectric spacer layer. We characterize the nanostructure by comparing different imaging methods involving low-temperature cathodoluminescence and photoluminescence as well as confocal laser reflection mapping. Supported by 3D numerical simulations, we find that resonant plasmonic wires can enhance the waveguide coupling efficiency.

[1] Wu et al., Nano Lett. 2017, 17, 7, 4291-4296

O 36: Overview Talk Hendrik Bluhm

Time: Wednesday 9:30–10:15 Location: S054

Invited Talk O 36.1 Wed 9:30 S054

Heterogeneous chemistry of liquid-vapor interfaces investigated with X-ray photoelectron spectroscopy — •Hendrik Bluhm — Fritz Haber Institute of the Max Planck Society, Berlin, Germany

Liquid-vapor interfaces drive numerous important processes in the environment and atmosphere, such as the sequestration of CO2 by the oceans and the uptake and release of trace gases by aerosol droplets. X-ray photoelectron spectroscopy is an excellent method for the investigation of liquid-vapor interfaces due to its interface sensitivity and chemical specificity. In this talk we will discuss the general approach in XPS experiments on liquid interfaces and present illustrative examples of the investigation of the heterogeneous chemistry of surfactants and ions at the interface of aqueous solutions.

O 37: Plasmonics and Nanooptics 1

Time: Wednesday 10:30–12:45 Location: H3

Topical Talk O 37.1 Wed 10:30 H3

Merging integrated photonics with free-electron beams — •Armin Feist — Max Planck Institute for Multidisciplinary Sciences, 37077 Göttingen, Germany

The coherent coupling of electrons and light is at the heart of free-electron quantum optics [1]. However, fully exploring its capabilities requires supreme control over the electron beam and optical quantum states involved. This suggests combining integrated photonics with electron microscopy.

In this talk, I will briefly introduce the basic principles and selected applications of inelastic electron-light scattering in an ultrafast transmission electron microscope (UTEM) [2,3], including the nanoscale imaging of confined optical modes and shaping of electron beams in space and time [4].

Recent progress in coupling single electrons to high-Q integrated photonic microresonators will be discussed, enabling highly efficient continuous-wave optical phase modulation of electron beams and nanoscale-μeV spectroscopy of a photonic mode [5]. Furthermore, spontaneous scattering at the empty resonator modes creates electron-photon pair states [6], opening a pathway towards a new class of heralded single-electron or Fock-state photon sources.

[1] K. Wang *et al.*, Optics & Photonics News **31**, 35 (2020). [2] Barwick *et al.*, Nature **462**, 902 (2009). [3] A. Feist *et al.*, Nature **521**, 200 (2015). [4] F.J. García de Abajo & V. Di Giulio, ACS Photonics **8**, 945 (2021). [5] J.W. Henke *et al.*, Nature **600**, 653 (2021). [6] A. Feist *et al.*, arXiv:2202.12821 (2022).

O 37.2 Wed 11:00 H3

Near-field spectroscopic predictions of low-temperature, gate-tunable plasmon-phonon coupling in the $LaAlO_3/SrTiO_3$ two-dimensional electron gas — •Julian Barnett[1], Yigong Luan[1], Felix Gunkel[2], and Thomas Taubner[1] — [1]I. Institute of Physics (IA), RWTH Aachen University — [2]Peter Grünberg Institute, Forschungszentrum Jülich

Heterointerfaces of $SrTiO_3$ and $LaAlO_3$ give rise to a buried two-dimensional electron gas (2DEG) between polar insulators, that is very sensitive to local defects but difficult to characterize on the nanoscale. It was recently shown that scanning near-field optical microscopy (SNOM) enables the extraction of local 2DEG properties with nanoscale lateral resolution by using phonon-enhanced near-field spectroscopy. Interestingly, the 2DEG mobility increases strongly at low temperatures and the charge carrier concentration can be tuned via gating, allowing control over the electronic properties.

Here, we predict that the plasmon-phonon-coupled near-field response will undergo a transition from a phonon-dominated regime to a plasmon-dominated regime with rising 2DEG mobility, which translates to a fundamentally different near-field spectrum. Additionally, we show that the plasmon-dominated regime could allow for the extraction of the 2DEG depth distribution with nanoscale lateral resolution, potentially enabling its mapping around defects. These insights can be directly transferred to the spectroscopic near-field investigation of 2DEGs, surface accumulation layers, and topologically protected surface states in a variety of bulk materials and heterostructures.

O 37.3 Wed 11:15 H3

Generation of Rotating Fields via Archimedean Spirals —

•Esra Ilke Albar, Heiko Appel, and Franco Bonafe — Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science,22761 Hamburg, Germany

Twisted light (optical vortex), has the distinctive feature that it carries orbital angular momentum. This special type of electromagnetic field promises exciting opportunities for the interaction with matter, and it is expected to enhance spectroscopic techniques. Twisted light already has a vast potential of applications ranging from astronomy and optical tweezers to spintronics. Given this broad spectrum of applications and exciting prospects, it becomes worthwhile to investigate new and efficient methods to generate optical vortices. In this sense, metallic Archimedean spirals in the micro- and nanoscale are good candidates to produce such vortices. We design Archimedean spirals and test their performance in terms of generating field vortices. We perform numerical simulations with the Octopus code which employs the Riemann-Silberstein representation to propagate Maxwell's equations in real-time. Circularly-polarized light is passed through the designed structure, which is modeled both as a non-dispersive linear medium, and as Drude medium. We found that the out-coming field's angular momentum is altered by the structure. By using two different material models the effect of the materials' optical properties and the sole geometrical factors on the angular momentum outcome could be distinguished.

O 37.4 Wed 11:30 H3

Theory of radial oscillations in metal nanoparticles driven by optically induced electron density gradients — •Robert Salzwedel[1], Andreas Knorr[1], Dominik Höing[2,3], Holger Lange[2,3], and Malte Selig[1] — [1]Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, 10623 Berlin, Germany — [2]Institut für Physikalische Chemie, Universität Hamburg, 20146 Hamburg, Germany — [3]The Hamburg Centre for Ultrafast Imaging, 22761 Hamburg, Germany

Upon optical excitation, metal nanoparticles oscillate in radial breathing modes. These oscillations are assumed to be driven mainly by the thermalization of hot electrons impulsively heating the lattice, which can be described classically [1,2]. Based on a combined approach of quantum hydrodynamics and Heisenberg equations of motion for the optical excitation of electron gas in metal nanoparticles and the associated electron-phonon interaction, we discuss the contribution of additional coherent sources to the radial breathing oscillations.

Our results reveal a more direct coupling mechanism between the field and phonons compared to the established interpretation of experiments: the optical pulse induces spatial gradients in the electron density that drive phonon oscillations coherently and directly on the time scale of the optical excitation. Therefore, thermal and coherent contributions must be considered in the early times of the oscillation.

[1] Hartland, G. V. et al., *JCP*, **116**, 8048 (2002)
[2] Hodak, J. H. et al., *JCP*, **111**, 8613 (1999)

O 37.5 Wed 11:45 H3

Near-field imaging of hyperbolic shear polaritons in gallium oxide — •Sören Wasserroth[1], Joseph R. Matson[2], Xiang Ni[3], Giulia Carini[1], Katja Diaz-Granados[2], Maximilan Obst[4], Enrico Maria Renzi[3], Emanuelle Galifi[3], Susanne Kehr[4], Lukas M. Eng[4], Martin Wolf[1], Thomas G. Folland[5], Andrea Alu[3], Joshua D. Caldwell[2], and Alexander Paarmann[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Vanderbilt University, Nashville, USA — [3]City University of New York, New York, USA — [4]Technische Universität Dresden, Dresden, Germany — [5]University of Iowa, Iowa, USA

Controlling the propagation direction and other properties of light in a material and at an interface is a very active field in current research. Strong anisotropy in crystals can lead to a hyperbolic dispersion featuring coupled light-matter interactions known as polaritons with highly directional propagation. Recently [1], it was shown that the additional anisotropy in monoclinic crystals, such as beta phase gallium oxide (bGO), gives rise to tilted wave fronts and asymmetric responses, called hyperbolic shear polaritons (HShPs).
Here, we will show mid-infrared free electron laser based near-field imaging of HShPs in bGO. Gold discs are used as local emitters on the bGO substrate. By changing the IR wavelength we observe the rotation and asymmetry of the HShPs. We compare the obtained images to simulated near-field contributions.

[1] Passler et al., Nature **602**, 595 (2022)

O 37.6 Wed 12:00 H3

Long-wave infrared super-resolution wide-field microscopy using sum-frequency generation — •Richarda Niemann[1], Sören Wasserroth[1], Guanyu Lu[2], Christopher R. Gubbin[3], Simone De Liberato[3], Joshua D. Caldwell[2], Martin Wolf[1], and Alexander Paarmann[1] — [1]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [2]Vanderbilt University, Nashville, USA — [3]University of Southampton, UK

We present our approach in infrared-visible (IR-VIS) sum-frequency generation (SFG) microscopy as a new tool for super-resolution imaging in the IR range.[1] Here, the imaging contrast in the microscope emerges from the IR resonances while the spatial resolution is provided by the SFG wavelength, i.e., well below the IR diffraction limit. As a proof of concept, we study localized phonon polaritons in subdiffractional nanostructures made of Silicon Carbide.[2,3] A free electron laser as IR light source offers high-power laser pulses with tunable wavelengths.[4] We are able to resolve phonon polariton modes in individual sub-diffractional nanostructures and achieve a spatial resolution of $\sim$1.4 μm corresponding to $\lambda_{IR}/9$.[1] Full spectral mapping over the whole SiC Reststrahlenband allows the spectroscopic identification of the polariton resonances, while the high spatial resolution reveals the microscopic origin of the SFG emission.

[1] R. Niemann et al., Appl. Phys. Lett. 120, 131102 (2022)
[2] J.D. Caldwell et al., Nanophotonics 4, 1 (2015)
[3] I. Razdolski et al., Nano Letters 16, 6954 (2016)
[4] W. Schöllkopf et al., Proc SPIE 9512, 95121L (2015)

O 37.7 Wed 12:15 H3

Observation of Hyperbolic Shear Polaritons in Monoclinic Crystals — •Giulia Carini[1], Nikolai C. Passler[1], Xiang Ni[2], Guangwei Hu[2,3], Joseph R. Matson[4], Martin Wolf[1], Mathias Schubert[5], Andrea Alù[2], Joshua D. Caldwell[4], Thomas G. Folland[6], and Alexander Paarmann[1] — [1]FHI, Berlin, Germany — [2]CUNY, NY, USA — [3]NUS, Singapore, Singapore — [4]Vanderbilt University, Nashville, TN,USA — [5]University of Nebraska, Lincoln, NE, USA — [6]University of Iowa, Iowa City, IA, USA

Surface phonon polaritons, light-matter coupled waves, have recently attracted much attention due to their versatility in nanophotonic applications in the infrared. In particular, highly anisotropic materials have been shown to support hyperbolic polaritons, which enable a deep sub-wavelength confinement of light [1,2].

In our contribution, we investigate the emergence of a new class of hyperbolic surface waves, that we call hyperbolic shear polaritons (HShPs), in the monoclinic crystal β-gallium oxide (bGO) [3]. The symmetry breaking in the in-plane hyperbolic propagation is directly linked to the frequency dependence of the optical axis directions within the monoclinic plane, resulting in shear phenomena in the non-diagonalizable dielectric permittivity tensor. We experimentally demonstrate the hyperbolic in-plane dispersion of these new polariton modes by means of Otto-type prism coupling.

[1] J. D. Caldwell, et al., Nat. Commun. 5, 5221 (2014).
[2] W. Ma, et al., Nature 562, 557-562 (2018).
[3] N. C. Passler, et al., Nature 602, 595-600 (2022).

O 37.8 Wed 12:30 H3

Transverse magnetic routing of light emission in hybrid plasmonic-semiconductor nanostructures: Towards operation at room temperature — •Lars Klompmaker[1], Alexander N. Poddubny[2], Eyüp Yalcin[1], Leonid V. Litvin[3], Ralf Jede[3], Grzegorz Karczewski[4], Sergij Chusnutdinow[4], Tomasz Wojtowicz[5], Dmitri R. Yakovlev[1,2], Manfred Bayer[1,2], and Ilya A. Akimov[1,2] — [1]Experimentelle Physik 2, Technische Universität Dortmund, 44221 Dortmund, Germany — [2]St. Petersburg — [3]Raith GmbH, Konrad-Adenauer-Allee 8, 44263 Dortmund, Germany — [4]Institute of Physics, Polish Academy of Sciences, PL-02668 Warsaw, Poland — [5]International Research Centre MagTop, PL-02668 Warsaw, Poland

We studied the transverse magnetic routing of light emission from a hybrid plasmonic-semiconductor quantum well (QW) structure where the exciton emission from the QW is routed into surface plasmon polaritons propagating along a nearby semiconductor-metal interface. In our diluted magnetic semiconductor (Cd,Mn)Te/(Cd,Mg)Te QW structure the magnitude of routing is governed by the circular polarization of the exciton optical transitions, induced by the magnetic field. A strong directionality of emission of 15 % was measured at low temperatures of 20 K, but for increasing temperatures it decreases to 4 % at about 65 K due to the decreasing magnetic susceptibility. To avoid this strong temperature dependence we also suggest an alternative design based on a non-magnetic (In,Ga)As/(In,Al)As QW structure.

O 38: Solid-Liquid Interfaces 4: Reactions and Electrochemistry

Time: Wednesday 10:30–12:30 Location: H4

Topical Talk O 38.1 Wed 10:30 H4
Electrochemical Microcalorimetry — •Rolf Schuster and Marco Schönig — Institute of Physical Chemistry, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany

We investigate electrochemical reactions at single electrodes by measuring the accompanying heat changes. The heat reversibly exchanged during an electrode process is directly correlated to the reaction entropy of the half-cell reaction including all side reactions, e.g., ordering processes of the solvent or coadsorption processes of anions. Thus, measuring the heat exchange during an electrochemical process provides independent information on the ongoing reaction, which is complementary, e.g., to the potential-current relation, usually measured by cyclic voltammetry or impedance spectroscopy.

In this contribution we will briefly discuss some theoretical aspects of electrochemical microcalorimetry of single electrodes and present our strategy to measure the heat evolution upon surface electrochemical processes. With our setup we are sensitive to heat effects originating from minute conversions of a few percent of a monolayer of an electroactive species.

We will present examples for entropy changes upon anion adsorption and double layer charging on Au(111) and discuss the effect of configurational entropy of the adlayer. The hydrogen adsorption on Pt-films deals as an example of a prototypical surface electrochemical reaction. Time-resolved studies of the heat evolution during Cu bulk deposition will demonstrate the implications from heat measurements on the subsequent reaction steps of this complex reaction.

O 38.2 Wed 11:00 H4
Direct Assessment of the Proton Affinity of Individual Surface Hydroxyls with AFM and DFT — •Bernd Meyer[1], Margareta Wagner[2], Martin Setvin[2], Michael Schmid[2], and Ulrike Diebold[2] — [1]Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany — [2]Institute of Applied Physics, TU Wien, Austria

The state of protonation/deprotonation of individual surface sites has far-ranging implications in all areas of chemistry. However, common experimental measurements of surface acidity are integral techniques which give only average quantities integrated over the whole surface. Here we show that an OH-functionalized tip of an atomic force microscope (AFM) can be used for quantitative insights into the acidity of individual surface OH groups [1]. The chosen model oxide, $In_2O_3(111)$, offers four types of surface O atoms with distinct properties, each giving rise to a characteristic force–distance curve after protonation. Density-functional theory (DFT) calculations demonstrate a linear correlation between the force minimum and the proton affinity of the surface hydroxyls. By benchmarking the calculations to known proton affinities and $\mathrm{p}K_\mathrm{a}$ values of gas-phase molecules, the force minima provide a direct measure of proton affinity distributions and $\mathrm{p}K_\mathrm{a}$ differences at the atomic scale.

[1] M. Wagner, B. Meyer, M. Setvin, M. Schmid, U. Diebold,
Nature **592** (2021) 722–725

O 38.3 Wed 11:15 H4
Revisiting the OH adsorption on Pt(111) in static water environments — •Alexandra C. Dávila López, Nicolas G. Hörmann, Thorben Eggert, and Karsten Reuter — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

The partial solvation of adsorbates typically leads to altered adsorption energies at solid-liquid interfaces as compared to vacuum. While such solvation effects can be treated most accurately based on *ab initio* molecular dynamics simulations, an according description is hardly feasible for a large number of systems e.g. across different substrates, adsorbates and adsorbate coverages due to prohibitive computational costs. As a result, many studies rely on an approximate treatment based on static water environments. In this work, we study solvation effects based on our previously introduced method [1] to generate a wide range of different static water adlayers. In particular, we analyze the adsorption energy of solvated OH on Pt(111) for a variety of explicit solvation environments and OH configurations. The results are benchmarked against available theoretical and experimental literature data [2, 3], and they highlight prevailing uncertainties in the description of solvation effects.

[1] A. C. Dávila *et al.*, J. Chem. Phys. **155**, 194702 (2021).
[2] V. Tripković *et al.*, Electrochim. Acta **55**, 7975*7981 (2010).
[3] H. H. Kristoffersen *et al.*, Chem. Sci., **9**, 6912-6921 (2018).

O 38.4 Wed 11:30 H4
Measuring the reaction volume of an electrochemical surface process: Cu underpotential deposition on Au(111) — •Lisa Hirsch, Bianca Krumm, Tamara Meyer, and Rolf Schuster — Karlsruhe Institute of Technology

Information about the reaction volume of electrochemical surface reactions is rather scarce [1-3] albeit this thermodynamic quantity would allow conclusions on solvent contribution and possible side processes. With a specifically designed electrochemical cell, we determined the variation of the cell potential of a (111)-textured Au-film in $CuSO_4$ / H_2SO_4 vs a Cu reference electrode, while applying pressure pulses of moderate amplitude (< 10 bar) and durations of several seconds, starting at different rest potentials. According to the pressure dependend potential variation, we calculated the reaction volume $\Delta_R\mathrm{V}$ of the processes at the working electrode. For rest potentials in the Cu UPD region, $\Delta_R\mathrm{V}$ varies only slightly (± $8\mathrm{cm}^3/\mathrm{mol}$) around $\Delta_R\mathrm{V} = 18\mathrm{cm}^3/\mathrm{mol}$, its value for Cu bulk deposition, moderately peaking upon formation of the $\left(\sqrt{3}\times\sqrt{3}\right)$ Cu UPD structure. Positive of the Cu UPD, in the sulfate adsorption region, $\Delta_R\mathrm{V}$ drops to smaller values. We check the integrity of the measured potential variations by their dependence on the pressure amplitude as well as by measuring the symmetrical $\mathrm{Cu}|\mathrm{Cu}^{2+},\mathrm{SO}_4^{2-}|\mathrm{Cu}$ cells.

[1] Conway, B. E., Currie, J. C., J. Chem. Soc., 74, 1978, 1390-1402. [2] Loewe, T., Baltruschat, H., Phys. Chem. Chem. Phys., 7, 2005, 379-384. [3] Heusler, K. E., Gaiser, L., Ber. Bunsenges. Phys. Chem., 73, 1969, 1059-1068.

O 38.5 Wed 11:45 H4
Enter the Void: Cavity Formation at Metal-Water Interfaces — •Thorben Eggert[1,2], Nicolas G. Hörmann[1], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Technical University of Munich, Munich, Germany

Cavity formation is an important concept when rationalizing the solvation of ions. However, most studies analyze cavities only in bulk liquids, omitting that their properties may change dramatically at solid-liquid interfaces.

Here, we study cavities at interfaces, particularly their free energy of formation based on molecular dynamics simulations. Specifically, we use a particle insertion approach, as well as the Multistate Bennett Acceptance Ratio method. We demonstrate that cavity formation at interfaces is dependent on the substrate material, which can be partially rationalized by the substrate-specific interfacial water structure. Furthermore, we observe stabilized cavities behind the first solvation layer of water.

These results might on the one hand rationalize recent theoretical suggestions of a non-electrostatic, attractive force on ions near interfaces, and on the other hand enable the improvement of implicit solvation models, which typically neglect substrate-specificity in their description.

O 38.6 Wed 12:00 H4
Impact of confined water on solvation and adsorption/desorption energetics of charged ions at the electrified interface — •Zhenyu Wang, Mira Todorova, and Jörg Neugebauer — Department of Computational Materials Design, Max-Planck-Institut für Eisenforschung, Max-Planck-Str.-1, D-40237, Düsseldorf, Germany

Understanding processes at electrified solid/liquid interfaces is crucial for many systems and a wide range of applications in electrochemical industry, catalytic sciences and biological engineering. Using a prototypical model system of a single ion in water confined between two charged electrodes, we perform nanosecond-scale atomistic molecular dynamics simulations to study the dielectric behavior of chemically pure water as well as the solvation of ions in the presence of an electric field. For weak electric fields we find that the screening charge density of water is proportional to the external electric intensity, in agreement with classical polarization theory. Probing the interface structure by the single Na+/Cl- ion we investigate the formation and evolution of the ion's solvation shell as function of the electrode-ion distance. Comparing potential profiles from Na+/Cl- calculations for different charge states and positions, we elucidate the role of screening and solvation shell size on reorganization energies and transmission barrier of the ions close to the interface.

O 38.7 Wed 12:15 H4

Are organic solvents key to enable CO_2 electro-reduction on Mo_2C as promised in theory? — •Thomas Mairegger[1], Christoph Griesser[1], Haobo Li[2], Nicolas Hörmann[3], Karsten Reuter[3], and Julia Kunze-Liebhäuser[1] — [1]Department of Physical Chemistry, Innsbruck, Austria — [2]University of Adelaide, Adelaide, Australia — [3]Fritz Haber Institute, Berlin, Germany

It has been proposed in active-site computational screening studies that Mo_2C is an effective electrocatalyst for the electrochemical CO_2 reduction reaction (CO_2RR) to valuable fuels, such as hydrocarbons and alcohols. However, the competing hydrogen evolution reaction (HER) has been found to exclusively take place.[1] Reason for this is the formation of a surface oxide film upon air exposure or immersion of Mo_2C into aqueous electrolytes that impedes the formation of the desired higher reduction products.[1]

Here we investigate the CO_2RR activity of polycrystalline hexagonal Mo_2C in non-aqueous electrolyte to avoid passivation of the electrode and circumvent the high HER activity. We show that Mo_2C is capable of reducing CO_2 in reasonable amounts in an acetonitrile electrolyte, with an onset at -1.08 V_{SHE}.[1] The nature of the products, among them gaseous CO, depends on the concentration of water in the electrolyte. Furthermore, we show that the acetonitrile has a stronger impact on the CO_2 electro-reduction than previously believed.

[1] Griesser, C., et. al., ACS Catalysis 11, 4920-4928 (2021).

O 39: Tribology

Time: Wednesday 10:30–12:15 Location: H6

O 39.1 Wed 10:30 H6

Automated calculation of surface energies of arbitrary crystals — •Firat Yalcin and Michael Wolloch — Computational Materials Physics, Faculty of Physics, University of Vienna, Kolingasse 14-16, 1090 Vienna

The surface energy is one of the most fundamental properties of the surface facets of a crystal, determining the stability, equilibrium shape, catalytic activity, and other phenomena like surface segregation and roughening. In this work, we present a high-throughput workflow to calculate the surface energies of arbitrary crystals, accounting for all possible Miller indices and different terminations, using density functional theory (DFT). By employing available open-source libraries and custom-made tools, every step from querying bulk structures from various databases to the generation of slabs with precise geometries and subsequent DFT calculations is performed automatically and with minimal user input. Surface energies, Wulff shapes, as well as all in- and outputs, are automatically saved into an easily accessible, OPTIMADE compliant database. Example results are presented for some non-trivial surfaces with complex geometries to showcase the capabilities and validate our approach.

O 39.2 Wed 10:45 H6

High-throughput calculations of heterogeneous interfaces for tribology — •Michael Wolloch[1,2], Gabriele Losi[3], Omar Chehaimi[3], Firat Yalcin[1], Mauro Ferrario[4], and M. Clelia Righi[3] — [1]CMP, University of Vienna, Vienna, Austria — [2]VASP software GmbH, Vienna, Austria — [3]DIFA, University of Bologna, Bologna, Italy — [4]FIM, UNIMORE, Modena, Italy

We have shown in the past that fundamental properties of sliding interfaces like adhesion, friction, and ultimate strength, are closely connected to one another and the re-arrangement of charge at the interface [1-3]. We extended the previously presented approach for homogeneous interfaces to nearly arbitrary interfaces of crystalline surfaces, allowing us to treat heterogeneous interfaces of multi component systems [4].

In this talk we will present the fault tolerant and fully automatic workflow we developed, as well as initial results of the ongoing high-throughput studies we are conducting.

Part of this work was supported by ERC grant 865633 (SLIDE); [1] Wolloch et al. Sci. Rep. 9, 17062 (2019), [2] Restuccia et al. Comput. Mater. Sci., 154:517-529 (2018), [3] Wolloch et al. PRL 121, 026804 (2018) [4] Wolloch et al. Comput. Mater. Sci., 207:111302 (2022)

O 39.3 Wed 11:00 H6

The Influence of Temperature and Wear on Nanoscale Friction Anisotropy — •Jennifer Konrad, Dirk Dietzel, and Andre Schirmeisen — Institute of Applied Physics, Justus-Liebig University Giessen, 35392 Giessen, Germany

On atomically flat surfaces, the nanoscale friction force measured by atomic force microscopy shows a distinct variations dependent on the sliding direction of the AFM-tip. This anisotropy occurs as a consequence of the surface structure and is related to different energy barrier heights along different directions of the sample surface. In this work, this anisotropy of friction force is now analyzed as a function of temperature under UHV conditions on hexagonal latices like HOPG or MoS2 and also on ionic crystals with 90 degree symmetry like NaCl. At low sample temperatures, the friction force as deduced from the thermally activated Prandtl Tomlinson Model increases, and thermally activated random jumps become more unlikely, which direct influences the stability of the different sliding directions. This can be confirmed for HOPG and MoS2, while, our results for the ionic crystals additionally show that the both the absolute friction and the anisotropy are not only influenced by temperature, but also reflect temperature dependent wear effects.

O 39.4 Wed 11:15 H6

Dissipative frictional mechanism over Moiré superstructure — •Yiming Song[1], Xiang Gao[2], Antoine Hinaut[1], Sebastian Scherb[1], Shuyu Huang[1], Oded Hod[2], Michael Urbakh[2], and Ernst Meyer[1] — [1]Department of Physics, University of Basel, Basel, Switzerland — [2]Department of Physical Chemistry, School of Chemistry, The Raymond and Beverly Sackler Faculty of Exact Sciences and The Sackler Center for Computational Molecular and Materials Science, Tel Aviv University, Tel Aviv, Israel.

Friction force microscopy experiments performed on graphene-coated platinum surfaces that exhibit a variety of moiré superstructures demonstrate that, in addition to the well-known atomic stick-slip dynamics, a new dominant energy dissipation mechanism emerges. The observed velocity dependence of friction displays two distinct regimes: (i) at low velocities, the friction force is small and nearly constant; whereas (ii) above some threshold, friction increases logarithmically with velocity. The threshold velocity, separating the two frictional regimes, decreases with increasing normal load and moiré superstructure period. Atomistic molecular dynamics simulations demonstrate that the main contribution to frictional energy dissipation results from the elastic deformation and subsequent relaxation of moiré ridges caused by the pushing action of the tip as it slides over the superstructure. Based on the simulation results we develop a semi-phenomological model, which makes it possible to calculate friction under measurement conditions and provides excellent agreement with the experimental observations.

O 39.5 Wed 11:30 H6

Investigating thermal and directional motion in molecular friction processes by photonic force microscopy — •Subhrokoli Ghosh and Alexander Rohrbach — Lab for Bio- and Nano-Photonics, Department of Microsystems Engineering (IMTEK), University of Freiburg, Georges-Koehler-Allee 102, 79110 Freiburg, Germany

Friction of a moving particle is a complex process of energy dissipation to the environment, which is important on most length scales, time scales and across disciplines. Several theories approach the molecular origin of friction, but a comprehensive understanding is still missing. Usually, the relation between dynamic friction and velocity is quantified by a coefficient, which depends on various parameters. Two main routes to determine the friction coefficient can be addressed by either from a directed particle motion or from its thermal motion. In both cases, Photonic Force Microscopy (PFM) has proven to be one of the most elegant techniques that can be utilized to better understand friction on mesoscopic length scales, specially at soft (-bio) interfaces. Towards this aim, we employ PFM with high-frequency axial tracking for directed and frequency dependent measurements of different bead-surface model systems, starting from simple poly-styrene (PS) bead-glass surface. From Brownian dynamics simulations, in combination with experiments, we obtain better insights on the friction coefficient and its dependency on different system parameters. From this, we developed a theoretical model describing the microscopic origin of friction through molecular on- and off-binding processes.

O 39.6 Wed 11:45 H6

Single Asperity Sliding Friction across the Superconducting Phase Transition — Wen Wang, •Dirk Dietzel, and Andrè Schirmeisen — Institute of Applied Physics, Justus Liebig University Giessen, Giessen, Germany

When analyzing sliding friction, it is usually an intriguing question to identify the different dissipation mechanisms contributing to friction. Usually, a number of different channels for dissipation are considered including phonon and electron systems, plastic deformation, and crack formation. Among these, especially the role of the electron system for energy dissipation often remains elusive. In this contribution, we now present experiments single asperity sliding friction monitored during AFM-measurements on a high-T_c BSCCO-superconductor. These measurements reveal a distinct temperature dependence of friction in a temperature range between 40 K and 300 K [1]. While the overall friction decreases with temperature as predicted by models about thermally activated friction, we find a distinct friction peak at about 95K. This peak can be explained by a superposition of different energy dissipation channels, where the influence of electronic contributions vanishes when cooling below the superconducting phase transition T_c. Our experiments thereby unambiguously link electronic friction effects to the number of normal state electrons in the superconducting phase below T_c. In addition, we analyze single asperity friction of BSCCO in the proximity of step-edges, where layer-defects reveal the potential influence of the topmost-layer on electronic friction contributions. [1] Wang, Dietzel, Schirmeisen, Science Advances, eaay0165 (2020)

O 39.7 Wed 12:00 H6

Fingerprint of a structural phase transition during superlubric sliding — •Ebru Cihan[1,2], Dirk Dietzel[1], and Andre Schirmeisen[1] — [1]Institute of Applied Physics, Justus-Liebig University Giessen, 35392 Giessen — [2]Institute for Materials Science, TU Dresden, 01062 Dresden

Although the fundamental concept of structural superlubricity (i.e. ultra-low friction observed between clean and atomically flat, incommensurate surfaces) is very straightforward, the effective energy barrier for lateral motion still depends on the exact structural dynamics at the sliding interface. In fact, it can be computationally predicted that the superlubricity of amorphous structures is less effective than that of crystalline structures, however this is not always easy to demonstrate experimentally. But we have now overcome this challenge by measuring the friction of antimony nanoparticles on highly oriented pyrolythic graphite in the high temperature regime, i.e. between 300 K and 750 K, where the interface can be restructured. At about 450 K, we trigger a phase transition in antimony nanoparticles, which also allows us to establish a direct link between friction and the interface structure. More specifically, our experiments reveal that the friction level decreases in the more crystalline state where the collective force cancellations are more effective. Due to the irreversible character of the phase transition, a reduced friction level can then also be observed after cooling back to room temperature. The reduction of friction can be associated with a decrease of the characteristic scaling factor of about 16%, as theoretically anticipated from the 'scaling law' for superlubricity.

O 40: Organic Molecules at Surfaces 4: Chemistry on Surfaces

Time: Wednesday 10:30–12:45 Location: S051

O 40.1 Wed 10:30 S051

On-surface synthesis of planar π-extended cycloparaphenylenes featuring an all-armchair edge topology — Feifei Xiang[1], Sven Maisel[2], Sumit Beniwal[1], Vladimir Akhmetov[2,3], Cordula Ruppenstein[3], Mirunalini Devarajulu[1], Andreas Dörr[1], Olena Papaianina[3], Andreas Görling[2], Konstantin Amsharov[2,3], and •Sabine Maier[1] — [1]FAU Erlangen-Nürnberg, Dept. of Physik — [2]FAU Erlangen-Nürnberg, Dept. of Chemistry and Pharmacy — [3]Institute for Chemistry, University Halle-Wittenberg

[n]cycloparaphenylenes ([n]CPPs) have attracted significant attention due to their unique cyclic structure and highly effective para-conjugation leading to a myriad of fascinating (opto-)electronic properties. However, their strained topology prevents the π-extension of CPPs to convert them either into armchair nanobelts or planarized CPP macrocycles. We have successfully tackled this long-standing challenge and present the bottom-up synthesis and characterization of atomically precise in-plane π-extended [12]CPP on Au(111) by low-temperature scanning probe microscopy/spectroscopy combined with density functional theory.[1] The planar π-extended CPP represents the first nanographene with an all-armchair edge topology. The exclusive para-conjugation at the periphery yields delocalized electronic states and the planarization maximizes the overlap of p-orbitals, which both reduce the bandgap compared to conventional CPP. Calculations predict ring currents and global aromaticity in the doubly charged system.

[1] F. Xiang et al., Nat. Chem. 2022, doi: 10.1038/s41557-022-00968-3.

O 40.2 Wed 10:45 S051

Electronic Properties of N-Heterotriangulene based Charge Transfer Complexes — •Mohsen Ajdari[1], Ronja Pappenberger[1], Ina Michalsky[2], Leonie Pap[1], Christian Huck[1], Marvin Hoffmann[3], Friedrich Maass[1], Milan Kivala[2], Andreas Dreuw[3], and Petra Tegeder[1] — [1]Physikalisch-Chemisches Institut, Universität Heidelberg — [2]Organisch-Chemisches Institut, Centre for Advanced Materials, Universität Heidelberg — [3]Interdisziplinäres Zentrum für Wissenschaftliches Rechnen, Universität Heidelberg

N-heterotriangulenes (N-HTAs) are a class of organic electron-transporting semiconductors that belong to N-heteropolycyclic aromatic compounds, which are promising candidates for a variety of (opto) electronic applications. In this study, electronic high-resolution electron energy loss spectroscopy (HREELS) in combination with quantum-chemical theory are utilized to investigate the electronic properties of two N-HTA derivatives, N-HTA-550 and N-HTA-557 on Au(111). In addition, formation of charge transfer complexes (CTCs) with N-HTAs, acting as donor molecules in combination with two well-known cyano-based electron acceptor molecules, TCNQ and F4TCNQ is identified. Our findings indicate that formation of the 7-membered ring in N-HTA-557 by adding the -C=C- bridge leads to a narrowing of the optical gap size by 0.9 eV and a decrease in the first triplet state energy by 1.2 eV. Moreover, all donor/acceptor bilayer systems on Au(111) exhibit low-lying electronic transitions between 0.9 and 2.2 eV, which are attributed to the formation of CTCs.

O 40.3 Wed 11:00 S051

N-Heterotriangulenes Donors and Charge Transfer Complexes formed with strong Electron Acceptors investigated with Two-Photon Photoemission Spectroscopy — •Jakob Steidel[1], Ina Michalsky[2], Milan Kivala[2], and Petra Tegeder[1] — [1]Institute of Physical Chemistry, Heidelberg University, Im Neuenheimer Feld 253, 69120 Heidelberg — [2]Institute of Organic Chemistry, Heidelberg University, Im Neuenheimer Feld 270, 69120 Heidelberg

Many opto-electronic devices such as organic photovoltaic cells or organic light emitting diodes utilize donor-acceptor-systems (D-A-systems). Triphenylamine derivatives are promising candidates for donors in D-A-systems.1 The introduction of an etheno-bridge in the planar triphenylamine derivative indolo[3,2,1-jk]carbazole (N-HTA-550) creates a seven membered antiaromatic ring in the resulting N-Heterotriangulene-557 (N-HTA-557). While the molecular geometry is mostly preserved, the electronic structure is strongly modified by this additional -C=C-moiety.

In the present contribution we study the electronic properties of N-HTA-550 and N-HTA-557 on a Au(111) surface using two-photon photoemission (2PPE) spectroscopy and temperature programmed desorption (TPD). Furthermore the formation of charge transfer complexes with strong electron acceptors (TCNQ and F4TCNQ) is investigated utilizing both 2PPE and TPD.

O 40.4 Wed 11:15 S051

Remarkably table metal-organic networks on an inert substrate: Ni-, Fe-, and Mn-TCNQ on graphene — •Zdeněk Jakub[1], Anna Kurowská[1], Ondrej Herich[1], Lenka Černá[1], Lukáš Kormoš[1], Azin Shahsavar[1], Pavel Procházka[1], and Jan Čechal[1,2] — [1]CEITEC, Brno University of Technology, Czechia — [2]Faculty of Mechanical Engineering, Brno University of Technology, Czechia

Potential applications of 2D metal-organic frameworks (MOF) require the frameworks to be monophase and well-defined at the atomic scale, to be decoupled from the supporting substrate, and to remain stable at the application conditions. Here, we present three systems meeting this elusive set of requirements: M-TCNQ (M = Ni, Fe, Mn) on epitaxial graphene/Ir(111). We study the systems experimentally by scanning tunneling microscopy, low energy electron microscopy and x-ray photoelectron spectroscopy. When synthesized on graphene, the 2D M-TCNQ MOFs are monophase with $\mathrm{M_1(TCNQ)_1}$ stoichiometry, and we demonstrate their remarkable chemical and thermal stability: All the studied systems survive exposure to ambient conditions, with Ni-TCNQ doing so without any significant changes to its atomic-scale structure or chemical state. Thermally, the most stable system is Fe-TCNQ which remains stable above 500 °C, while all the tested MOFs survive heating to 250 °C. Overall, the modular M-TCNQ/graphene system combines the atomic-scale definition required for fundamental studies with the robustness needed for applications, thus it presents an ideal model for research in single atom catalysis or spintronics.

O 40.5 Wed 11:30 S051

Control of overlayer-substrate coupling via alkali doping in two-dimensional metal-organic networks — •Billal Sohail[1], Phil J. Blowey[2], Tien-Lin Lee[3], Paul T. P. Ryan[3], David A. Duncan[3], Giovanni Constantini[1], D. Phil Woodruff[1], and Reinhard J. Maurer[1] — [1]University of Warwick — [2]University of Leeds — [3]Diamond Light Source

We characterise a two-dimensional donor-acceptor network formed by coadsorption of alkali atoms and the prototypical acceptor molecule TCNQ (7,7,8,8-tetracyanoquinodimethane) at different orientations of Ag surfaces. We characterise the adsorption structure with a combination of normal incidence x-ray standing wave (NIXSW) measurements, Scanning Tunnelling Microscopy, and dispersion-inclusive Density Functional Theory, which we find to be in excellent agreement with experiment. The adsorption structure sensitively depends on an inter- play of molecule-metal charge transfer and long-range dispersion forces, which are influenced by the co-adsorption ratio between alkali atoms and TCNQ. In general, alkali atom co-adsorption reduces the molecule-substrate interaction strength, yet is energetically favoured compared to TCNQ co-adsorption with Ag adatoms. We show that the donor-acceptor ratio in the network is able to control the overlayer-substrate interaction, which strongly affects electronic properties such as the work function and the level alignment at the interface. Therefore, the concentration of alkali donor atoms can be used to tune electronic properties of the interface.

O 40.6 Wed 11:45 S051

Analysis of the 3-dimensional adsorption configuration of organic molecules by SPM manipulation and imaging — •Alexander Ihle[1], Daniel Ebeling[1], Daniel Kohrs[2], Hermann A. Wegner[2], and Andre Schirmeisen[1] — [1]Institute of Applied Physics, Justus Liebig University Giessen — [2]Institute of Organic Chemistry, Justus Liebig University Giessen

On-surface chemistry is a powerful tool for building covalent molecular structures such as chains, networks, or graphene nanoribbons (GNRs). In particular, the catalytic properties of the metal substrate as well as the 2D confinement facilitate the synthesis of new structures that are not accessible via solution chemistry. The Ullmann type coupling is one of the most frequently applied on-surface reaction for synthesizing C-C bonded structures. However, only limited information is available about the three dimensional adsorption conformation of the molecular educts and how this affects the reaction pathway. Here, we studied on-surface reactions of 9-X-10-(1,1':3',1''-terphenyl-5'-yl)anthracene (X= bromine, iodine) on Cu(111), Ag(111) and Au(111). Using low-temperature atomic force microscopy with CO-functionalized tips, we can identify the precise adsorption conformation of the pristine molecules as well as the intermediate and final products. In particular, the three dimensional conformation of the intermediates strongly inhibits the intermolecular coupling reaction between the educts on all surfaces. Instead, intramolecular bond formation is observed.

O 40.7 Wed 12:00 S051

STM growth studies of 5,14-ol-5,14-diborapentacyclo on low-index coinage metal surfaces — •Wun-Chang Pan[1], Jing Qi[1], Carina Mützel[2], Paula Weber[1], Frank Würthner[2], and Matthias Bode[1] — [1]Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany — [2]Institut für Organische Chemie & Center for Nanosystems Chemistry (CNC), Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

In recent studies [1, 2], heteroatoms-doped precursors have frequently been used to polymerize graphene nanoribbons with a large variety of structures or dopant heteroatoms. Using cryogenic scanning tunneling microscopy, we investigated the structure of self-assembled 5,14-ol-5,14-diborapentacyclo (CM218) on low-index coinage metal surfaces. The main focus of our study is on CM218 on Ag(111), where we find that molecular clusters and chains coexist with molecular islands. At low annealing temperature $T_{ann} < 100°C$, the islands exhibit a rail track-like structure with a rhomboid-shaped unit cell. Besides, we find irregular clusters and molecular chains. At higher $T_{ann} \geq 180°C$, islands with a honeycomb (HC) structure are observed. Topographic images of these HCs display a pronounced bias dependence. Molecule-functionalized tips allow for high-resolution images of these structures for which we suggest structural models.

[1] L. Grill and S. Hecht, Nature Chemistry 12, 115 (2020).
[2] Q. Zhong *et al.*, Nature Chemistry 13, 1133 (2021).

O 40.8 Wed 12:15 S051

Surface chemistry of dibromoindigo - effects of temperature and type of surface — •Manuela Hocke[1,2], Michael Schmittel[3], Wolfgang Heckl[1,2], and Markus Lackinger[1,2] — [1]Deutsches Museum, 80538 München — [2]Technische Universität München, Physics Department, 85748 Garching — [3]Center of Micro and Nanochemistry, Universität Siegen, 57068 Siegen, Germany

Mother Nature offers a great variety of suitable halogenated compounds, which can readily be used as precursor for on-surface Ullmann couplings. Here, we study the thermally activated surface chemistry of the famous dye Tyrian purple (6,6'-dibromoindigo) comparatively on Ag(111) and Au(111) by Scanning-Tunneling-Microscopy. On Au(111) we observed two distinct self-assembled structures comprised of fully intact molecules. Covalent structures were obtained either by deposition onto hot surfaces or subsequent heating, where the heating rate is crucial. On Ag(111) only one self-assembled structure was observed. The organometallic structures obtained upon debromination rarely exhibited the anticipated linear structure. Instead, we find remarkably diverse structures. The additional functionalization of 6,6'-dibromindigo with potent H-bond donor and acceptor groups renders the on-surface polymerization of this compound particularly prone to additional influences of supramolecular bonds with a vivid contribution of the co-adsorbed dissociated bromine atoms. Moreover, the surprisingly large variation of the organometallic structures on Ag(111) and the profound dependence on preparation parameters indicate an unexpectedly large kinetic influence.

O 40.9 Wed 12:30 S051

Interface-driven Assembly of Pentacene/MoS_2 Lateral Heterostructures: A Combined STM and DFT Study — •Sergio Tosoni[1], Francesco Tumino[2], Andi Rabia[2], Andrea Li Bassi[2], and Carlo Casari[2] — [1]Dipartimento di Scienza dei materiali, Università di Milano-Bicocca, via Roberto Cozzi 55, I-20125 Milano, Italy — [2]Dipartimento di Energia, Politecnico di Milano, via G. Ponzio 34/3, Milano, I-20133, Italy

Mixed-dimensional van der Waals heterostructures formed by molecular assemblies and 2D materials provide a novel platform for fundamental nanoscience and future nanoelectronics applications. Here we investigate a hybrid heterostructure between pentacene molecules and 2D MoS_2 nanocrystals deposited on Au(111) by means of Scanning Tunneling Microscopy and Density Functional Theory calculations.

In the MoS_2/Au interface, the lattice mismatch leads to the growth of extended monolayer films displaying a non-commensurate lattice with the metal substrate and typical features of point defects, identified as single sulfur vacancies.

The formation of atomically thin pentacene/MoS_2 lateral heterostructures is observed on the Au substrate. Interestingly, the inner regions of the MoS_2 layer are not covered by pentacene. The density of states changes sharply across the pentacene/MoS_2 interface indicating a weak interfacial coupling, which leaves unaltered the electronic signature of MoS_2 edge states. This work unveils the growth of abrupt lateral heterostructures toward hybrid devices based on organic/inorganic one-dimensional junctions.

O 41: Graphene: Growth, Substrate Interaction, Intercalation, and Doping

Time: Wednesday 10:30–12:30 Location: S052

O 41.1 Wed 10:30 S052

Using polyaromatic hydrocarbons for graphene growth — •Matthew A. Stoodley[1,2], Benedikt P. Klein[1,2], Luke A. Rochford[2], Matthew Edmondson[3], Marc Walker[2], Tien-Lin Lee[1], Alexander Saywell[3], Reinhard J. Maurer[2], and David A. Duncan[1] — [1]Diamond Light Source, Didcot, United Kingdom — [2]University of Warwick, Coventry, United Kingdom — [3]University of Nottingham, Nottingham, United Kingdom

Graphene, due to its widespread potential applications, has been one of the most studied materials this century. The most common approach to produce high quality graphene is through epitaxial growth utilising chemical vapour deposition (CVD) on copper surfaces. This method exploits the catalytic activity of copper to form highly crystalline, large area graphene. Traditionally, precursors used to grow graphene on Cu(111) require high pressures and elevated substrate temperatures, however, many cutting edge characterisation techniques cannot be used at such conditions. In this work, polyaromatic hydrocarbons (PAH) are used to grow high quality graphene on Cu(111) via CVD at relatively low surface temperatures. Furthermore, the low vapour pressure of the PAH permits us to synthesise graphene films in ultra-high vacuum conditions. We present a characterization of the grown graphene, by utilising a wide variety of techniques including Scanning Tunnelling Microscopy, X-ray photoelectron spectroscopy, normal incidence x-ray standing waves, and near edge X-ray adsorption fine structure.

O 41.2 Wed 10:45 S052

Stone-Wales defect: molecular model system reveals increased interaction with Cu(111) surface — •Benedikt P. Klein[1,2,3], Alexander Ihle[4], Stefan R. Kachel[1], Lukas Ruppenthal[1], Samuel J. Hall[2], Daniel Ebeling[4], Ralf Tonner-Zech[1], Reinhard J. Maurer[2], Andre Schirmeisen[4], and J. Michael Gottfried[1] — [1]Philipps-Universität Marburg, Germany — [2]University of Warwick, Coventry, UK — [3]Diamond Light Source, Didcot, UK — [4]Justus-Liebig-Universität Gießen, Germany

The properties of the graphene/metal interface are crucially influenced by the enhanced interaction of defects in the graphene layer with the metal substrate. However, due to experimental and computational constraints, it is difficult to investigate this interaction directly. We combine calculations with experimental analysis of large organic molecules adsorbed on metal surfaces as molecular model systems. We chose two model molecules, azupyrene and pyrene, which have the same aromatic topology as the prototypical Stone-Wales defect and the ideal graphene lattice, respectively. When adsorbed on the Cu(111) surface, we could show using TPD that the model defect binds much stronger to the surface. nc-AFM and NIXSW results furthermore prove a reduction in adsorption height while XPS, UPS, and NEXAFS show an increased electronic hybridisation between molecule and surface. DFT results agree with these findings and show a localized interaction with the metal surface, both for the molecular model systems and the defect embedded into the graphene layer.

O 41.3 Wed 11:00 S052

Determining the stability and catalytic formation of graphene on liquid Cu using machine-learning potentials — •Hao Gao[1], Valentina Belova[2], Maciej Jankowski[2], Hendrik H. Heenen[1], Gilles Renaud[2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]ESRF, Grenoble, France

The recently discovered rapid, high-quality synthesis of graphene (Gr) on liquid Cu catalysts is microscopically still poorly understood. This is due to the difficult characterization of the Cu liquid surface. Especially in atomistic simulations, the large length and time scales necessary to reliably emulate the temporal evolution of the liquid are a major challenge. Corresponding molecular dynamics simulations require large simulation cells and need to span well into the nanosecond regime – an endeavor presently intractable via first-principles methods. In this work we use computationally efficient machine-learning potentials (MLPs) trained to density-functional theory (DFT) data in order to extrapolate the first-principles predictive power to the required scales. Detailed benchmarking confirms that our MLP captures the involved physics well, accurately reproducing the experimentally determined Gr adsorption height. We apply the MLP to further study the catalytic mechanism of Gr synthesis in order to rationalize distinct reaction kinetics found experimentally. Our work draws a path for the use of reliably trained MLPs as a multiscale modeling technique to explore previously unchartered computational problems. In that we provide new insight into the domain of liquid metal catalysts which generally lack atomic-scale understanding.

O 41.4 Wed 11:15 S052

Properties of epitaxial graphene on various SiC terminations and polytypes investigated by low-energy electron microscopy — •Philip Schädlich[1,2], Davood Momeni Pakdehi[3], Florian Speck[1,2], Klaus Pierz[3], and Thomas Seyller[1,2] — [1]Technische Universität Chemnitz, Chemnitz, Germany — [2]Center for Materials, Architectures and Integration of Nanomembranes (MAIN), Chemnitz, Germany — [3]Physikalisch-Technische Bundesanstalt, Braunschweig, Germany

The epitaxial growth of graphene on SiC has been improved from UHV growth to ambient pressure synthesis [1], and recently to the polymer-assisted sublimation growth (PASG) [2]. It results in an enhanced nucleation of the buffer layer and suppressed step bunching, which usually occurs upon the graphene formation by sublimation. Thus, PASG leads to homogeneous monolayer graphene with minimum step size equivalent to one or two SiC-bilayers. As a result of the latter, graphene on various surface terminations of the SiC substrate can now be systematically studied.

We utilize low-energy electron microscopy (LEEM) to identify the underlying substrate structure and to elucidate its delicate impact on the characteristic I(V) curves for different SiC polytypes. In addition, we investigate how the decoupling of the buffer layer from the substrate by hydrogen intercalation [3] influences the aforementioned effects.

[1] K. V. Emtsev et al., Nat. Mat. 8, 203 (2009). [2] M. Kruskopf et al., 2D Mater. 3 (4), 041002 (2016). [3] C. Riedl et al., PRL 103, 246804 (2009).

O 41.5 Wed 11:30 S052

Quasiparticle Interference Effects Revealed in Potassium-Intercalated Epitaxial Monolayer Graphene — Tobias Huempfner, •Felix Otto, Roman Forker, and Torsten Fritz — Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany

Epitaxial graphene is known for its fascinating physical properties, such as a linear dispersion at the K point, an anomalous quantum Hall effect and even superconductivity. These effects are based on the unique electronic structure of graphene, which can be tuned, e.g., by intercalation of metals. In this study, we scrutinize samples of epitaxial monolayer graphene (EMLG) on SiC(0001) intercalated with potassium by means of local and area-averaging experimental methods at low temperatures. For the highest accessible K concentration we find a highly ordered (2×2) superstructure that the K atoms form below the uppermost graphene layer. Further, we observe that the K atoms also reside below the buffer layer of the EMLG on SiC(0001) sample that is located between the quasi-freestanding graphene sheet and the SiC substrate. This causes an effective decoupling of the buffer layer and a transformation to K-intercalated epitaxial bilayer graphene (EBLG). This configuration promotes strong *n*-doping of the graphene sheets, where besides a rigid shift of the Dirac bands to higher binding energies also filling of two parabolic interlayer bands is observed.

O 41.6 Wed 11:45 S052

Structural and electronic properties of a van der Waals heterostructure of two-dimensional Pb and epitaxial graphene — •Bharti Matta, Philipp Rosenzweig, Olaf Bolkenbaas, Kathrin Küster, and Ulrich Starke — Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany

Intercalation is an established technique for manipulating the electronic properties of epitaxial graphene. Moreover, it is a way to confine otherwise unstable two-dimensional (2D) layers of elements, leading to unique physical properties compared to their bulk counterparts due to quantum confinement. In this work, we show uniformly Pb-intercalated quasi-freestanding monolayer graphene on SiC, which turns out to be essentially charge neutral. Additional bands – some crossing the Fermi level – can be clearly discerned, demonstrating the metallic character of 2D Pb sandwiched within the graphene/SiC heterointerface. The band velocity of the graphene Dirac cone changes at binding energies of around 0.6 eV and 1.1 eV, which points towards hybridization with these Pb bands. Low-energy electron diffraction reveals a 10×10 Moiré periodicity with respect to graphene. This is consistent with a triangular lattice of intercalated Pb that is strained by less than 2% relative to bulk-truncated Pb(111) and of which a $(\sqrt{3}\times\sqrt{3})$R30° supercell matches 2×2-SiC. However, finding direct signatures of the corresponding periodicities in the available band structure data is challenging. Our experimental results will provide a solid ground for further theoretical assessment of this system and better understanding of its properties.

O 41.7 Wed 12:00 S052

Surface Transport Properties of Pb- and Sn-intercalated Graphene — •M. Gruschwitz, C. Ghosal, Z. Mamiyev, J. Koch, S. Wolff, T. Seyller, and C. Tegenkamp — Institut für Physik, Technische Universität Chemnitz, Chemnitz, Germany

Intercalation experiments on epitaxial graphene are attracting a lot of attention at present as a tool to boost further the electronic properties of 2D graphene. In this work we studied the intercalation of Pb using buffer layers on 6H-SiC(0001). Recent DFT calculations suggest Pb atoms to act as electron donors thus allowing the doping level of the quasi-freestanding graphene layer to be tuned by the amount of intercalated material [1].

In this work we present results about the large-area intercalation of Pb and Sn, investigated by means of electron diffraction, scanning tunneling microscopy, photoelectron spectroscopy and in-situ surface transport. The intercalation of Pb and Sn results in formation of almost charge neutral graphene. In case of Pb, the intercalated layer consists of 2 ML and shows a strong structural corrugation. The epitaxial heterostructure provides an extremely high conductivity of $\sigma = 100$ mS/□. However, below 70 K we found a metal-insulator transition which we assign to the formation of minigaps in epitaxial graphene, possibly induced by a static distortion of graphene following the corrugation of the interface layer [2]. Sn intercalation does not cause such a transition while yielding a conductivity of $\sigma = 2.5$ mS/□.

[1] J. Wang et al., PRB 103, 085403 (2021); [2] M. Gruschwitz et al., Materials 14, 7706 (2021)

O 41.8 Wed 12:15 S052

Electron correlation effects in highly-doped single-layer graphene — •Vivien Enenkel[1], Philipp Rosenzweig[2], Hrag Karakachian[2], Fabian Paschke[1], Ulrich Starke[2], and Mikhail Fonin[1] — [1]Fachbereich Physik, Universität Konstanz, 78457 Konstanz, Germany — [2]Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany

Electronic correlations in graphene are expected to be strongly enhanced when there is a very high density of states at the Fermi level, giving rise to many-body states such as superconductivity or charge density waves (CDW) [1]. While the latter have been reported for example in Ca-intercalated bilayer graphene [2,3], in case of epitaxial monolayer graphene no direct evidence of correlated electronic ground states has yet been reported. We investigate heavily n-doped monolayer graphene on SiC(0001), obtained by Yb intercalation underneath zero-layer graphene. Here, a van Hove singularity (VHS) is pushed to the Fermi level, giving rise to an extended VHS, effectively pinning an almost non-dispersive flat band at EF [4]. Low-temperature STM reveals several distinct structures of Yb-intercalated graphene, which we attribute to differing arrangements of the Yb at the interface. dI/dU spectra show a pronounced gap feature centered at EF, whose response to field and temperature variations allows the interpretation of the feature as a CDW state. [1] M. L. Kiesel et al., Phys. Rev. B 86, 020507 (2012); [2] R. Shimizu et al., Phys. Rev. Lett., 114, 146103 (2015); [3] S. Ichinokura et al., ACS Nano 10, 2, 2761 (2016); [4] P. Rosenzweig et al., Phys. Rev. B 100, 035445 (2019).

O 42: Metal substrates 2

Time: Wednesday 10:30–11:45 Location: S053

O 42.1 Wed 10:30 S053

Atomic structure determination of epitaxially grown tin perovskite CsSnBr$_3$ on gold surfaces — •Madad Abbasli[1], Jenny Schrage[1], Jiaqi Cai[1], Jeremy Hieulle[2], Alex Redinger[2], Carsten Busse[1], and Robin Ohmann[1] — [1]University of Siegen, Department of Physics, Germany — [2]University of Luxembourg, Department of Physics and Materials Science, Luxembourg

Tin perovskites can be efficient and environmentally friendly substitutions to lead perovskites, but they have a drawback due to oxidation from Sn^{2+} to Sn^{4+}. Here, we present an in-situ study of epitaxially grown CsSnBr$_3$ on Au(111) and Au(100) in ultrahigh vacuum (UHV) using scanning tunneling microscopy (STM), low energy electron diffraction (LEED) and x-ray photoelectron spectroscopy (XPS). By co-evaporation of the precursor molecules CsBr and SnBr$_2$ submonolayers up to a few monolayers are obtained. On Au(111), CsSnBr$_3$ grows in three differently oriented domains due to the hexagonal symmetry of the substrate. On Au(100), which has square symmetry, identical to CsSnBr$_3$, but with about half of the lattice constant of the perovskite, we observe a (2x2) superstructure. Chemical analysis shows no indication of tin oxidation, which demonstrates the importance of preparation of tin perovskites in vacuum using evaporation.

O 42.2 Wed 10:45 S053

Peculiar growth of Mn on Ir (111) investigated by SP-STM — •Arturo Rodríguez Sota, Vishesh Saxena, André Kubetzka, Jonas Spethmann, Roland Wiesendanger, and Kirsten von Bergmann — Institut für Nanostruktur- und Festkörperphysik, Universität Hamburg

The growth of metallic layers has been studied for many years, from single atom coverages via clusters and islands to extended films. The experimental parameters used during the growth, such as deposition temperature or amount of material, determine the resulting phase. Usually, only one phase is present for a given preparation, but for europium on graphene the coexistence of clusters and islands of has been documented. This was attributed to several effects including charge transfer from the metal clusters to the substrate in a precise window of coverage and temperature [1].

Here we study the growth of Mn on Ir(111) using spin polarized scanning tunneling microscopy (SP-STM). In this metallic system we have found the coexistence of clusters and islands for any Mn submonolayer coverage. We have characterized the clusters in detail at low temperature and found several different configurations made out of three atoms. The Mn islands exhibit a reconstruction that disappears when the layer is completely closed. The monolayer presents the Néel state as its magnetic ground state.

[1] D. F. Förster et al., New J. Phys. 14, 023022 (2012).

O 42.3 Wed 11:00 S053

Breaking down of Stoner band ferromagnetism induced by interface formation — •David Janas[1], Andrea Droghetti[2], Iulia Cojocariu[3], Vitaliy Feyer[3], Stefano Ponzoni[1], Milos Radonjić[4], Ivan Rungger[5], Liviu Chioncel[6], Giovanni Zamborlini[1], and Mirko Cinchetti[1] — [1]TU Dortmund University, Dortmund, Germany — [2]Trinity College, Dublin, Ireland — [3]Forschungszentrum Jülich, Jülich, Germany — [4]University of Belgrade, Belgrade, Serbia — [5]National Physics Laboratory, Teddington, United Kingdom — [6]University of Augsburg, Augsburg, Germany

Interfaces between ferromagnetic transition metals and molecules or atoms display intriguing spin properties that have been invoked to explain the peculiar behavior of a variety of molecular spintronic and spin-optoelectronic devices [1]. Despite its relevance, a satisfying model for the hybridization occurring at such interfaces has not yet been presented. Experimentally, this is due to the lack of information on the interface band structure in the whole Brillouin zone, while, theoretically, the role of electron correlation has up to now mostly been neglected. Here, we overcome these limitations using state-of-the-art experimental and theoretical methods to explore the model interface between iron and an ordered oxygen layer. We use spin-resolved momentum microscopy to provide access to the complete spin-resolved interface band structure and explain our findings using DFT+Σ_2 calculations that go beyond the one-electron approximation [2].

[1] Cinchetti, M. et al. *Nature Mater* **16**, 507-515 (2017).

[2] Droghetti, A. et al. *Phys. Rev. B* **105**, 115129 (2022).

O 42.4 Wed 11:15 S053

Computational screening of self-intercalated transition metal dichalcogenides for enhanced electrocatalytic activity — •Stefano Americo and Kristian S. Thygesen — Center for Atomic-scale Materials Design, Department of Physics, Technical University of Denmark, DK - 2800 Kongens Lyngby, Denmark

Self-intercalation of metal atoms in bilayer transition metal dichalcogenides (TMD) has been recently realized experimentally through chemical vapor deposition (CVD) and molecular beam epitaxy (MBE), giving rise to completely new phases of the materials with controlled stoichiometry. However, little is known yet about the possible applications for this novel class of compounds. In this work, we use density functional theory to evaluate the catalytic activity towards electrochemical water splitting for 22 self-intercalated TMDs, including both metallic and semiconducting ones. We assess the thermodynamic and chemical stability of the compounds at different pH values and perform a systematic study of the hydrogen and oxygen adsorption energies at 33%, 66% and 100% intercalation degrees, identifying potential anode and cathode materials. Temperature-pressure phase diagrams are also calculated in order to guide the synthesis of compounds with the desired intercalation degree through CVD.

O 42.5 Wed 11:30 S053

A μ-Photoreactor for Investigating Planar Hydrogen Evolution Catalysts at Ambient Condition — •Clara Aletsee, Martin Tschurl, and Ueli Heiz — Physical Chemistry, Department of Chemistry & Catalysis Research Center, Technical University of Munich

Photocatalytic water splitting and alcohol reforming offer promising alternatives for clean H_2 production, in contrast to the industrially applied steam reforming of fossil fuels. However, the understanding of the reaction mechanisms ,which is indispensable for tailored performance improvement, is complicated by the structural complexity of powdered catalyst. In contrast, well-defined planar catalysts are already used for fundamental studies in ultra-high vacuum, but a simple extrapolation of the results to applied conditions is impractical due to a significant pressure difference.

Herein, we present a μ-photoreactor for the evaluation of planar model catalysts like single crystals or epitaxially grown semiconductors at ambient conditions in a continuous gas flow. In this system, the catalyst serves as the bottom of the reactor, while a 200 μm O-ring on top defines the reactor volume of 12 μL. It is sealed by a UV-vis transparent lid with lithographically implemented channels for the gas flow toward the quadrupole mass spectrometer for product analysis. The functionality is demonstrated by ethanol photoreforming over a Pt loaded TiO_2 catalyst. This concept will not only enable the evaluation of planar photocatalysts, but also the investigation of the transferability of UHV results to applied systems.

O 43: Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 1

Time: Wednesday 10:30–13:00 Location: S054

O 43.1 Wed 10:30 S054

Structure of Amorphous Phosphorus from Machine Learning-Driven Simulations — •Yuxing Zhou, William Kirkpatrick, and Volker L. Deringer — Department of Chemistry, Inorganic Chemistry Laboratory, University of Oxford Oxford OX1 3QR, UK

Amorphous phosphorus (a-P) has long attracted interest because of its complex atomic structure, and more recently as an anode material for batteries. However, accurately describing and understanding a-P at the atomistic level remains a challenge. In this talk, we show that a general-purpose Gaussian approximation potential (GAP) model for phosphorus can be created by machine learning (ML) from a suitably chosen ensemble of quantum-mechanical results. Its accuracy in describing the amorphous phase is demonstrated via large-scale molecular-dynamics simulations on the atomic structure of a-P: the calculated structure factors yield good agreement with earlier experimental evidence. Abundant five-membered rings are found in the structural model, which are the building block of more complex clusters. We provide new insights into the cluster fragments under pressure: an analysis of cluster fragments, large rings, and voids suggests that moderate pressure (up to about 5 GPa) does not break the connectivity of clusters, but higher pressure does. Changes in the simulated first sharp diffraction peak during compression and decompression indicate a hysteresis in the recovery of medium-range order. Our work provides a starting point for further computational studies of a-P, and more generally it exemplifies how ML-driven modeling can accelerate the understanding of disordered functional materials.

O 43.2 Wed 10:45 S054

Realistic Structural Properties of Amorphous SiNx from Machine-Learning-Driven Molecular Dynamics — •Ganesh Kumar Nayak[1], Prashanth Srinivasan[2], Juraj Todt[3], Rostislav Daniel[1], and David Holec[1] — [1]Department of Materials Science, Montanuniversität Leoben, Leoben, Austria — [2]Franz-Josef-Strasse 18 — [3]Erich Schmid Institute of Materials Science of the Austrian Academy of Sciences, Jahnstrasse 12, Leoben, Austria

Machine-learning(ML)-based interatomic potentials can enable simulations of extended systems with an accuracy that is largely comparable to DFT, but with a computational cost, that is orders of magnitude lower. Molecular dynamics simulations further exhibit favorable linear (order N) scaling behavior.

Amorphous silicon nitride (a-SiNx) is a widely studied noncrystalline material, and yet the subtle details of its atomistic structure and mechanical properties are still unclear. Due to the small sizes of representative models, DFT cannot reliably predict its structural properties and hence left an anisotropic order parameter. Here, we show that accurate structural models of a-SiNx can be obtained using an ML-based inter-atomic potential. Our predictions of structural properties are validated by experimental values of mass density by X-ray reflectivity measurements and by radial distribution function measured by synchrotron X- ray diffraction.

Our study demonstrates the broader impact of ML potentials for elucidating structures and properties of technologically important amorphous materials.

O 43.3 Wed 11:00 S054

Combined experimental-computational directed sampling approach to modelling amorphous alumina — •Angela Harper[1], Steffen Emge[2], Pieter Magusin[2,3], Clare Grey[2], and Andrew Morris[4] — [1]Cavendish Laboratory, University of Cambridg — [2]Department of Chemistry, University of Cambridge — [3]Institute for Life Sciences & Chemistry, Hogeschool Utrecht — [4]School of Metallurgy and Materials, University of Birmingham

Understanding the electronic and atomic level structure of materials is imperative for discovering the next generation of solid state electronic devices. Yet for amorphous materials, it is non-trivial to determine the exact local ordering. In this talk, I outline a method for modelling disordered materials, using experimentally directed sampling of static configurations from *ab initio* molecular dynamics[1]. We calculate experimentally relevant spectra and properties including X-ray absorption edges, nuclear magnetic resonance chemical shieldings, and the electronic density of states, with first principles accuracy. This model is validated on amorphous alumina, a widely used coating material in electronic devices, and identify two distinct five-fold coordinated geometries of AlO_5, as well as localised states at the conduction band minimum. By leveraging both experimental and computational data in our approach we highlight the need for experimentally informed calculations which lead to a more detailed understanding of complex materials, and develop an approach that is widely applicable to the modelling community.

[1] Harper, AF et al., Under review (2021)
`doi.org/10.33774/chemrxiv-2021-qjzbj`

O 43.4 Wed 11:15 S054

Structural phases and thermodynamics of $BaTiO_3$ from an integrated machine learning model — •Lorenzo Gigli[1], Max Veit[1], Michele Kotiuga[2], Giovanni Pizzi[2], Nicola Marzari[2], and Michele Ceriotti[1] — [1]Laboratory of Computational Science and Modeling (COSMO), Institute of Materials, École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland — [2]Theory and Simulation of Materials (THEOS) and National Centre for Computational Design and Discovery of Novel Materials (MARVEL), École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

Modeling the ferroelectric transition of any given material requires three key ingredients: (1) a model of the potential energy surface, that describes the energetic response to a structural distortion; (2) the free energy surface sampled at the relevant, finite-temperature conditions; and; (3) the polarization of individual configurations that determines the observed polarization and the phase transitions. To this aim, we make use of an integrated machine-learning framework, based on a combination of an interatomic potential and a microscopic polarization model, which we use to run Molecular Dynamics simulations of ferroelectrics with the same accuracy of the underlying DFT method, on time and length scales that are not accessible to direct ab-initio modeling. This allows us to uncover the microscopic nature of the ferroelectric transition in barium titanate ($BaTiO_3$) and to identify the presence of an order-disorder transition as the main driver of ferroelectricity. The framework also allows us to reconstruct the temperature-dependent $BaTiO_3$ phase diagram, with first-of-its-kind accuracy.

O 43.5 Wed 11:30 S054

Dielectric properties of $BaTiO_3$ from an integrated machine-learning model — •Max Veit[1], Lorenzo Gigli[1], Michele Kotiuga[2], Giovanni Pizzi[2], Nicola Marzari[2], and Michele Ceirotti[1] — [1]Laboratory for Computational Science and Modeling (COSMO), Ecole Polytechnique Fédérale de Lausanne, Lausanne, CH — [2]Laboratory for Theory and Simulation of Materials (THEOS), Ecole Polytechnique Fédérale de Lausanne, Lausanne, CH

Modeling the finite-temperature behavior of ferroelectric materials from first principles has always been challenging due to the large supercells and long simulation times required for adequate sampling. Here we demonstrate the use of an integrated machine learning (ML) model of the potential energy and polarization surfaces of barium titanate ($BaTiO_3$) to overcome these difficulties and run long MD simulations with DFT accuracy. We use these simulations to compute the frequency-dependent dielectric response function, finding a spectrum qualitatively similar that obtained with previous effective-Hamiltonian simulations as well as to experimentally measured profiles, with some remaining discrepancies that we trace back to the underlying DFT model. Finally, we discuss possible extensions of the model to explicitly include long-range interactions, previously included only in an implicit, emergent manner. We expect this integrated, generally applicable modeling technique to become a valuable tool for elucidating the ferroelectric behavior of a wide variety of materials.

O 43.6 Wed 11:45 S054

The first-principles phase diagram of monolayer nanoconfined water — •Venkat Kapil[1], Christoph Schran[1], Andrea Zen[2], Ji Chen[3], Chris Pickard[1], and Angelos Michaelides[1] — [1]University of Cambridge, UK — [2]Universitá di Napoli Federico II, Italy — [3]Peking University, Beijing, China

Water in nanoscale cavities is ubiquitous and of central importance to everyday phenomena in geology and biology, and at the heart of current and future technologies in nano-science. A molecular-level picture of the structure and dynamics of nano-confined water is a prerequisite to understanding and controlling the behavior of water under confinement. Here we explore a monolayer of water confined within a graphene-like channel using a framework that combines developments in high-level electronic structure theory, machine learning, and statistical sampling. This approach enables a treatment of nano-confined water at unprecedented accuracy. We find that monolayer water exhibits surprisingly rich and diverse phase behavior that is highly sensitive to temperature and the van der Waals pressure acting within the nano-channel. Monolayer water exhibits numerous molecular ice phases with melting temperatures that vary by over 400 degrees in a non-monotonic manner with pressure. In addition, we predict two unexpected phases: a *hexatic-like* phase, which is an intermediate between a solid and a liquid, and a superionic phase with a high electrical conductivity exceeding that of battery materials. Our work suggests that nano-confinement could be a promising route towards superionic behaviour at easily accessible conditions.

O 43.7 Wed 12:00 S054

Exploring amorphous graphene with empirical and machine-learned potentials — •Zakariya El-Machachi[1], Mark Wilson[2], and Volker L. Deringer[1] — [1]Department of Chemistry, Inorganic Chemistry Laboratory, University of Oxford, Oxford OX1 3QR, UK — [2]Department of Chemistry, Physical and Theoretical Chemistry Laboratory, University of Oxford, Oxford OX1 3QZ, UK

The structure of amorphous graphene (aG) lacks long range order whilst having short and medium range order yielding a rich and complex configurational space, which is yet to be fully understood. Here we report on an atomistic modelling study of aG using a machine learning (ML) based force field. ML force fields are typically "trained" on data from highly accurate but computationally costly density functional theory (DFT) computations. Atomistic models created by such ML methods can achieve near DFT accuracy at a fraction of the computational cost. One key assumption is that the global energy can be separated into sums of local energies. The physical interpretation of ML local energies is an interesting research question. We find that local and nearest neighbour (NN) ML energies can inform the generation of aG models from crystalline graphene via a Monte–Carlo bond switching algorithm. Bond switches are introduced as Stone–Wales defects, with the local energies of the defect pair and its NNs used in the acceptance criterion. Established empirical force fields are used in the same way and the resulting structures are studied. Our results provide insight into the modelling of amorphous graphene and into the nature of ML potential-energy models.

O 43.8 Wed 12:15 S054

Machine learning for estimation of spin models in undoped cuprates — •Denys Y. Kononenko[1], Ulrich K. Rössler[1], Jeroen van den Brink[1,2], and Oleg Janson[1] — [1]Institute for Theoretical Solid State Physics, IFW Dresden, Dresden, Germany — [2]Institute for Theoretical Physics, TU Dresden, Dresden, Germany

Undoped cuprates tailor a fascinating variety of low-dimensional and frustrated spin models, which can be indirectly characterized by the transfer integrals. The estimation of transfer integrals is related to a relatively complicated computational procedure which includes besides DFT calculation also a Wannierization. We propose a data-driven approach to replace this computationally demanding procedure.

We employ the Gaussian Process Regression model, trained on the results of high-throughput DFT calculations to estimate transfer integrals in undoped cuprates. The model learns from data the dependency between the local crystal environment of copper atoms pair and the corresponding value of transfer integral. The site position function of the local crystal environment is represented as a finite-dimensional vector composed of decomposition coefficients in the truncated basis of Zernike 3D functions [1]. The vector descriptor incorporates the spatial configuration and chemical composition of the local crystal environment. The proposed approach can be utilized for a rapid assessment of the spin models of new cuprates using structural information as the only input.

[1] M. Novotni and R. Klein, Computer Aided Design 36, 1047 (2004)

O 43.9 Wed 12:30 S054

Machine-learning Based Screening of Lead-free Perovskites for Photovoltaic Applications — •Elisabetta Landini[1,3], Harald Oberhofer[2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut

der Max-Planck-Gesellschaft, Faradayweg 4-6, D-14195 Berlin, Germany — [2]Chair for Theoretical Physics VII, Physikalisches Institut Universität Bayreuth, 95440, Bayreuth, Germany — [3]Chair for Theoretical Chemistry, Technische Universität München, Lichtenbergstr. 4, D-85747 Garching, Germany

Lead-free halide double perovskites are promising stable and non-toxic alternatives to methylammonium lead iodide in the field of photovoltaics. In this context, the most commonly used double perovskite is $Cs_2AgBiBr_6$, due to its favorable charge transport properties [1]. However, the maximum power conversion efficiency obtained for this material does not exceed 3%, as a consequence of its wide indirect gap and its intrinsic and extrinsic defects [2]. On the other hand, the materials space that arises from the substitution of different elements in the 4 lattice sites of this structure is large and still mostly unexplored.

In this work a neural network is used to predict the band gap of double perovskites from an initial space of 7056 structures and select candidates suitable for visible light absorption. Successive hybrid DFT calculations are used to evaluate the thermodynamic stability and the power conversion efficiency of the selected compounds, and propose novel potential solar absorbers.

[1] E.T. McClure *et al.*, *Chemistry of Materials* **28**, 1348 (2016).
[2] X. Yang *et al.*, *Energy & Fuels* **34**,10513 (2020).

O 43.10 Wed 12:45 S054

Equivariant graph neural network for linear scaling electron density estimation and applications in battery materials — •Arghya Bhowmik and Peter Jorgensen — 301 Anker Engelunds vej, Kgs. Lyngby, DK-2800

We present a machine learning framework for the prediction of ρ(r) based on equivariant graph message passing neural networks. The electron density is predicted at special query point vertices that are part of the message passing graph, but only receive messages. The model is tested across multiple data sets of molecules (QM9), liquid ethylene carbonate electrolyte (EC) and LixNiyMnzCo(1-y-z)O2 lithium ion battery cathodes (NMC). The model is used to explore large materials phase space for safer battery materials and uncovering new understanding how redox mediated diffusion occurs and battery materials.

O 44: Focus Session: Surfaces and Interfaces of (Incipient) Ferroelectrics (joint session O/KFM)

Ferroelectricity is a property of materials that allows spontaneous, switchable electric polarization. Recently, many surface-related applications have been proposed where ferroelectric or incipient-ferroelectric materials exhibit superior properties. These include catalysis, electron-hole separation in light harvesting, unique electronic properties such as a negative capacitance in heterostructures of ferroelectric materials, to name just a few. While (incipient) ferroelectrics clearly perform well in the aforementioned applications, there is very limited fundamental understanding of the processes involved on the surfaces of these materials.

Organizers: Martin Setvin (Charles University, Prague), Chiara Gattinoni (London South Bank University), and Michele Reticcioli (University of Vienna)

Time: Wednesday 15:00–18:30 Location: H3

Topical Talk O 44.1 Wed 15:00 H3

In search of electrostatic happiness at surfaces — •Nicola Spaldin — Materials Theory, ETH Zurich

We review the concept of surface charge in ionic insulators, first, in the context of the polarization in ferroelectric materials (traditionally discussed in the ferroelectrics community) and, second, in the context of layers of charged ions (traditionally discussed in the surface science community). In both cases, the surface charge leads to an electrostatic instability if it is not compensated, which is usually detrimental for the development of electronic devices based on ferroelectrics, but favorable for applications such as catalysis where surface reactivity is desirable.

Using the prototypical multiferroic bismuth ferrite, BiFeO3, as an example, we show how the spontaneous ferroelectric polarization and the charged ionic layers can in fact combine to yield stable, uncharged “happy” (100) surface geometries. Switching the polarization causes these (100) surfaces considerable electrostatic distress, which must be compensated by the introduction of charged point defects or adsorbates. We demonstrate that the relative happiness or unhappiness of the surfaces enables a cycle of alternating charged then neutral adsorbates on polarization switching, which can be exploited for water remediation and water splitting.

We close with a proposal that these physics can induce polarization in thin films of certain usually non-polar materials, and give a recipe for determining likely candidates.

In collaboration with Chiara Gattinoni, Ipek Efe and Marta Rossell

Topical Talk O 44.2 Wed 15:30 H3

Synthesis and Characterisation of Ultra-thin Aurivillius Phase Multiferroics — •Lynette Keeney — Tyndall National Institute, University College Cork, Lee Maltings Complex, Dyke Parade, Cork, Ireland, T12 R5CP

Multiferroic materials, possessing simultaneous ferroelectric and ferromagnetic memory states, have been road-mapped as promising multistate architectures for memory scaling beyond current technologies. In recent years, my team reported the design of such a novel room temperature multiferroic material with an Aurivillius phase structure that could ideally be suited to future fabrication of revolutionary memory devices. Electrostatic strain and elastic energy variations close to defect regions increase the extent of magnetic partitioning and also influence the formation of exotic charged domain walls and polar vortices. This further initiates technology prospects in ultra-compact data storage, energy-efficient neuromorphic computing and ultrahigh speed data processing. As miniaturisation of electronic devices continues, a crucial requirement is the enhancement of their functional properties at very small dimensions. In this presentation, I will discuss how direct liquid injection chemical vapour deposition allows for frontier-development of ultra-thin films at fundamental thickness. Via a two-dimensional layer-by-layer growth mode, films equating to half of one unit-cell (2.5 nm) of the Aurivillius structure are grown. The persistence of stable ferroelectricity, even when pushed to ultra-thin thicknesses, demonstrates the recent progress in the optimisation of Aurivillius phase materials for utilisation in future miniaturised multiferroic-based devices.

O 44.3 Wed 16:00 H3

Influence of Nb dopants on the polarization and screening on cleaved $SrTiO_3$(001) surfaces — •Igor Sokolović[1], Alexander Hoheneder[1], Jesus Redondo[2], Dominik Wrana[2], Michael Schmid[1], Ulrike Diebold[1], and Martin Setvín[2] — [1]Institute of Applied Physics, TU Wien, Vienna, Austria — [2]Faculty of Physics and Mathematics, Charles University, Prague, Czech Republic

The incipient ferroelectric $SrTiO_3$ can turn ferroelectric even at room temperature under the application of strain, and this quantum phase transition can be utilized to cleave $SrTiO_3$ single crystals that otherwise posses no preferable cleavage planes [1]. This cleaving procedure creates truly bulk-terminated $SrTiO_3$(001) surfaces that come the closest to being pristine [2]. In this talk, I will present how the SrO- and TiO_2-terminated surface domains of opposite polarity can be influenced by the small changes in the amounts of Nb doping. The cleaved $SrTiO_3$(001) surfaces with varying Nb doping levels were systematically studied with atomic resolution using noncontact atomic force microscopy (ncAFM) and scanning tunneling microscopy (STM). It was observed that Nb doping does not affect the magnitude of the strain-induced polarization, yet still significantly affects the morphology, the electronic structure, and the domain-wall structure on cleaved

SrTiO$_3$(001) surfaces. Beside demonstrating the interplay between the domain distribution and electrostatic screening, these results show how the properties of these heterogeneous surfaces can be tuned.

[1] Sokolović *et al.*, Phys. Rev. Mater. 3, 034407 (2019)

[2] Sokolović *et al.*, Phys. Rev. B 103, L241406 (2021).

O 44.4 Wed 16:15 H3

Polaronic Properties of the weakly-polar SrTiO$_3$(001) Surface — •Florian Ellinger[1], Michele Reticcioli[1], Igor Sokolović[2], Ulrike Diebold[2], Martin Setvín[3], and Cesare Franchini[1,4] — [1]University of Vienna — [2]Technische Universität Wien — [3]Charles University Prague — [4]Università di Bologna

The SrTiO$_3$(001) surface shows ferroelectric-like distortions on the bulk-like termination, an out-of-plane dipole moment, and so-called "weak polarity". Recent experiments propose that these effects are compensated by Sr-adatoms and -vacancies, stabilizing the unreconstructed surface. [1]

We investigate the 1×1 SrTiO$_3$(001) surface with TiO$_2$- and SrO-terminations by means of density functional theory (DFT) simulations. Our calculations confirm the experimental interpretation and show that these polarity-compensating surface defects introduce additional charge. Adsorbing Sr-adatoms and doping with Nb leads to excess electrons in the crystal, facilitating the formation of electron-polarons. On the other hand, by creating Sr-vacancies on the surface we introduce excess holes to the system, which can localize as hole-polarons. For both kinds of polarons we analyze their general properties, e.g., the preferred localization site or stability. Further, we compare results of different structural phases of this crystal to achieve a comprehensive understanding of mentioned physical phenomena.

[1] Sokolović et al., *Incipient ferroelectricity: A route towards bulk-terminated SrTiO$_3$*, Phys. Rev. Materials 3, 034407 (2019)

O 44.5 Wed 16:30 H3

Ferroelectric domain imaging in barium titanate using infrared-visible sum-frequency generation microscopy — •Dorothee Mader[1], Daniel Lourens[2], Maarten Kwaaitaal[2], Richarda Niemann[1], Sören Wasserroth[1], Sandy Gewinner[1], Marco De Pas[1], Wieland Schöllkopf[1], Martin Wolf[1], Andrei Kirilyuk[2], Sebastian F. Maehrlein[1], and Alexander Paarmann[1] — [1]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [2]Radboud Universiteit, Nijmegen, The Netherlands

Phonons exhibit a mostly unexplored leverage on the mechanisms and dynamics of domain formation in ferroics. Here, a new method is employed combining resonant phonon excitation and ferroelectric domain imaging of barium titanate (BTO) using infrared-visible (IR-VIS) sum-frequency generation (SFG) microscopy [1]. BTO is a non-centrosymmetric perovskite oxide with a strong ferroelectric polarization in its tetragonal phase. Typically, BTO samples exhibit a multi-domain structure. In this contribution, SFG microscopy is shown to naturally provide domain contrast due to the polarization-induced local variation of the nonlinear susceptibility. Additionally, our spectral analysis of the SFG response reveals the domain-selective phonon resonances for all high-frequency phonons in the IR spectral range of 500-800 cm^{-1}. By locally mapping phonon resonances in domains and domain walls, this approach may enable in-depth understanding of the underlying physics of domain formation and its dynamics. [1] R. Niemann et al., Appl. Phys. Lett. 120, 131102 (2022).

Topical Talk O 44.6 Wed 16:45 H3

Water-oxidation catalysis on surfaces of ferroelectrics — •Ulrich Aschauer[1], Nathalie Vonrüti[1], Zhenyun Lan[2], Didrik R. Småbråten[1], Tejs Vegge[2], and Ivano E. Castelli[2] — [1]University of Bern, Bern, Switzerland — [2]Technical University of Denmark, Kgs. Lyngby, Denmark

Surfaces of ferroelectrics have unique properties for catalysis since the binding strength of reaction intermediates can be modulated by switching the ferroelectric polarization. This could allow to overcome the limitations of the Sabatier principle and enable dynamical catalysts operation. In this talk, we will focus on the interplay between screening charge transfer to surfaces, the adsorbate coverage and the (photo)electrochemical water-oxidation activity of ferroelectric surfaces. We will compare different ferroelectric materials such as BaTiO$_3$, strained LaTiO$_2$N and the hexagonal improper ferroelectric oxynitride InSnO$_2$N. Our results indicate that ferroelectric switching can indeed provide an economically interesting route to enhance the catalytic activity but that material-specific intricacies of the surface adsorbate coverage need to be understood and controlled to exploit the full potential of ferroelectric switching in (photo)electrocatalysis.

O 44.7 Wed 17:15 H3

The polar KTaO$_3$ (001) surface: Electronic structure and CO adsorption — Zhichang Wang[1], Michele Reticcioli[2], Zdenek Jakub[1], Michael Schmid[1], Gareth Parkinson[1], Ulrike Diebold[1], Cesare Franchini[2], and •Martin Setvin[3] — [1]TU Wien, Vienna, Austria — [2]University of Vienna, Vienna, Austria — [3]Charles University, Prague, Czech Republic

Polar surfaces offer intriguing physical and chemical properties applicable in electronics or catalysis. Cleaving the KTaO$_3$ perovskite along its polar (001) plane provides a well-defined, bulk-terminated surface with KO and TaO$_2$ terminations [1]. As-cleaved surfaces exhibit a high concentration of in-gap states; these electrons predominantly reside at the TaO$_2$-terminated parts of the surface. These electrons can affect surface chemistry, as is demonstrated for CO molecules. CO has two adsorption configurations on the TaO$_2$ termination, and the CO differs in how it couples to the excess electrons. DFT calculations indicate that CO preferentially couples to electron bipolarons.

The work was supported by FWF project P32148-N36, by GACR 20-21727X and GAUK Primus/20/SCI/009.

[1] M. Setvin, M. Reticcioli, F. Poelzleitner et al., Science 359, 572 (2018)

O 44.8 Wed 17:30 H3

Polarons and ferroelectricity: tip-induced phenomena on oxide perovskite surfaces — •Dominik Wrana[1], Igor Sokolović[2], Jesus Redondo[1], Pavel Kocán[1], Aji Alexander[1], Llorenç Albons[1], and Martin Setvin[1] — [1]Department of Surface and Plasma Science, Charles University, Prague, Czech Republic — [2]Institute of Applied Physics, TU Wien, Vienna, Austria

In this talk, I will present the similarities and differences between two representative perovskite oxide surfaces: KTaO$_3$(001) and BaTiO$_3$(001), showcasing the manifestation of the (incipient-) ferroelectricity on the atomic and electronic structure. Both surfaces were prepared by cleaving single crystals in situ and characterized by means of qPlus nc-AFM at temperatures ranging from 4K to 100K.

Bulk-terminated KTaO$_3$(001) develops two alternating domains of KO and TaO$_2$ [1]. Excess electrons injected from the AFM tip form quasiparticles called polarons (charges coupled with lattice distortions) which can be further shaped into 1D or 2D structures by emerging electric fields.

Different mechanism applies in the case of BaTiO$_3$(001), where at low temperatures titanium atoms can easily break the symmetry causing a spontaneous polarization. Hence, a biased tip allows for reversible manipulation of individual atoms on the surface: writing and erasing polarized ferroelectric domains.

[1] Setvin, Martin, et al. Science 359.6375 (2018): 572-575

O 44.9 Wed 17:45 H3

Optimisation and miniaturisation of naturally-layered multiferroic thin films — •Lynette Keeney — Tyndall National Institute, University College Cork, Lee Maltings Complex, Dyke Parade, Cork, Ireland, T12 R5CP

Multiferroic materials, possessing simultaneous ferroelectric and ferromagnetic memory states, are road-mapped as promising multistate architectures for memory scaling beyond current technologies. In recent years, my team reported the design of such a novel room temperature multiferroic material with an Aurivillius phase structure that could ideally be suited to future fabrication of revolutionary memory devices. In this presentation, I will discuss how electrostatic strain and elastic energy variations close to bismuth oxide interfaces and defect regions are key to promoting magnetic cation partitioning and multiferroic behaviour. These also influence the formation of exotic charged domain walls and polar vortices, further initiating technology prospects in ultra-compact data storage. As miniaturisation of electronic devices continues, a crucial requirement is the enhancement of their functional properties at very small dimensions. Direct liquid injection chemical vapour deposition allows for frontier-development of ultra-thin films at fundamental thickness. Via a two-dimensional layer-by-layer growth mode, films equating to half of one unit-cell (2.5 nm) of the Aurivillius structure are grown. The persistence of stable ferroelectricity, even when pushed to ultra-thin thicknesses, demonstrates the recent progress in the optimisation of Aurivillius phase materials for utilisation in future miniaturised multiferroic-based devices.

Topical Talk O 44.10 Wed 18:00 H3
Spin-orbitronics and superconductivity in KTaO3 twodimensional electron gases — •Srijani Mallik[1], Gerbold Ménard[2], Guilhem Saiz[2], Hugo Witt[1,2], Sara Varotto[1], Luis M. Vicente-Arche[1], Julien Bréhin[1], Annika Johansson[3], Börge Göbel[4], Raphaël Salazar[5], Ingrid Mertig[4], Lara Benfatto[6], Nicolas Bergeal[2], and Manuel Bibes[1] — [1]Unité Mixte de Physique CNRS/Thales, Palaiseau, France — [2]LPEM ESCPI, Paris, France — [3]MPI, Halle, Germany — [4]Martin-Luther-Universitat Halle-Wittenberg, Germany — [5]Synchrotron SOLEIL, France — [6]Sapienza University of Rome, Italy

Similar to SrTiO3 (STO) recent research has shown that KTaO3 (KTO) may also harbor a 2DEG at interfaces with several oxide materials. Due to the presence of Ta (5d element), it is expected that the Rashba spin-orbit coupling in KTO 2DEGs should be larger than in STO 2DEGs. Further, (110) and (111)-oriented KTO 2DEG show superconductivity at temperature a factor of ca. 10 higher than in STO 2DEGs. In this talk we will show that 2DEGs can be generated by the simple deposition of Al metal on KTO single crystals. We will report their electronic band structure by angle-resolved photoemission spectroscopy, evidencing a peculiar Rashba splitting. We will show that this Rashba state can be harnessed to achieve very efficient spin-charge interconversion. Finally, we will present microwave impedance spectroscopy measurements of the superconducting condensate and discuss the nature of superconductivity in these systems.

O 45: Focus Session: Catalysis at Liquid Interfaces

Catalysis at liquid Interfaces has emerged as novel approach to explore the highly dynamic, anisotropic environment of liquid interfaces to create, tailor, and stabilize catalytically active sites with unique reactivity and performance. With this concept, fundamental problems of catalyst science and technology can be addressed to develop novel catalytic materials that combine selectivity, productivity, robustness, and ease of processing at the highest possible level. Thus, the concepts of "Supported Catalytically Active Liquid Metal Solutions" (SCALMS), interface-enhanced "Supported Ionic Liquid Phase" (SILP) and advanced "Solid Catalysts with Ionic Liquid Layer" (SCILL) systems are of particular focus.

Organizers: Marcus Bär (Helmholtz-Zentrum Berlin), Jörg Libuda (FAU Erlangen-Nürnberg), Christian Papp (FAU Erlangen-Nürnberg)

Time: Wednesday 15:00–18:00 Location: H4

Topical Talk O 45.1 Wed 15:00 H4
Addressing Electronic Effects in Catalysis by Intermetallic Compounds — •Marc Armbrüster — Chemnitz University of Technology

The catalytic properties of materials are determined by their electronic and geometric structure. A reliable development addresses both influences separately and then combines the ideal electronic and structural properties resulting in superior catalytic materials.

Unfortunately, this is only straightforward on paper. The implementation faces hurdles concerning the control of the materials. E.g. the electronic modification of metallic materials by addition of a second metal with a different electron count can result in substitutional alloys with (in theory) random distribution of the elements. However, the sought-for electronic modification is not reachable due to segregation. Another intrinsic challenge is the geometric and electronic entanglement, i.e. when substituting one element by another, the structure might change and with it, the electronic properties.

A new approach is the partial substitution of main group elements in isostructural intermetallic compounds, enabling excellent electronic control with minimal geometric variation. Taking InPd2 and substituting In by Sn alters the valence electron count, while the distances are mostly not affected. Tuning the In/Sn ratio allows to address the electronic influence with very high resolution. Results in gas-phase hydrogenation and electrocatalysis are presented as examples of the broadly applicable approach.

O 45.2 Wed 15:30 H4
Gallium droplets on nanostructured surfaces as matrix material for supported liquid metal solutions — •André Hofer, Nicola Taccardi, Peter Wasserscheid, and Julien Bachmann — FAU Erlangen, Germany

A large variety of organic compounds are produced industrially using a dehydrogenation reaction which removes H_2 from a molecule and hence converts it into a more reactive and valuable product. Usually, such a reaction is endothermic and needs to be catalysed or performed at elevated temperatures. Our approach for improving the availability of the catalyst is the utilization of supported catalytically active liquid metal solutions (SCALMS). Here, the catalytically active material is dissolved in a dynamic liquid matrix, protecting it from undesired deactivation. Ga is used in our approach as the matrix material due to its outstanding properties. So far, SCALMS catalysis has been performed with very limited control of particle sizes. Here, we present a model system to determine the fundamental kinetic parameters of SCALMS reactions. We fabricate highly ordered, nanostructured substrates by electrochemical anodization of Al, generating indentations in a hexagonal arrangement with a tunable diameter. These substrates are coated with acidic or alkaline metal oxides by ALD to affect the surface tension and wetting behaviour of the Ga droplets. Subsequently, a Ga complex is spin-coated from organic solution on the surfaces and the elementary Ga droplets are obtained upon thermal treatment. Parameters relevant to SCALMS application of the surface fabricated are investigated systematically as they depend on the preparative parameters.

O 45.3 Wed 15:45 H4
Extending the SCALMS catalytic concept to ternary systems — •Michael Moritz[1], Sven Maisel[1], Narayanan Raman[2], Haiko Wittkämper[1], Christoph Wichmann[1], Nicola Taccardi[2], Peter Wasserscheid[2], Andreas Görling[1], Hans-Peter Steinrück[1], and Christian Papp[1] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg, Department Chemie und Pharmazie, Egerlandstraße 3, 91058 Erlangen, Germany — [2]Friedrich-Alexander-Universität Erlangen-Nürnberg, Department Chemie- und Bioingenieurwesen, Egerlandstraße 3, 91058 Erlangen, Germany

Over the last few years, supported catalytically active liquid metal solutions (SCALMS) were shown to be remarkably efficient catalysts for alkane dehydrogenation. The active phase of these catalysts consists of a liquid metal matrix, in which catalytically active transition metals are dissolved. So far, only Gallium was used as a liquid matrix material, because of the low toxicity and low melting point. Since extensive research has already been made on Gallium-based SCALMS systems with different catalytically active transition metals, varying the matrix material is the logical next step to further improve the catalyst performance. These efforts are now extended to ternary systems such as GaInPt and GaSnPt. This extension opens up new questions as it increases the complexity, but also brings exciting new properties and insights into the dynamics of liquid SCALMS catalysts. In this talk, the most recent results regarding these ternary systems will be presented, with an emphasis on photoemission studies. Funded by SFB 1452.

O 45.4 Wed 16:00 H4
Investigation of Pt-Ga SCALMS model particles using photoelectron spectroscopy — •Christoph Wichmann[1], Haiko Wittkämper[1], Micheal Moritz[1], Tzung-En Hsieh[2], Johannes Frisch[2], Mingjian Wu[3], Marcus Bär[2], Erdmann Spiecker[3], Hans-Peter Steinrück[1], and Christian Papp[1] — [1]Universität Erlangen, Physikalische Chemie II, Germany — [2]Helmholtz-Zentrum Berlin, Germany — [3]Universität Erlangen, Lehrstuhl für Werkstoffwissenschaften, Germany

Supported catalytically active liquid metal solutions (SCALMS) recently gained much attention as highly active and stable catalysts for the dehydrogenation of alkanes. SCALMS are metal solutions composed of low amounts of catalytically active transition metals like Rh, Pd, or Pt, alloyed with low melting metals like Ga, which act as matrix. Their stability is attested to a notable resistance against deactivation by coking, which is the prevalent deactivation route for dehydrogenation catalysts. To obtain a fundamental understanding, we make these complex materials systems accessible for a surface science approach. To gain insight in the catalytic properties of such nano-solutions, we work with supported PtGa particle systems. Herein, supported PtGa particle systems with low Pt content were investigated. The influence of the deposition sequence, the effects of annealing, and exposure to oxygen on the alloying and morphology of the resulting particle were studied with XPS using different excitation energies to obtain a depth-profile of the first few layers of the liquid.

Funded by SFB 1452.

Topical Talk O 45.5 Wed 16:15 H4

Understanding liquid metal catalysts for graphene synthesis using machine learning interatomic potentials — •Hendrik H. Heenen — Fritz-Haber-Institut der MPG, Berlin, Germany

High-quality, near defect-free graphene can be synthesized on the levelled and uniform surfaces of liquid metal catalysts. This smoothness on the microscale is sometimes accompanied by seemingly different catalytic properties, the determination of which is, however, ambiguous. Assessing distinct catalytic properties of liquid metal catalysts by first principles atomistic simulations has so far been challenged due to the intractable long length and time scales necessary to model the liquid phase. Using computationally efficient machine learning interatomic potentials (MLIPs) trained to first principles data allows to extrapolate predictability to necessary scales and opens an avenue for obtaining the desired microscopic insight.

In this talk I will present strategies to train and employ MLIPs for the simulation of graphene synthesis on liquid metal catalysts. I will introduce the data-efficient training of MLIPs via fairly automatic workflows as a tool to extend the predictive accuracy of e.g. density functional theory to larger scales. On basis of these potentials, large-scale simulations can be performed to compute experimental observables and elucidate microscopic processes relevant to graphene synthesis. Further, one can identify trends between different metals and directly compare between the solid and liquid states of a catalyst. Findings based on these simulation approaches shed new light on the role of the liquid state of liquid metal catalysts.

Topical Talk O 45.6 Wed 16:45 H4

Ionic liquids and deep eutectic solvents - sustainable media for selective molecular recognition and adsorption — •Jan Blasius, Leonard Dick, and Barbara Kirchner — Mulliken Center for Theoretical Chemistry, University of Bonn, Beringstrasse 4+6, D-53115 Bonn, Germany

The effective reduction of CO2 emission and chemical waste production are key disciplines on our way towards a more sustainable future. Ionic liquids (ILs) and deep eutectic solvents (DESs) offer new opportunities for addressing these challenges, as their properties can be adjusted in order to fulfill specific purposes. To efficiently optimize their usage for various applications, it is vital to understand the underlying molecular behavior and inherent structuring inside these liquids. Using state-of-the-art theoretical methods, we shed light on molecular adsorption and recognition mechanisms in ILs and DESs. This enables us to determine the reasons for specific capacities and allows a theoretical design of solvents with precisely tuned geometric, energetic and kinetic parameters in order to enhance the processes of interest. In this presentation, we will highlight our most recent research concerning molecular recognition and adsorption in ILs and DESs which can support the development of novel chemical processes that pave our way towards a more sustainable future.

O 45.7 Wed 17:15 H4

Surfaces of ionic liquids studied by ARXPS and UHV Pendant Drop — •Ulrike Paap, Florian Maier, and Hans-Peter Steinrück — Chair of Physical Chemistry II, FAU Erlangen-Nürnberg, Egerlandstraße 3, 91058 Erlangen, Germany

Ionic liquids (ILs) are characterized by a low melting point, low vapor pressure and low surface tension. Many ILs exhibit high gas solubility along a high chemical and thermal stability. Such properties are beneficial in many areas such as gas absorbents, refrigerants, lubricants, antistatics and surfactants. In this work, we studied various imidazolium-based ILs with different functional PEG chains attached to the cation using angle-resolved X-ray photoelectron spectroscopy (ARXPS) and a new pendant drop setup, which enables to measure the surface tension also in ultrahigh vacuum (UHV). With ARXPS, we determine the surface composition of the topmost IL nanometers on the molecular level. We combine these results with our surface tensions studies in UHV and atmosphere. Our results allow for a deeper understanding of enrichment and molecular orientation processes at the outermost surface of these systems in a microscopic and macroscopic range.

This work was supported by the European Research Council (ERC) through an Advanced Investigator Grant (ILID 693398) to HPS and the Collaborative Research Center (CRC) 1452: Catalysis at Liquid Interfaces.

O 45.8 Wed 17:30 H4

Mixed Films of protic and non-protic Ionic Liquids on Metal Surfaces – on-surface metathesis on Ag(111) — •Stephen Massicot[1], Tomoya Sasaki[2], Matthias Lexox[1], Sunghwan Shin[1], Florian Maier[1], Susumu Kuwabata[2], and Hans-Peter Steinrück[1] — [1]Lehrstuhl für Physikalische Chemie 2, Friedrich-Alexander-Universität Erlangen-Nürnberg — [2]Department of Applied Chemistry, Graduate School of Engineering, Osaka University

Ionic liquids (ILs) are salts that are composed of poorly coordinating ions and thus exhibit melting points typically well below 100°C. Thin films of ILs are of utmost interest in many applications such as in catalysis and electrochemistry. In this context, we investigate mixed ultrathin films of the protic IL diethylmethylammonium trifluoromethanesulfonate ([dema][TfO]) and the aprotic IL 1-methyl-3-octylimidazolium hexafluorophosphate ($[C_8C_1Im][PF_6]$) on a Ag(111) surface. The films are prepared by *in situ* physical vapor deposition in UHV. The molecular composition of the IL/solid and IL/vacuum interfaces is studied by angle-resolved and temperature-programmed X-ray photoelectron spectroscopy. We observe thermally induced phenomena of ion exchange and preferential enrichment at these interfaces. Furthermore, ions desorb selectively which leads to the on-surface formation of a new IL by metathesis at the IL/metal interface.

Supported through an ERC Advanced Investigator Grant (ILID 693398) to HPS and by the DFG through SFB 1452.

O 45.9 Wed 17:45 H4

SCILL model catalysis - butadiene hydrogenation studied with molecular beam techniques — •Leonhard Winter, Stephen Massicot, Afra Gezmis, Florian Maier, and Hans-Peter Steinrück — Lehrstuhl für Physikalische Chemie II, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), Egerlandstr. 3, 91058 Erlangen

Ionic Liquids (ILs) are low temperature melting salts, often liquid even below room temperature. In “Solid Catalyst with Ionic Liquid Layer (SCILL)” systems, IL thin films are used to coat the catalytically active metal and therefore, modify the reactivity and the selectivity of the catalyst. Selectivity is crucial e.g., in the industrially important hydrogenation of 1,3-butadiene to butenes since further hydrogenation to *n*-butane must be suppressed.

We used a supersonic molecular beam to investigate the dynamical details of this reaction in a UHV model study. The method of King and Wells was used to measure sticking coefficients of 1,3-butadiene and 1-butene on bare Pt(111) and Pt(111) coated with ultrathin layers of the IL 1,3-dimethylimidazolium bis[(trifluoromethyl)sulfonyl]imide ($[C_1C_1Im][Tf_2N]$). The hydrogenation reaction was also modelled by pre-adsorbing and co-feeding hydrogen gas.

Supported by the DFG through the Collaborative Research Center (CRC)/ Sonderforschungsbereich (SFB) 1452.

O 46: New Methods and Developments 3: Theory

Time: Wednesday 15:00–17:00 Location: H6

O 46.1 Wed 15:00 H6

Atomistic and coarse-grained modelling of liquid-liquid and liquid-gas interfaces — •Jakob Filser[1,2], Karsten Reuter[1,2], and Harald Oberhofer[1,3] — [1]Chair of Theoretical Chemistry, TU Munich — [2]Theory Department, Fritz Haber Institute Berlin — [3]Chair for Theoretical Physics VII, University of Bayreuth

Modelling of dielectric interfaces remains a central challenge in computational chemistry. Liquid-liquid and liquid-gas interfaces have so far received relatively little attention, compared to solid surfaces. We present a new method to incorporate solvation effects into density-functional theory calculations of large organic adsorbates at liquid-liquid and liquid-gas interfaces. Simulating a large number of solvent molecules explicitly at this first-principles level is not computationally tractable. We therefore resort to an implicit solvation approach, treating the solvent as a structureless dielectric medium. Specifically, we advance the multipole-expansion method, in which we model the interface as the boundary of two semi-infinite media with different permittivity. Additionally, we introduce a piecewise expansion of the dielectric response. While the previous version of the MPE method could solve the electrostatic problem only for small solute molecules up to ≈ 10 non-hydrogen atoms, this development now allows us to simulate larger solutes with an overall non-convex hull, with electrostatic interaction energies converged up to few meV. Validating first results of our model for octanoid acid at a water-gas interface by explicit force-field level molecular dynamics simulation provides insight into the role of the atomistic structure of the solvent in the adsorption.

O 46.2 Wed 15:15 H6

Quantum feedback at the solid-liquid interface: flow-induced electronic current and negative friction — •Baptiste Coquinot[1,2], Lydéric Bocquet[1], and Nikita Kavokine[2] — [1]Laboratoire de Physique de l'École Normale Supérieure, ENS, Université PSL, CNRS, Paris, France — [2]Center for Computational Quantum Physics, Flatiron Institute, New York, NY 10010, USA

An electronic current driven through a conductor can induce a current in another conductor through the famous Coulomb drag effect. Similar phenomena have been reported at the interface between a moving fluid and a conductor, but their interpretation has remained elusive. Here, we develop a quantum-mechanical theory of the intertwined fluid and electronic flows, taking advantage of the non-equilibrium Keldysh framework. We predict that a globally neutral liquid can generate an electronic current in the solid wall along which it flows. This hydrodynamic Coulomb drag originates from both direct Coulomb interactions and interactions mediated by the solid's phonons. We derive explicitly the Coulomb drag current in terms of the solid's electronic and phononic properties, as well as the liquid's dielectric response, a result which quantitatively agrees with recent experiments at the liquid-graphene interface. Furthermore, we show that the current generation counteracts momentum transfer from the liquid to the solid, leading to a reduction of the hydrodynamic friction coefficient through a quantum feedback mechanism. Our results provide a roadmap for controlling nanoscale liquid flows at the quantum level, and suggest strategies for designing materials with low hydrodynamic friction.

O 46.3 Wed 15:30 H6

A fully periodic treatment of the chemisorption function for the analysis of adsorbate-substrate interactions — •Simiam Ghan[1], Karsten Reuter[1], and Harald Oberhofer[2] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany. — [2]University of Bayreuth, Bayreuth, Germany

We discuss the extension of our improved Projection-Operator Diabatization scheme POD2GS[1] to the study of electronic coupling on surfaces within periodic density-functional theory (DFT) simulations. Using POD2GS, we calculate diabatic electronic couplings H_{ab} between adsorbate and surface bands throughout k-space. It becomes thus possible to directly calculate the chemisorption function of Newns and Anderson[2], a weighted density of states which appears often in the theoretical description of surface interactions.

Interpreting the chemisorption function within the Fermi Golden Rule yields ultrafast (spin)electron transfer lifetimes directly comparable to experiment, as we demonstrate for the case of core-excited Argon monolayers on ferromagnetic substrates Fe(110), Co(0001) and Ni(111)[3]. Our scheme reveals the importance of sampling the Brillouin zone to gain accurate and convergent electron transfer rates, and offers an array of further applications.

[1] J. Chem. Theory Comput. 2020, 16, 12, 7431-7443.
[2] Phys. Rev. 1969, 178, 1123.
[3] Phys. Rev. Lett. 2014, 112, 086801.

O 46.4 Wed 15:45 H6

A Revised Fourth-Generation Neural Network Potential for the Accurate Representation of Multiple Charge States — •Alexander Knoll, Tsz Wai Ko, and Jörg Behler — Georg-August-Universität Göttingen, Institut für Physikalische Chemie, Theoretische Chemie, Göttingen, Germany

Machine learning potentials (MLPs) have become a mature tool for large-scale atomistic simulations in chemistry and materials science. Recently, a fourth-generation high-dimensional neural network potential (4G-HDNNP) has been introduced, in which the atomic charges are determined in a charge equilibration step enabling the description of long-range charge transfer. The quality of the charge prediction depends on environment-dependent electronegativities expressed by atomic neural networks, which poses a challenge for structures with differing total charges but nearly-identical nuclear positions. Here, we propose a generalized method applicable to these situations, and for a series of model systems we demonstrate that this extension leads to additional flexibility of the atomic electronegativities, ultimately resulting in more accurate atomic charges, energies, and forces.

O 46.5 Wed 16:00 H6

Machine learning potentials for complex aqueous systems made simple — •Christoph Schran[1], Fabian L. Thiemann[1,2], Patrick Rowe[1], Erich A. Müller[2], Ondrej Marsalek[3], and Angelos Michaelides[1] — [1]University of Cambridge, UK — [2]Imperial College London, UK — [3]Charles University, Czech Republic

Simulation techniques based on accurate and efficient representations of potential energy surfaces are urgently needed for the understanding of complex systems such as solid-liquid interfaces. Here we present a machine learning framework that enables the efficient development and validation of models for complex aqueous systems. Instead of trying to deliver a globally optimal machine learning potential, we propose to develop models applicable to specific thermodynamic state points in a simple and user-friendly process. After an initial ab initio simulation, a machine learning potential is constructed with minimum human effort through a data-driven active learning protocol. Such models can afterward be applied in exhaustive simulations to provide reliable answers for the scientific question at hand or to systematically explore the thermal performance of ab initio methods. We showcase this methodology on a diverse set of aqueous systems. Highlighting the accuracy of our approach with respect to the underlying ab initio reference, the resulting models are evaluated in detail with an automated validation protocol that includes structural and dynamical properties and the precision of the force prediction of the models. Finally, we demonstrate the capabilities of our approach for providing insight into the solid-liquid interface for various systems.

O 46.6 Wed 16:15 H6

Quantifying the breakdown of electronic friction theory during molecular scattering of NO from Au(111) — •Connor L. Box[1], Yaolong Zhang[2], Rongrong Yin[2], Bin Jiang[2], and Reinhard J. Maurer[1] — [1]Department of Chemistry, University of Warwick, United Kingdom — [2]Department of Chemical Physics, University of Science and Technology of China, Hefei, China

The Born-Oppenheimer approximation fails to capture the extent of multiquantum vibrational energy loss recorded during molecular scattering from metallic surfaces.[1] NO scattering on Au(111) has been one of the most studied examples in this regard, providing a testing ground for developing various nonadiabatic theories. The exact failings compared to experiment and their origin from theory are not established for any system, particularly since dynamic properties are affected by many compounding simulation errors, of which the quality of nonadiabatic treatment is just one. We use a high-dimensional machine learning representation of the energy and the electronic friction tensor to minimize errors that arise from quantum chemistry.[1,2]

This allows us to perform a comprehensive quantitative analysis of the performance of molecular dynamics with electronic friction in describing state-to-state scattering. We find that electronic friction theory accurately predicts elastic and single-quantum energy loss, but underestimates multi-quantum energy loss and overestimates molecular trapping at high vibrational excitation. Our analysis reveals potential remedies to these issues. [1] C. L. Box & Y. Zhang et al, JACS Au, 2020 [2] R. Yin et al, J. Phys. Chem. Lett, 2019

O 46.7 Wed 16:30 H6

Accurate computation of chemical contrast in field ion microscopy — •Shalini Bhatt — Max Planck Institut für Eisenforschung GmbH Düsseldorf Germany

Field ion microscopy (FIM) was the first microscopy technique to image individual atoms on a metal surface with near atomic spatial resolution. In short an imaging gas (e.g. He, Ne) is ionized above a surface subject of a few field 10^{10} V/m. The imaging contrast is dominated by the ionization probability at 5-10 Å above the surface. To simulate this within density-functional theory (DFT), we adapt the Tersoff-Hamann theory known from scanning tunneling microscopy (STM).

The gigantic electric field leads to very fast decay of wavefunctions into the vacuum. At the ionization height, they run into a regime that is dominated by numerical noise. To address this noise challenge inherent to any global-scale Kohn-Sham solver employed in DFT codes, wavefunction tails must be recomputed. To solve 3D Schrödinger equation at local scale we develop the extrapolated tail via reverse algorithm (EXTRA). The decaying tails are obtained by reverse integration (from outside in) using a Numerov-like algorithm. The starting conditions are then iteratively adapted to match the values of planewave DFT wavefunctions close to the surface. We demonstrate chemical contrast for Ta and W at Ni surface using this new technique.

O 46.8 Wed 16:45 H6

Designing Covalent Organic Frameworks Through Active Machine Learning — •Yuxuan Yao[1,2], Christian Kunkel[3], Karsten Reuter[3], and Harald Oberhofer[2] — [1]Chair for Theoretical Chemistry and Catalysis Research Center, Technical University Munich — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft — [3]Chair for Theoretical Physics VII, University of Bayreuth

Covalent organic frameworks(COFs) are a class of materials with potential applications in many fields such as catalysis, sensing, or optoelectronics. It is well known that their design space is far too large to sample one by one. Focusing on their electronic properties, we modify an earlier active machine learning(AML) approach that explores the molecular design through the use of surrogate models for charge injection and transport descriptors. In this method, the Gaussian Process Regression(GPR) and AML are combined to train the molecular space. This way we ensure that only promising molecules or candidates that are very different from already explored ones have their descriptors evaluated on a comparatively expensive quantum mechanical level. Specifically, we modify molecular generation rules in order to produce three-fold rotational symmetric candidates molecules for use in COFs. In the future this approach can be generalized for any other symmetries, to potentially even allow for 3-dimensional network generation. The generation of a candidate space with well defined symmetries together with AML ensures a high efficiency in the detection of promising COFs with superior charge conduction properties and demonstrates the utility of this approach.

O 47: Focus Session: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy 1

Magnetic single atoms and molecules are intensively studied as the smallest building blocks for potential applications in spintronic devices and quantum information processing. Detecting and controlling single spin states and their spin interactions require both high energy resolutions and atomic scale imaging capability. Scanning tunneling microscopes provide not only the spatial resolutions but also the bottom up approach to build magnetic structures atom by atom. Recently, unprecedented spin sensitivities have been reached by functionalizing the STM tips or by combining STM with electron spin resonance. The goal of this symposium is to highlight the recent developments in the rapidly evolving field of atomic scale sensing and quantum control of spins on surfaces. Researchers that use STM to investigate atomic or molecular spins on surfaces will present their most recent results. Sharing technical advances and addressing current issues will create synergies to foster future progress in this field and a deeper understanding of the underlying physics.

Organizer: Andreas Heinrich (Center for Quantum Nanoscience, Seoul, Republic of Korea)

Time: Wednesday 15:00–18:00 Location: S051

Topical Talk O 47.1 Wed 15:00 S051

Quantum control of multi-spin architectures on a surface — •Yujeong Bae — Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS), 03760 Seoul, South Korea — Department of Physics, Ewha Womans University, Seoul 03760, South Korea

The spin-polarized scanning tunneling microscopy (STM) combined with electron spin resonance (ESR) [1] enables us to achieve single spin sensitivity with atomic precision. Employing ESR-STM, a single spin on a surface can be coherently probed and controlled [1,2], where the magnetic tip is positioned directly on the target spin. Here, we demonstrate a new approach to coherently control multi-electron spins in a quantum spin architecture crafted atom-by-atom. We found the remote spins, which are outside the tunnel junction, can be controlled by the local oscillating magnetic fields created by a single-atom magnet placed next to them in oscillating electric fields. The readout of multi-electron spins is achieved by a sensor atom weakly coupled to them. The resonances of the sensor spin are separated in the frequency domain so that we can independently and simultaneously control the sensor and remote spins. Our work shows the enhanced coherent properties of the remote spins as well as fast controlled operations of multi-electrons in an all-electrical fashion. Our development widens the approaches to the multi-spin control in tailored spin structures on a surface.

[1] S. Baumann et al., Science, 350 (2015) 417.
[2] K. Yang et al., Science, 366 (2019) 509.

O 47.2 Wed 15:30 S051

Longitudinal and transverse electron paramagnetic resonance in a scanning tunneling microscope — •Tom S. Seifert[1], Stepan Kovarik[2], Pietro Gambardella[2], and Sebastian Stepanow[2] — [1]FU Berlin — [2]ETH Zürich

Combining scanning tunneling microscopy (STM) with electron-paramagnetic resonance (EPR) allows for sensitive probing magnetic interactions at atomic scales [1]. However, the experimental requirements for driving the EPR transitions are not obvious [2,3]. In-depth understanding of what drives EPR-STM is mandatory to explore novel material systems with high sensitivity. Here, we acquire and model EPR spectra of single Fe and hydrogenated Ti atoms on bilayer MgO on Ag using a radio frequency (RF) antenna close to the STM junction [4]. We investigate in a systematic way the impact of RF excitation strength and tunneling parameters on the EPR signal and find strong evidence for a piezoelectric coupling mechanism [5]. Specifically, transverse magnetic field gradients drive the spin-1/2 hydrogenated Ti, whereas longitudinal magnetic field gradients drive the spin-2 Fe. Finally, we demonstrate how the choice of specific tip-sample distances allows one to minimize the impact of tip magnetic fields on the EPR-

STM measurements thereby excluding a major experimental uncertainty when determining single-atom magnetic moments [6].

[1] S. Baumann et al., Science 350 (2015) [2] K. Yang et al., PRL 122 (2019) [3] P. Willke et al., Nano Lett. 19 (2019) [4] T.S. Seifert et al., PRR 2 (2020) [5] T.S. Seifert et al., Sci. Adv. 6 (2020) [6] T.S. Seifert et al., PRR 3 (2021)

O 47.3 Wed 15:45 S051

Experimental Determination of a Single Atom Ground State Orbital through Hyperfine Anisotropy — •Laëtitia Farinacci[1], Lukas M. Veldman[1], Philip Willke[2], and Sander Otte[1] — [1]Delft University of Technology, Delft, The Netherlands — [2]Karlsruhe Institute of Technology, Karlsruhe, Germany

Electron spin resonance has long been a powerful tool for electronic analysis. Its recent combination with scanning tunneling microscopy [1] gives exceptional energy resolution for the investigation of the magnetism of single atoms in addition to a very good characterization of their spatial surroundings.

Here, we provide a full angle-dependent investigation of the anisotropy of the hyperfine splitting of single Ti atoms on MgO/Ag(100). We find that the anisotropy of the hyperfine splitting is related to that of the g factor: spin-orbit coupling leads to a partially unquenched angular momentum which couples to the electron spin and thereby affects its interaction both with an external magnetic field and the nuclear spin of the nucleus. Combining the symmetry properties of the atoms binding site with a simple point charge model, we provide a method to predict the shape of the ground state orbital of the Ti atom. Relying on experimental values only, this analysis paves the way for a new protocol for electronic structure analysis for spin centers on surfaces.

[1] Baumann et al., Science 350 (2015)

O 47.4 Wed 16:00 S051

Anisotropic hyperfine interaction of surface-adsorbed single atoms — •Jinkyung Kim[1,2], Kyungju Noh[1,2], Yi Chen[1,3], Christoph Wolf[1,3], Andreas Heinrich[1,2], and Yujeong Bae[1,2] — [1]Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS), Seoul 03760, South Korea — [2]Department of Physics, Ewha Womans University, Seoul 03760, South Korea — [3]Ewha Womans University, Seoul 03760, Republic of Korea

Hyperfine interactions between electron and nuclear spins provide sensitive probes to the chemical environment of atoms, molecules, and crystal defects as well as an alternative way to control the nuclear spins. While a variety of experimental techniques have been applied to the detection and control of single nuclear spins in various environments, simultaneous investigations of the single nuclear spin*s resonance and the atomic scale imaging are more demanding. Using the electron spin resonance (ESR) technique in a scanning tunneling microscope (STM), we investigated hyperfine interaction of hydrogenated titanium (Ti) on MgO/Ag(100) and its local environment at the atomic scale. By means of atom manipulation in vector magnetic fields, we identified the hyperfine interaction of Ti along three principal axes, which shows a large anisotropy of hyperfine interaction. As a sensitive probe of chemical environment, the observed hyperfine interaction reflects the anisotropic orbital configuration of the electronic ground state, which is further supported by the density functional theory calculations.

O 47.5 Wed 16:15 S051

Electron Paramagnetic Resonance of individual Alkali Metal Atoms and Dimers on Ultrathin MgO — •Stepan Kovarik[1], Roberto Robles[2], Richard Schlitz[1], Tom Sebastian Seifert[1,3], Nicolas Lorente[2,4], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]Department of Materials, ETH Zurich, Switzerland — [2]Centro de Física de Materiales, San Sebastian, Spain — [3]Department of Physics, FU Berlin, Germany — [4]Donostia International Physics Center, San Sebastian, Spain

Electron paramagnetic resonance (EPR) provides unique insight into the chemical structure and magnetic properties of dopants in oxide and semiconducting materials that are of interest for applications in electronics, catalysis, and quantum sensing. We demonstrate that EPR in combination with scanning tunneling microscopy (STM) [1, 2] allows for probing the spin and charge state of alkali metal atoms on an ultrathin magnesium oxide layer on a Ag(100) substrate. We identify a magnetic moment of 1 μ_B for Li_2, LiNa, and Na_2 dimers corresponding to spin radicals with a charge state of +1e [3]. Individual alkali atoms have the same charge state and no magnetic moment. The ionization of the adsorbates is attributed to the charge transfer of one electron through the oxide to the metal substrate. Our work highlights the potential of EPR-STM to provide insight into dopant atoms, which are relevant for the control of the electrical properties of surfaces and represent a suitable platform for studying quantum nanomagnets.

[1] S. Baumann et al., Science 350 (2015) [2] T. S. Seifert et al., PRR 2 (2020) [3] S. Kovarik et al., Nano Lett. 22 (2022)

Topical Talk O 47.6 Wed 16:30 S051

Free coherent evolution of a coupled atomic spin system initialized by electron scattering — •Sander Otte[1], Lukas Veldman[1], Laetitia Farinacci[1], Rasa Rejali[1], Rik Broekhoven[1], Jeremie Gobeil[1], David Coffey[1], and Markus Ternes[2,3] — [1]Delft University of Technology, Delft, The Netherlands — [2]RWTH Aachen University, Aachen, Germany — [3]Forschungszentrum Jülich, Jülich, Germany

Full insight into the dynamics of a coupled quantum system depends on the ability to follow the effect of a local excitation in real-time. Here, we trace the free coherent evolution of a pair of coupled atomic spins by means of scanning tunneling microscopy. Rather than using microwave pulses, we use a direct-current pump-probe scheme to detect the local magnetization after a current-induced excitation performed on one of the spins. By making use of magnetic interaction with the probe tip, we are able to tune the relative precession of the spins. We show that only if their Larmor frequencies match, the two spins can entangle, causing angular momentum to be swapped back and forth. These results provide insight into the locality of electron spin scattering and set the stage for controlled migration of a quantum state through an extended spin lattice.

O 47.7 Wed 17:00 S051

Controlled migration of a coherent spin excitation through atomically assembled nanomagnets — •Lukas Veldman[1], Laëtitia Farinacci[1], Rasa Rejali[1], Rik Broekhoven[1], Jeremie Gobeil[1], David Coffey[1], Markus Ternes[2,3], and Sander Otte[1] — [1]TU Delft, Delft, The Netherlands — [2]RWTH Aachen, Aachen, Germany — [3]Forschungszentrum Jülich, Jülich, Germany

Tracing single coherent spin excitations in low dimensional nanomagnets has been a long standing goal in experimental solid state physics. Scanning tunneling microscopy offers a promising platform to chase this goal due to its capability to build nanomagnets atom-by-atom and address each atomic spin individually. Here, we show the possibility of inducing a single spin-flip excitation using electron scattering and the measurement of the resulting flip-flop interaction between two atomic spins [1]. Next, we use the same principle to observe coherent spin dynamics in multiple atomically assembled nanomagnets. By tuning the interaction between tip and nanomagnet, we are able to address different magnetic resonances in each atomic structure. In engineered branched structures, the spin excitation can be sent to different directions using the tip interaction. These techniques can serve as a platform for dynamical quantum simulation and can form a foundation for atomically assembled spintronic applications.

[1] Veldman, L. M., Farinacci, L., Rejali, R., ... & Otte, A. F. (2021) Science, 372

O 47.8 Wed 17:15 S051

Electric field control of spin transitions in a single molecule using ESR-STM — •Maneesha Ismail, Piotr Kot, and Christian R. Ast — Max-Planck-Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart, Germany

Since the demonstration of the control of spins on the atomic scale, the technique of ESR-STM has been used extensively to explore the field of spintronics. Here, we present an extension to the ESR-STM parameter space, which uses the bias voltage to tune the energy of the Zeeman transition. We emonstrate electronic control of spin resonance transitions in a single TiH molecule. We were able to observe a strong dependency of the g-factor along with a tip-field shift as function of the electric field. Finally, we manipulate a TiH dimer by continuously changing the bias voltage such that the dimer moves through an avoided crossing of the energy levels. This could be an important step towards pump-probe control of spin states through the bias voltage and opens new possibilities for coherent manipulation.

O 47.9 Wed 17:30 S051

Isotope detection inside single molecules in scanning-probe based electron spin resonance — •Lisanne Sellies, Raffael Spachtholz, Philipp Scheuerer, and Jascha Repp — University of Regensburg, Regensburg, Germany

Electron spin resonance (ESR), a versatile technique to study materials with unpaired electrons, was recently combined with scanning tunneling microscopy (STM), bringing atomic-scale spatial resolution to ESR [1]. ESR-STM even allowed detecting strong hyperfine interactions between the single electron spin under study with the atom's nuclear spin [2], due to its largely improved energy resolution compared to conventional STM, reaching the nano-electron-volt regime. This energy resolution could even be improved further if the tunneling current as a read-out was avoided, since this current is the dominating decoherence source for the probed electron spin.

We propose, therefore, a new ESR scanning probe method based on atomic force microscopy (AFM). Since our technique does not rely on the tunneling current to read-out the ESR signal, we increase the coherence times of the electron spins and, consequentially, the energy resolution. Therefore, we can resolve the hyperfine interaction inside organic molecules and distinguish molecules only differing in the isotopic composition. Thus, our technique allows the chemical fingerprinting of molecules and their surroundings.

References: [1] S. Baumann et al., Science 350, 417 (2015) [2] P. Willke et al., Science 362, 336 (2018)

O 47.10 Wed 17:45 S051

A home-built scanning tunneling microscope combined with electron spin resonance — •Andreas Heinrich — Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Korea

Scanning tunneling microscopy is a powerful tool to characterize the electronic and magnetic properties of atomic scale structures on a surface. Recently, improved spectral energy resolution has been achieved by functionalizing the STM tip with a well-characterized molecule at the apex [1,2] or by combining electron spin resonance with STM (ESR-STM) [3]. Here, we present the design and operation of an optimized, home-built ESR-STM with a specially designed mechanical damper, a Joule-Thomson refrigerator, and 2-axes vector magnets. This system provides outstanding performance of STM for nanoscale measurements. We further describe a new design of a microwave antenna to increase the transmission of RF voltages to the junction, which performs better than the direct connection to the tip. Applying RF power through the antenna terminated at 50 Ohm results in the reduction of standing waves and increases the available frequency range (5-40 GHz), which allows us to measure ESR of surface spins at elevated temperatures up to 10 K. References: [1] M. Ormaza et al., Nano let. 17, 1877-1882 (2017) [2] G. Czap et al., Science, 364, 670-673 (2019) [3] S. Baumann et al., Science, 350, 417-420 (2015)

O 48: 2D Materials 1: Electronic Structure of Transition Metal Dichalcogenides

Time: Wednesday 15:00–17:30 Location: S052

O 48.1 Wed 15:00 S052

the stability of point defects in 2D monolayer transition metal dichalcogenides and their impact on the electronic structure — •alaa akkoush[1,2] and mariana rossi[1,2] — [1]Fritz Haber Institute of the Max Planck Society, Faradayweg 4-6, Berlin — [2]MPI for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22765 Hamburg

Defects can strongly influence the electronic, optical and mechanical properties of 1D materials. However, their stability and distribution under different conditions of temperature, pressure and strain are not well characterized from an atomistic perspective. We have investigated the structural and electronic properties, as well as the thermodynamic stability of point defects (vacancies and adatoms) in monolayer transition metal dichalcogenides MX_2 with M=Mo/W and X= S/Se, through density-functional theory (DFT) simulations with hybrid exchange correlation functional, as implemented in the all-electron package FHI-aims [1]. These calculations are carried out using a supercell approximation to model localised defects using periodic boundary conditions. We show quantitatively that X adatom is most favorable in rich X conditions while in poor X environment X monovacancy is most favorable. Interestingly, an interplay between adatom and divacancies takes place as temperature increases. To gauge the importance of vibrational free energy contributions on the engineering of gap states in the 2D monolayers, we compare the formation energies of point defects with an adsorbed F6TNAP at various thermodynamic conditions.

[1] S.V. Levchenko , et al., *Comp. Phys. Comm.* **192** 60-69 (2015)

O 48.2 Wed 15:15 S052

Non-linear optical response of TMD Nanotubes using Wannier interpolation — •Jyoti Krishna and Julen Ibañez-Azpiroz — Centro de Fisica de Materiales (CSIC-UPV/EHU), Donostia-San Sebastian, Spain

Single crystals lacking an inversion center display a non-vanishing second-order response known as shift current- a dominant contribution to the bulk photovoltaic effect. There has been a renewed interest in novel materials with large non-linear absorption capabilities[1]. Here we focus on transition metal dichalcogenide nanotubes, which have recently exhibited a short-circuit intensity showing orders of magnitude enhancement over the monolayer value[2]. We systematically explore the implications of the different chiral indexes on both linear and quadratic optical responses for single-walled TMD NT employing the Wannier interpolation technique. We classify the allowed symmetry and calculate the dependence of the magnitude of the response as a function of NT diameter and chirality. Finally, we discussed our results in the context of the experimental measurements.

Funding provided by the European Union's Horizon 2020 research and innovation programme under the European Research Council (ERC) grant agreement No 946629. (1) L. Z Tan, F. Zheng, F. Wang and A. M. Rappe, npj Comput. Mater. 2, 16026 (2016). (2) Y. J. Zhang et. Al Nature 570, 349 (2019). (3) G. Pizzi et. Al, J. Phys. Cond. Matt 32, 165902 (2020).

O 48.3 Wed 15:30 S052

Structural, electronic and optical properties of strained MoS_2 — •Jan-Hauke Graalmann and Michael Rohlfing — Institute of Solid State Theory, University of Münster, 48149 Münster, Germany

Experimental and theoretical studies have shown that the spectrum of MoS_2 changes when the system gets strained. Stretching a monolayer leads to a shift of the optical absorption spectrum to lower energies. In case of the bilayer under hydrostatic stress, the interlayer interaction plays a major role.

In this talk we investigate the behaviour of a MoS_2 monolayer and bilayer theoretically using DFT, *GdW* and the Bethe-Salpeter equation. Our results show a transition of the monolayer from a direct semiconductor to an indirect one when the in-plane lattice constant is increased. Furthermore, the fundamental gap at the K point is reduced while the binding energies of the A and B excitons remain approximately constant. These effects lead to an effective shift of the excitation energies of both excitons towards lower energy with similar gauge factors.

We also find a significant influence of interlayer interaction within the bilayer. The effect of a decreasing gap by biaxial shrinking of each single layer under an external hydrostatic pressure gets counterbalanced by the reduction of the interlayer distance.

O 48.4 Wed 15:45 S052

Excitons in TMDC Bilayers — •Jan Nellesen and Michael Rohlfing — Westfälische Wilhelms-Universität, Münster, Germany

Semiconducting Transition-Metal-Dichalcogenides (TMDCs) have gained a lot of attention in the past few years due to possible new applications in optoelectronics. In particular, the excitonic properties of TMDC mono- and bilayers have been studied extensively.

This work focuses on interlayer excitons in TMDC heterostructures which combine large binding energies with relatively long lifespans. The exciton energies are investigated in different systems using the Bethe-Salpeter-Equation.

In order to describe excitons in twisted bilayers, which exhibit long-range moiré structures, *ab initio* approaches are no longer feasible because of their high computational demand. With the goal of studying excitons in such systems in mind, the TMDC bandstructure is modeled within a tight-binding approximation. For describing the electron-hole interaction, a model potential is used.

O 48.5 Wed 16:00 S052

Unoccupied electronic states of 1T-TiSe$_2$: Band dispersions and CDW-induced changes at $\overline{\Gamma}$ — •Patrick Geers, Marcel

Holtmann, and Markus Donath — University of Münster, Germany

The transition metal dichalcogenide 1T-TiSe$_2$ shows a phase transition into a charge density wave (CDW) below a critical temperature T_{CDW} [1]. This transition leads to a half-sized Brillouin zone and backfolded electronic bands. For the occupied electronic structure, photoemission results show a backfolding of Se-4$p_{x,y}$ valence bands below T_{CDW} from $\overline{\Gamma}$ to $\overline{M}$ [2].

We present angle-resolved inverse-photoemission (IPE) measurements for the unoccupied electronic structure above and below T_{CDW}. Our data for the $\overline{\Gamma}\,\overline{M}$ azimuth resemble literature data for the energy vs. momentum dispersion of Ti-3d states as well as an image-potential-induced surface state [3]. In addition, we report on IPE measurements for the $\overline{\Gamma}\,\overline{K}$ azimuth. Finally, we give special attention to the changes in the electronic structure, which are caused by the CDW phase transition. Measurements around $\overline{\Gamma}$ below T_{CDW} show modifications in the spectra which are attributed to backfolded electronic bands from $\overline{M}$ to $\overline{\Gamma}$ as a direct consequence of the CDW phase.

[1] Di Salvo *et al.*, Phys. Rev. B **14**, 4321 (1976).
[2] Watson *et al.*, Phys. Rev. Lett. **122**, 076404 (2019).
[3] Drube *et al.*, J. Phys. C **20**, 4201 (1987).

O 48.6 Wed 16:15 S052

Nontrivial Doping Evolution of Electronic Properties in Ising-Superconducting Alloys — •Wen Wan[1], Darshana Wickramaratne[2], Paul Dreher[1], Rishav Harsh[1], Igor I. Mazin[3], and Miguel Ugeda[1] — [1]Donostia International Physics Center (DIPC), Paseo Manuel de Lardizábal 4, 20018 San Sebastián, Spain — [2]Center for Computational Materials Science, U.S. Naval Research Laboratory, Washington, DC 20375, USA — [3]Department of Physics and Astronomy, George Mason University, Fairfax, VA 22030, USA

TMDs offer unprecedented versatility to engineer 2D materials with tailored properties to explore novel structural and electronic phase transitions. Here, we present the atomic-scale evolution of the electronic ground state of a monolayer of Nb1-δMoδSe2 ($0<\delta<1$) using STM/STS measurements at 300 mK. We investigate the atomic and electronic structure of this 2D alloy throughout the metal to semiconductor transition (NbSe2 to MoSe2). Our measurements let us extract the effective doping of Mo atoms, the bandgap evolution and the band shifts, which are monotonic with δ. Furthermore, we demonstrate that collective phases (CDW and superconductivity) are remarkably robust against disorder. We further show that the superconducting TC changes non-monotonically with doping. This contrasting behavior in the normal and superconducting state is explained using first-principles calculations. We show that Mo doping decreases the DOS at EF and the magnitude of pair-breaking spin fluctuations as a function of Mo content. (1) W. Wan, et al. Advanced Materials, accepted (2022).

O 48.7 Wed 16:30 S052

Surface spin texture derived from a single mirror plane of WTe$_2$ — Tristan Heider[1], Gustav Bihlmayer[2], Jakub Schusser[3,4], Friedrich Reinert[4], Jan Minar[3], Stefan Blügel[2], Claus M. Schneider[1], and •Lukasz Plucinski[1] — [1]PGI-6 Forschungszentrum Jülich — [2]PGI-1 Forschungszentrum Jülich — [3]University of West Bohemia, Pilsen, Czech Republic — [4]Experimentelle Physik VII, Universität Würzburg

WTe$_2$ is an important semi-metallic quantum material that exhibits non-saturating magnetoresistance and potentially hosts Weyl type-II nodes [1]. Through the laser-driven spin-polarized ARPES Fermi surface mapping, we demonstrate highly asymmetric spin textures of electrons photoemitted from the surface states of WTe$_2$. Such asymmetries are not present in the initial state spin textures, which are bound by the time-reversal and crystal lattice mirror plane symmetries. The findings are reproduced qualitatively by theoretical modeling within the one-step model photoemission formalism, while a simple toy-model suggests that a similar effect shall be observed in other materials with low symmetry.

Our spin-polarized maps with detail comparable to the previous spin-integrated maps [2] have been measured using the newly developed high-resolution instrument at PGI-6 in Jülich that is based on a hemispherical analyzer with the scanning lens, an exchange-scattering spin detector, and a *cw* 6 eV laser.

[1] P. K. Das et. al. Electron. Struct. 1, 014003 (2019) and refs. therein. [2] F. Y. Bruno et al., Phys. Rev. B 94, 121112 (2016).

O 48.8 Wed 16:45 S052

Effect of gold substrate on the excitonic properties of MoS$_2$: a final state sum frequency spectroscopy study — •Tao Yang, Erik Pollmann, Stephan Sleziona, Marika Schleberger, Richard Kramer Campen, and Yujin Tong — Fakultät für Physik Universität Duisburg-Essen, 47057 Duisburg, Germany

Monolayer transition metal dichalcogenides(TMDCs) are promising candidates for applications in electronics, optoelectronics, and photocatalysts due to their good thermodynamic stability, ease of preparation, tunable bandgap in the visible region, and pronounced activity for photoelectrochemical water splitting. To take advantage of their excellent properties to build the devices and catalysts, a metallic surface is required to combine them. However, compared to the well-explored properties of monolayer TMDCs on dielectric substrates, less attention has been paid to these properties on metal substrates due to the challenges associated with linear spectroscopies probing, such as quenching. Here we use a final state sum frequency spectroscopy(FSSFG) to study the optical properties of MoS$_2$ exfoliated on a gold surface. Relative to the well-known six-fold symmetry of MoS$_2$ on SiO$_2$, the azimuthal dependent FSSFG show significantly different patterns at different polarization combinations. The current study provides important insights into the significant changes in electronic structure when MoS$_2$ comes into contact with gold, and the possibility to selectively switch resonance on and off through azimuthal tuning, as revealed by the robust FSSFG.

O 48.9 Wed 17:00 S052

Time-resolved momentum microscopy of moiré interlayer excitons in twisted TMD heterostructures — •David Schmitt[1], Jan Philipp Bange[1], Wiebke Bennecke[1], AbdulAziz AlMutairi[2], Giuseppe Meneghini[3], Daniel Steil[1], R. Thomas Weitz[1], Sabine Steil[1], G. S. Matthijs Jansen[1], Samuel Brem[3], Ermin Malic[3], Stephan Hofmann[2], Marcel Reutzel[1], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Friedrich-Hund-Platz 1, Göttingen, Germany — [2]Department of Engineering, University of Cambridge, Cambridge CB3 0FA, U.K. — [3]Fachbereich Physik, Philipps-Universität, 35032 Marburg, Germany

Transition metal dichalcogenides (TMDs) are extensively studied because of their exceptional material properties. The stacking of different TMDs can lead to even more intriguing electronic properties: In type-II band aligned TMD stacks, novel excitonic states can be created where the electron and the hole contribution to the exciton are separated between the van-der-Waals-coupled TMDs. A twist angle between these layers allows further manipulation of the electronic properties and gives rise to moiré-patterns which leads to distinct patterns in momentum space. Here, we make use of our setup for time-resolved momentum microscopy that is perfectly suited to probe these excitonic features with energy- and in-plane momentum resolution. We present femtosecond evaluation of the momentum-fingerprints of the excitonic features that are created on a type II aligned heterostructure. Schmitt *et al.*, arXiv:2112.05011 (2021).

O 48.10 Wed 17:15 S052

***In Operando* Soft X-Ray Photoemission Spectroscopy of 2D Material Devices** — •Alena Nierhauve[1,2], Matthias Kalläne[1,2], Tammo Zimmermann[1], Jens Buck[1,2], Philipp Kagerer[3], Zhansong Geng[4], Chao Zhang[4], Frank Schwierz[4], Martin Ziegler[4], Rok Venturini[5], and Kai Rossnagel[1,2,6] — [1]IEAP, CAU Kiel, 24098 Kiel, Germany — [2]Ruprecht-Haensel-Labor, DESY and CAU Kiel, 22607 Hamburg and 24098 Kiel, Germany — [3]Dep. of Exp. Physics VII, JMU Würzburg, 97074 Würzburg, Germany — [4]Dep. of Electr. Engineering and Information Techn., TU Ilmenau, 98684 Ilmenau, Germany — [5]Jožef Stefan Institute, 1000 Ljubljana, Slovenia — [6]DESY, 22607 Hamburg, Germany

Layered transition-metal dichalcogenides (TMDCs) are a particularly promising platform for low-dimensional electronic devices due to their two-dimensional nature and richness regarding physical phenomena. This includes non-linear conductance behavior due to, e.g., metal-insulator transitions and memristive behavior, which is of particular interest to in-memory computing designs for neuromorphic systems. By combination of micrometer position- and angle-resolved photoemission spectroscopy (μ-ARPES) in the soft X-ray range with *in operando* electrical control, we attempt to study the electronic structure changes concomitant with thickness changes and non-equilibrium conditions in device-like structures based on TMDCs. A portrayal of conductance-governing mechanisms at a fundamental level could help to evolve towards the understanding and engineering of novel band structures, transport phenomena, and device functionality.

O 49: Oxide Surfaces 1

Time: Wednesday 15:00–17:30 Location: S053

O 49.1 Wed 15:00 S053

Revised Chen's derivative rule for efficient simulations of scanning tunneling microscopy: New results on surface oxides — •Krisztián Palotás[1], Yunjae Lee[2], Taehun Lee[2], and Aloysius Soon[2] — [1]Wigner Research Center for Physics, Budapest, Hungary — [2]Yonsei University, Seoul, Republic of Korea

Advanced simulation tools of scanning tunneling microscopy (STM) are vital for the proper understanding of various physical and chemical processes at material surfaces. For this reason Chen's derivative rule for electron tunneling has been revised [1] to build a computationally efficient STM simulation tool, based on ab initio electronic structure. This STM simulation method enables (i) the weighting of tunneling matrix elements of arbitrary tip-orbital composition, (ii) arbitrary tip geometrical orientations to mimic asymmetric tip-sample relations, and (iii) the possibility of quantitative analysis of tip-orbital interference contributions to the tunneling current. Recently, this method has been applied to diverse complex surface oxides [2,3,4], where a better agreement with STM experiments has been achieved than obtained with the Tersoff-Hamann (spherical tip orbital) model.

[1] G. Mándi and K. Palotás, Phys. Rev. B 91, 165406 (2015). [2] T. Lee et al., Nanoscale 11, 6023 (2019). [3] T. T. Ly et al., J. Phys. Chem. C 123, 12716 (2019). [4] Y.-J. Lee et al., Appl. Surf. Sci. 562, 150148 (2021).

O 49.2 Wed 15:15 S053

Hydrogen Atom Scattering at Aluminium Oxide — •Martin Liebetrau and Jörg Behler — Universität Göttingen, Institut für Physikalische Chemie, Theoretische Chemie, Tammannstraße 6, 37077 Göttingen, Germany

The adsorption of atomic hydrogen is important in many fields, from heterogeneous catalysis via hydrogen storage to nuclear fusion. Here, we report molecular dynamics simulations of high-energy hydrogen atom scattering at the α-Al_2O_3(0001) surface. Employing a high-dimensional neural network potential, which allows us to include the full-dimensional thermal motion of the surface atoms, we are able to calculate a large number of trajectories with the accuracy of density-functional theory at a small fraction of the computational costs. Investigating different kinetic energies, surface temperatures and incident angles, we are able to characterize the scattering process in detail.

O 49.3 Wed 15:30 S053

Adsorption of Gases on β-Ga_2O_3 Surfaces — •Jonathan K. Hofmann[1,2], Celina S. Schulze[1], Martin Franz[1], Nipin Kohli[1], Dorothee Rosenzweig[1], Zbignew Galazka[3], and Holger Eisele[1] — [1]Technische Universität Berlin, Institut für Festkörperphysik, Germany — [2]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — [3]Institut für Kristallzüchtung, Germany

β-Ga_2O_3 is a wide band gap material, which is promising for high power and UV (opto-)electronics. The typical n-type doping is controllable via the growth parameters, intentional doping, or post-growth heat treatment. In this contribution, we address the question of its surface properties under typical ambient conditions, i. e., under H_2O and O exposure. The β-Ga_2O_3 single crystals were grown with the Czochralski method [1]. Using Auger electron spectroscopy (AES), and scanning tunnelling microscopy/spectroscopy (STM/STS), we show how the different adsorbed atoms/molecules change the structure and electronic properties of β-Ga_2O_3 (100) and (001) surfaces under UHV-conditions. On the (100) surface, large clusters of H_2O with an undisturbed surface in between were observed. However, STS showed no change in the electronic states. On the (001) surface, oxygen covered almost the complete surface. STS showed that O lifts the band bending inherent in clean β-Ga_2O_3 surfaces.

The project was supported by the Leibnitz Association, Leibnitz Science Campus GraFOX, project C2-3.

[1] Z. Galazka *et al.* ECS J. Solid State Sci. Technol. **6** (2017) Q3007

O 49.4 Wed 15:45 S053

Atomic scale studies of chromium species on Fe_3O_4(001) — •Moritz Eder[1], Panukorn Sombut[1], Chunlei Wang[1], Matthias Meier[2], Jiri Pavelec[1], Cesare Franchini[2,3], Michael Schmid[1], Ulrike Diebold[1], and Gareth Parkinson[1] — [1]Institute of Applied Physics, TU Wien, Austria — [2]Faculty of Physics and Center for Computational Materials Science, University of Vienna, Austria — [3]Department of Physics and Astronomy, Alma Mater Studiorum, Università di Bologna, Bologna, Italy

Chromium (Cr) ferrite catalysts are industrially employed for the water gas shift reaction in order to provide large amounts of molecular hydrogen.[1] However, the European REACH legislation requires the removal of hexavalent Cr from all catalysts used in industrial processes due to its toxicity. Consequently, alternative metals are sought to fulfill the role of Cr species in the corresponding compounds.[2] To this end, one needs a fundamental understanding of Cr on iron oxides. We present the investigation of Cr species on Fe_3O_4(001) by means of STM and XPS at room temperature. We show the behavior during exposure to water gas shift reactants and upon high-temperature treatment. The results are compared to the behavior of other transition metals[3] and discussed with respect to implications for applied catalysis.

[1] Häussinger, Lohmüller, Watson, Hydrogen, 2. Production. In: Ullmann's Encyclopedia of Industrial Chemistry, 2011

[2] Glassner, Int. Surf. Technol.14, p. 36, (2021)

[3] Bliem et al., Phys. Rev. B 92, p. 075440 (2015)

O 49.5 Wed 16:00 S053

Adsorption and structural behaviour of thymine on Ce2O3 and Ce6WO12 — •Sascha Mehl — Elettra Sincrotrone, Trieste, Italy

Reducible oxides are of particular interest in the field of biological systems and nanotechnology application for instance in biosensors, bio medicine and catalysis. The aim is to design novel nano-size structured thin cerium oxide films that possess bio recognition and bio catalytic properties to detect macro molecules e.g. nucleic acids. Model studies of simplified systems such as nucleobases on well-defined oxide surfaces play a major role for understanding and development of these highly promising bio analytical devices. We utilized synchrotron radiation photo electron spectroscopy (SRPES), resonant photo electron spectroscopy (RPES) and X-ray absorption spectroscopy (XAS) which provide ideal conditions to obtain oxidation states, structural and geometrical information on thin organic ad layers. Furthermore, the focus was on the adsorption behavior, electronic structure and thermal stability in the temperature range of 25 * 250 °C of thymine on reduced cerium oxide model systems: thymine/Ce2O3(111)/Cu(111) and thymine/Ce6WO12(100)/W(110). We distinguished two different adsorbed species of thymine on cerium oxide: one strongly bound chemisorbed type and a second weakly bound physisorbed species in the multi layer regime which desorbs at 75 °C. Further investigations have shown that chemisorbed thymine binds to the surface via N-atoms independent of elementary composition and stoichiometry of the substrate.

O 49.6 Wed 16:15 S053

Interaction of organic acids with magnetite surfaces - the DFT perspective — Kai Sellschopp[1], Wernfried Mayr-Schmölzer[1], Somak Banerjee[1], Johann Fleischhaker[1], Robert Meissner[2], and •Gregor Vonbun-Feldbauer[1] — [1]Institute of Advanced Ceramics, TU Hamburg, Germany — [2]Institute of Polymers and Composites, TU Hamburg, Germany

Magnetite nanoparticles have a high potential for diverse applications like waste-water treatment, catalysis, and hybrid materials. Formic acid and phosphoric acid can be viewed as the smallest representatives of the acids, which are used to functionalize magnetite nanoparticles. Here, we present results from density functional theory (DFT) calculations on the adsorption of such acids on magnetite low index surfaces. For modelling the adsorption, the configuration space is a challenge and here an approach is presented which allows to sample the adsorption configuration space and to select configurations as input for DFT calculations using unsupervised machine learning approaches. The resulting structures allow for in-depth analyses of the systems including structural and electronic effects.

O 49.7 Wed 16:30 S053

Role of surface termination and orientation on the activity of $CoFe_2O_4$(001) and (100) surfaces for water oxidation —

•shohreh rafiezadeh and rossitza pentcheva — Department of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47057 Duisburg

CoFe$_2$O$_4$ plays an important role as an anode material for electrochemical water splitting, necessitating detailed understanding of the mechanism of oxygen evolution reaction (OER). This inverse spinel contains alternating layers of octahedral Co and Fe in the (001) direction and mixed Co and Fe layers along the (100) orientation. Here, we employ density functional theory calculations with an onsite Hubbard U term (DFT+U) to investigate the OER performance of CoFe$_2$O$_4$(100) and iron-rich (001) surfaces and compare to the cobalt-rich (001) surface [1]. While the overpotentials of Fe reaction sites are above 0.44 V, octahedral Co shows the lowest overpotential: 0.38 V at the Co-rich (001) surface and even 0.20 V at the mixed Co-Fe (100) surface when terminated with an additional tetrahedral Fe-layer. This reduction of overpotential correlates with a Co^{2+} oxidation state at the surface and a stabilization of the *OOH intermediate due to hydrogen bonding to neighboring sites. Support by the German Science Foundation (DFG), CRC/TRR 247, project B04 and a computational grant at MagnitUDE are gratefully acknowledged.

[1] H. Hajiyani, R. Pentcheva, ACS Catalysis, 8, 11773-11782 (2018).

O 49.8 Wed 16:45 S053

Complexion Induced Active Phase Evolution in High-Temperature Solid Oxide Cells — •Hanna Türk, Franz-Philipp Schmidt, Thomas Götsch, Robert Schlögl, Axel Knop-Gericke, Thomas Lunkenbein, Christoph Scheurer, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin, Germany

Solid oxide cells (SOCs) are among the most efficient technologies for energy-to-hydrogen conversion from fluctuating renewable electricity sources. While SOCs are in principle well adapted to intermittent operation, cell performance and lifetime in electrolysis mode is severely limited by degradation of the anode. This degradation goes hand in hand with the oxygen evolution reaction (OER) taking place at the triple-phase boundary (TPB) between the anode, the solid electrolyte and the gas phase. Up to now, the atomistic structure of this active catalyst region is essentially unknown though, which prevents a detailed analysis of the actual degradation mechanisms.

Recently, we took the first step in elucidating the TPB structure by revealing a complexion at the underlying solid|solid interface of the sintered anode[1], featuring partial amorphization and varying elemental distributions deviating from the confining bulk phases. Based on this finding, we now expand our force field based Monte-Carlo simulations to the OER active site. Our experimentally validated results show unexpected compositional changes with respect to the thermodynamic equilibrium, that combined with a spatially resolved diffusion study indicate a hitherto unknown deactivation mechanism of the anode.

[1] H. Türk et al., Adv. Mater. Interfaces 8, 2100967 (2021).

O 49.9 Wed 17:00 S053

Synergistic Effects of Co and Fe on the Oxygen Evolution Reaction Activity of LaCo$_x$Fe$_{1-x}$O$_3$ — •Achim Füngerlings and Rossitza Pentcheva — Universität Duisburg-Essen, Fakultät für Physik, Lotharstr. 1, 47057 Duisburg

The efficiency of the perovskite LaCo$_x$Fe$_{1-x}$O$_3$ as a catalyst for the oxygen evolution reaction was investigated by DFT+U calculations. The overpotential required to drive the reaction is significantly reduced upon Co incorporation, with a subsequent nonmonotonic behaviour for larger amounts of Co. This is supported by electrocatalytic measurements of phase-pure LaCo$_x$Fe$_{1-x}$O$_3$ samples [1]. With the deprotonation of adsorbed *OH being the potential determining step in all cases, the reason for the observed trend is twofold: Co turns out to be a more favorable reaction site than Fe, whereas the overpotential of the latter is decreased upon Co substitution. Variations of the magnetic moments of the Co and Fe cations during OER reveal the participation of several, particularly Co, cations up to several layers below the surface. This extends the concept of the active site.

[1] A. Füngerlings, A. Koul, M. Dreyer, A. Rabe, D. M. Morales, W. Schuhmann, M. Behrens, R. Pentcheva, Chem. Eur. J. 27, 17145-17158 (2021)

O 49.10 Wed 17:15 S053

Hydrophobic pockets on an oxide surface: In$_2$O$_3$(111) — Hao Chen[1,2], Matthias Blatnik[1,3], Michael Schmid[1], Bernd Meyer[4], Ulrike Diebold[1], and •Margareta Wagner[1,3] — [1]TU Wien, Vienna, Austria — [2]University of the Chinese Academy of Sciences, Beijing, China — [3]CEITEC BUT, Brno, Czechia — [4]FAU Erlangen-Nürnberg, Erlangen, Germany

Indium oxide, a transparent conductive oxide (TCO), is widely used in semiconductor industry but it also displays promising performance in electro- and photocatalytic reactions. In all applications, surrounding water molecules may influence chemical processes at the atomic scale, and understanding the interaction of water with In$_2$O$_3$ is important.

We focus on In$_2$O$_3$(111), which has an intrinsically large unit cell composed of a hydrophilic and a hydrophobic area. We test the reactivity of these areas by unraveling the interfacial water structures for the whole range of water coverages in UHV, from single dissociated molecules to multilayers, employing TPD, XPS, STM and AFM. Even at high coverages we clearly see hydrophilicity and hydrophobicity within the unit cell, both in experiments and calculations. Local accumulation and depletion of water is confirmed by DFT calculations and ab initio molecular dynamics (MD) simulations for ordered structures consisting of up to 18 water molecules per unit cell. This first water layer shows ordering into nanoscopic 3D water clusters separated by hydrophobic pockets. Going beyond UHV conditions, our MD simulations of a liquid layer show the robustness of the strongly hydrophobic area in the unit cell.

O 50: Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 2

Time: Wednesday 15:00–18:00 Location: S054

O 50.1 Wed 15:00 S054

Automating the Generation of Linearized Augmented Planewave Basis Functions — •Hannah Kleine[1], Sven Lubeck[1], Andris Gulans[2], and Claudia Draxl[1] — [1]Humboldt-Universität zu Berlin — [2]University of Latvia

The linearized augmented planewave (LAPW) and local orbitals (LO) method is known to be a highly precise scheme for solving the Kohn-Sham equations of density-functional theory (DFT) for solids. One drawback, however, is that the LAPW and LO basis functions depend on linearization energies which are material dependent. We propose an approach that allows us to compute the linearization energies from the number of nodes of the respective atomic function. It is implemented in the all-electron full-potential computer package exciting [1]. Following this prescription, we are able to automatize the generation of linearly independent basis functions for any material. This approach not only improves the usability of the method, it also leads to better reproducibility of results, and prepares the code for high-throughput calculations.

[1] A. Gulans et al., J. Phys. Condens. Matter 26, 363202 (2014).

O 50.2 Wed 15:15 S054

Hybrid Density-Functional Theory at the Limit: All-electron Exact Exchange beyond 10,000 Atoms — Florian Merz[1], •Sebastian Kokott[2], Christian Carbogno[2], Yi Yao[3], Markus Rampp[4], Matthias Scheffler[2], and Volker Blum[3] — [1]Lenovo HPC Innovation Center, Stuttgart — [2]The NOMAD Laboratory at the FHI-MPG and HU, Berlin — [3]Duke University, North Carolina, USA — [4]Max Planck Computing and Data Facility, Garching

The computational bottleneck of hybrid density functionals, such as HSE [1], is the evaluation of the exact exchange (EXX) contribution. In this work, we present algorithmic advances in the resolution-of-identity [2] based, real-space implementation of EXX in *FHI-aims* [3,4]. By exploiting MPI-3 intra-node shared memory and enhancing the parallelization scheme, scalability and workload distribution has been drastically improved, which results in memory efficiency and performance increases of up to two orders of magnitude compared to the original implementation, for both total energies and forces/stresses. [3,4] We discuss the details of our implementations and demonstrate the performance as well as scalability for a balanced test set covering inorganic solids, large molecules, and organic crystals with up to 10,000 atoms.

Eventually, we show how these advancements enable insights for the design and optimization of hybrid organic/inorganic perovskites.
[1] Heyd, Scuseria, and Ernzerhof, *J. Chem. Phys.* **118.18**, (2003).
[2] Ihrig *et al.*, *New J. Phys.* **17.9**, (2015).
[3] Levchenko *et al.*, *Comp. Phys. Commun.* **192**, (2015).
[4] Knuth *et al.*, *Comp. Phys. Commun.* **190**, (2015).

O 50.3 Wed 15:30 S054

Studying nuclear quantum effects on water splitting at a charged interface — •Karen Fidanyan and Mariana Rossi — Max Planck Institut for the Structure and Dynamics of Matter

Interfaces of water with charged metallic surfaces are relevant for multiple technological processes.[1] In particular, in electrolyzers, the water splitting reaction happens at charged metallic electrodes. The atomistic mechanisms of this fundamental reaction is determined not only by the surface charge induced by the potential bias, but also by nuclear quantum effects (NQE), which are known to impact such reactions strongly.[2] In this work, we present steps towards a framework for an *ab initio* evaluation of the interplay between these two effects. By making use of a new implementation of the nudged-elastic-band method in the i-PI software,[3] we are able to calculate forces for this reaction from a variety of electronic-structure codes. At first, we approximate the electrode by a slab using density-functional theory with an applied homogeneous electric field and study how NQEs are changed through the water dissociation paths under different field strengths. Taking advantage of the capabilities of i-PI to connect to other codes, our framework can be extended to more accurate models of the potential bias, as for example grand-canonical electronic structure methods.
[1] G. Gonella et al., *Nat Rev Chem* **5**, 466–485 (2021).
[2] Y. Litman, D. Donadio, M. Ceriotti and M. Rossi, *J. Chem. Phys.* **148** 102320 (2018).
[3] V. Kapil et al., *Comp. Phys. Comm.* **236** 214–223 (2019).

O 50.4 Wed 15:45 S054

All-Electron BSE@GW Method for K-Edge Core Electron Excitation Energies — •Yi Yao[1,2], Dorothea Golze[3,4], Patrick Rinke[4], Volker Blum[2,5], and Yosuke Kanai[1] — [1]Department of Chemistry, University of North Carolina at Chapel Hill, Chapel Hill, North Carolina 27599, United States — [2]Thomas Lord Department of Mechanical Engineering and Materials Science, Duke University, Durham, North Carolina 27708, United States — [3]Faculty of Chemistry and Food Chemistry, Technische Universität Dresden, 01062 Dresden, Germany — [4]Department of Applied Physics, Aalto University, P.O. Box 11100, FI-00076 Aalto, Finland — [5]Department of Chemistry, Duke University, Durham, North Carolina 27708, United States

We present an accurate computational approach to calculate K-edge core electron excitation energies, achieved by combining all-electron GW and Bethe-Salpeter equation (BSE) methods. We assess the BSE@GW approach for calculating K-edge X-ray absorption spectra using a set of small organic molecules and also a medium-sized sulfur-containing molecule, which was used in a past benchmark of an equation-of-motion coupled-cluster (EOM-CC) method by Peng and coworkers [Peng et al., J. Chem. Theory Comput., 11, 4146 (2015)]. We present the influence of different numerical approximations. We assess the basis set dependence and convergence. We identify the importance of core-correlation basis functions as well as the augmenting basis functions. Compared to the experimental values, the predicted mean absolute error by BSE@GW is as low as 0.6-0.7 eV.

O 50.5 Wed 16:00 S054

Hybrid excitations at the interface between MoS_2 monolayer and organic molecules from first-principle calculation — •Ignacio Gonzalez Oliva[1], Fabio Caruso[1,2], Pasquale Pavone[1], and Claudia Draxl[1] — [1]Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, 12489 Berlin, Germany — [2]Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, 24118 Kiel, Germany

Hybrid materials composed of organic and two dimensional (2D) inorganic semiconductors are receiving increasing attention due to the interesting physical processes happening at the interface. We present a first-principles investigation of the electronic and optical properties of hybrid organic-inorganic interfaces consisting of a MoS_2 monolayer and the π-conjugate molecules pyrene and pyridine. Employing the G_0W_0 approximation to obtain the quasi-particle band structures, and solving the Bethe-Salpeter equation, we compute the absorption spectra. The latter reveal intralayer excitons on the MoS_2 side, and also hybrid as well as charge-transfer excitons at the interface. Our findings indicate that hybrid systems consisting of semiconducting transition-metal dichalcogenides and organic π-conjugate molecules can host a rich variety of optical excitations and thus provide a promising venue to explore exciton physics in low dimensionality.

O 50.6 Wed 16:15 S054

Mott Metal-Insulator Transition from Steady-State Density Functional Theory — •David Jacob[1,2], Gianluca Stefanucci[3], and Stefan Kurth[1,2,4] — [1]Universidad del País Vasco UPV/EHU, San Sebastian, Spain — [2]Ikerbasque Foundation, Bilbao, Spain — [3]Università di Roma Tor Vergata, Rome, Italy — [4]Donostia International Physics Center, San Sebastian, Spain

We present a computationally efficient method to obtain *many-body* spectral functions of bulk systems in a *density functional theory* framework [1]. To this end we generalize a recently developed method for computing many-body spectral functions of nanoscale systems [2], based on steady-state density functional theory (i-DFT) and using an idealized scanning tunneling microscope (STM) setup, to the case of bulk systems. In this setup the spectral function can be obtained from the finite-bias differential conductance of the current through the STM tip. The fictitious noninteracting system of i-DFT features an exchange-correlation (XC) contribution to the bias which guarantees the same current as in the true interacting system. Exact properties of the XC bias are established using Fermi-liquid theory and subsequently implemented to construct approximations for the Hubbard model. We show for two different lattice structures that our method captures the Mott metal-insulator transition.

References: [1] D. Jacob, S. Kurth, G. Stefanucci, Phys. Rev. Lett. **125**, 216401 (2020); [2] D. Jacob and S. Kurth, Nano Lett. **18**, 2086 (2018).

O 50.7 Wed 16:30 S054

Network of 1d edge channels and localized states emerging in moiré system — •Jeyong Park, Jinhong Park, and Achim Rosch — Institute of theoretical physics, University of Cologne, Germany

For the general moiré system, we theoretically study the effective Hamiltonian for single layer considering the smooth periodic moiré potential induced by other layer, which appears because of distance difference between each pair of atoms located on different layer. For moiré potential larger than tunable moiré energy scale, we find the coexistence of 1D channels and localized state in real space. The 1D channels emerge along the three 1D lines where the mass gap of single layer coming from moiré potential becomes zero, which is showing that these channels are the edge states. Localized states emerge at the crossing point of three 1d lines. We construct the toy model which describes the coexistence of these states by symmetry analysis and consider the RKKY interaction between localized states which is mediated by 1D channels by including whole spin and valley degrees of freedom. Expanding the effective action for the toy model and controlling chemical potential to suppress oscillation coming from fermi momentum gives three spin interaction $S \cdot (S \times S)$, which can give chiral spin liquid phase. We show how the analytical result can be seen in our model numerically and propose the graphene on top of AB stacking bilayer with twist as the candidate material for realization.

O 50.8 Wed 16:45 S054

Incorporating First-Principles Electronic Friction in Instanton Rate Theory — •Yair Litman[1], Eszter S. Pos[1], Connor L. Box[2], Rocco Martinazzo[3], Reinhard J. Maurer[2], and Mariana Rossi[1] — [1]MPI for the Structure and Dynamics of Matter, Hamburg, Germany. — [2]Department of Chemistry, University of Warwick, Coventry, United Kingdom — [3]Department of Chemistry, Università degli Studi di Milano, Milano, Italy

Reactions involving impurities in bulk metals are ubiquitous in a wide range of technological applications. The theoretical modelling of such reactions present a challenge for theory because nuclear quantum effects (NQEs) can play a prominent role and the coupling of the atomic motion with the electrons in the metal gives rise to important non-adiabatic effects (NAEs). In this work, we present a theoretical framework for the calculation of reaction rates capable of capturing both NQEs and NAEs for high-dimension realistic systems [1,2]. This is achieved by combining the ring polymer instanton (RPI) formalism with *ab initio* electronic friction from Ref. [3]. We derive equations that incorporate the spatial and frequency dependence of electronic friction, and name the new method RPI with explicit friction (RPI-EF). We validate RPI-EF against numerically exact results and find, quantitatively, how the friction modifies reaction rates and tunnelling pathways. Finally, we present *ab initio* results for H-hopping in selected bulk metals. [1] Y. Litman, E. S. Pós, C. L. Box, R. Martinazzo, R. J. Maurer, and M. Rossi, *JCP* **156**, 194106 (2022) [2] *id. JCP* **156**, 194107 (2022) [3] M. Head-Gordon, J. C. Tully, *JCP* **96**, 3939 (1992).

O 50.9 Wed 17:00 S054

Real-Time Time-Dependent Density Functional Theory within the FHI-aims code — Joscha Hekele[1], Yi Yao[2], Volker Blum[2], Yosuke Kanai[3], and •Peter Kratzer[1] — [1]Faculty of Physics, University Duisburg-Essen, 47057 Duisburg, Germany — [2]Thomas Lord Department of Mechanical Engineering and Materials Science, Duke University, Durham, North Carolina 27708, USA — [3]Department of Chemistry, University of North Carolina, Chapel Hill, North Carolina 27599, USA

We present a high-precision all-electron RT-TDDFT implementation using numerical atom-centered orbital (NAO) basis functions into FHI-aims. First, RT-TDDFT results are validated against linear-response TDDFT results for the molecules of Thiel's test set and the importance of basis augmentation for adequate convergence is confirmed. Adopting a velocity-gauge formalism, dielectric properties of crystalline materials are calculated. Taking advantage of the all-electron full-potential implementation, we present applications to soft X-ray spectra of H_2O using hybrid functionals. Moreover, numerical performance tests are presented showing almost linear scaling on parallel computers.

O 50.10 Wed 17:15 S054

***Ab initio* study of many-body interacting nonlinear optical photoconductivity tensors** — •Peio Garcia-Goiricelaya[1], Jyoti Krishna[1], and Julen Ibañez-Azpiroz[1,2] — [1]Centro de Física de Materiales, University of the Basque Country UPV/EHU, Spain — [2]IKERBASQUE Basque Foundation for Science, Spain

We present a general scheme for calculating the many-body interacting second-order optical photoconductivity tensor from first-principles. Our practical implementation starts from the length-gauge formulation of the single-particle non-interacting second-order response tensor [1] that is efficiently calculated using Wannier interpolation [2]. In a second step, we make use of the TD-CDFT in order to extract the many-body interacting response tensor, taking into account collective excitations, i.e. excitonic effects for semiconductors and plasmonic effects for metals. We employ this scheme to assess the impact of many-body excitations on second harmonic generation and benchmark our results with experiments on the semiconducting GaAs [3] and the semimetal TaAs [4], as well as with early calculations [5].

Funding provided by the European Union's Horizon 2020 research and innovation programme under the European Research Council (ERC) grant agreement No 946629.

[1] J. E. Sipe and A. I. Shkrebtii, Phys. Rev. B **61**, 5337 (2000).
[2] G. Pizzi *et al.*, J. Phys. Cond.Matt. **32**, 165902 (2020)
[3] S. Bergfeld and W. Daum, Phys. Rev. Lett. **90**, 036801 (2003).
[4] Shreyas Patankar *et al.*, Phys. Rev. B **98**, 165113 (2018).
[5] E. Luppi *et al.*, Phys. Rev. B **82**, 235201 (2010).

O 50.11 Wed 17:30 S054

Electron-phonon drag in MgB_2 — •Nakib Protik and Claudia Draxl — Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany

MgB_2 is a well known phonon-mediated superconductor with a transition temperature around 40 K. The strong electron-phonon interactions present in the material also give rise to strong drag phenomena, as has been demonstrated experimentally in Refs. [1, 2]. While the measured thermopower in the normal phase shows hole-like transport, there is, however, no consensus regarding the sign and temperature dependence of the drag contribution. In this work, we carry out anisotropic and band-resolved *ab initio* transport calculations to investigate this puzzle. Our findings will shed light on the roles of the microscopic interactions on the strong drag behavior in metals.

[1] Schneider, Matthias, Dieter Lipp, Alexander Gladun, Peter Zahn, Axel Handstein, Günter Fuchs, Stefan-Ludwig Drechsler, Manuel Richter, Karl-Hartmut Müller, and Helge Rosner. "Heat and charge transport properties of MgB_2." Physica C: Superconductivity 363, no. 1 (2001): 6-12.

[2] Putti, M., E. Galleani d'Agliano, D. Marre, F. Napoli, M. Tassisto, P. Manfrinetti, A. Palenzona, C. Rizzuto, and S. Massidda. "Electron transport properties of MgB_2 in the normal state." The European Physical Journal B-Condensed Matter and Complex Systems 25, no. 4 (2002): 439-443.

O 50.12 Wed 17:45 S054

***Ab initio* simulations of vibrational sum frequency generation without molecular decomposition** — •Paolo Lazzaroni, Alan Lewis, and Mariana Rossi — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

Hydrogen evolution through heterogeneous catalysis is an increasingly popular solution for storage of energy generated from renewable sources, and as such is becoming a central part of green energy grids. Understanding the structure of water at solid interfaces is vital to find more efficient and cost-effective catalysts of this reaction, and sum-frequency generation spectroscopy provides a surface-specific method of investigating these structures [1]. In this work we present a new framework for *ab initio* calculations of the sum-frequency response which avoids the need for an arbitrary molecular decomposition of the polarizability and dipole moment. Within this approach, based on Density Functional Theory, the observables are evaluated using real space integrals directly on the whole system [2]. This allows us, for example, to unravel the effect of this decomposition on the sum-frequency response of the much-studied water/vacuum interface. This scheme also enables us, in the context of water-solid interfaces, to account explicitly for the contribution of the solid to the polarizability and dipole moment of the interface, which are commonly neglected.
[1] A. Morita, J. Hynes, J. Phys. Chem. B 106, 673 (2002)
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O 51: Poster Wednesday: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy

Time: Wednesday 18:00–20:00 Location: P4

O 51.1 Wed 18:00 P4

Nickelocene molecule as an STM magnetic sensor — •Andres Pinar Sole, Oleksandr Stetsovych, Pavel Jelinek, Ales Cahlik, Christian Wakerlin, and Jindric Kolorenc — Czech Institute of Physics, Prague, Czech Republic, Email: pinar@fzu.cz

Functionalization of the scanning probe of a scanning tunnelling microscopy (STM) with metallocene molecule allows performing spin-sensitive measurements on magnetic systems. Here, we used a nickelocene molecule (NiCp2), consisting of a Ni atom sandwiched between two cyclopentadienyl rings. As an S=1 system, it presents magnetic-induced spectral features due to the inelastic electron spin-flip.

First, we examined 1D metallorganic coordination polymers (2,5-diamino-1,4-benzoquinonediimines) on Au(111) with Co or Cr atoms as metal sites respectively. Nickelocene IETS conductance spectrum deformation was only sensitive to the out-of-plane magnetic anisotropy of the Cr.

Secondly, the Nc functionalized probe was also used to measure the magnetism emerging from the unpaired electron on the edge of a wave-like graphene nanoribbon (GNR) on Au(111). Here, we observed spectral convolution between a Kondo feature from the edge-state and the nickelocene spectrum.

To understand the IETS from the magnetic sensor, a many-body Hubbard model was proposed, and the system was simplified to a two-site model consisting of the partially filled 3d shell of the nickelocene and a 3d shell in the probed magnetic center.

O 51.2 Wed 18:00 P4

Yu-Shiba-Rusinov band dispersion of infinite Mn chains on top of a semi-infinite Nb(110) surface — •Rik Broekhoven, Artem Pulkin, Antonio Manesco, Sander Otte, Anton Akhmerov, and Michael Wimmer — Delft University of Technology, Delft, The Netherlands

Chains of magnetic atoms on s-wave superconductors have been proposed to have a topological non-trivial phase, when the induced in-gap

Yu-Shiba-Rusinov (YSR) bands are p-wave gapped by for example spin-orbit coupling. In order to determine the topological phase diagram, experiments were previously limited to probing local quantities of the chain. Recent improvements in STM methodology have made it possible however to now also probe the YSR band dispersion relation. It was found to be highly dependent on the chain spacing and orientation with respect to the superconductor lattice.

Motivated by this discovery, here we present a method to evaluate the in-gap dispersion relation of chains with different orientations and spacing. In contrast to previous works, we work with infinite chains on a semi-infinite surface to make sure the system is larger than the superconductor coherence length. We focus on Mn atoms on top of superconducting Nb(110). First, we derive an effective tight-binding model from ab initio calculation, and subsequently we use the multidimensional Green's function formalism to extend to a semi-infinite system and solve for the corresponding in-gap YSR bands.

O 51.3 Wed 18:00 P4

Unravelling the Magnetic Ground State of All-Organic Diradicals on Au Substrates — •Alessio Vegliante[1], Saleta Fernandez[2], Ricardo Ortiz[3], Niklas Friedrich[1], Andrea Aguirre[1], Francisco Romero[1], Diego Peña[2], and Thomas Frederiksen[3] — [1]CIC NanoGUNE, San Sebastian, Spain — [2]CiQUS-USC, Santiago de Compostela, Spain — [3]Donostia International Physics Center, San Sebastian, Spain

Open-shell organic molecules have received considerable interest as potential candidates for molecular spintronics. Magnetism can emerge in all-organic molecules due to the presence of one or more unpaired π-electrons. Organic diradicals, hosting two spin centers, are particularly interesting as model systems for investigating and manipulating magnetic interactions at the atomic scale.

Here we study the magnetic state of a Chichibabin*s hydrocarbon diradical deposited on a gold substrate, by using scanning tunneling microscopy (STM) and spectroscopy (STS). We investigate the interaction between the two spin centers of the molecule, focusing on the influence of the molecular geometry. With the support of theoretical simulations, we show that the adsorption on the metal surface changes the ground state of the molecule from a triplet, expected in the gas phase, to a singlet state. Furthermore, we demonstrate that it is possible to modify the magnetic state of the diradical through conformational changes induced by the STM tip. These findings represent an important step towards the control of magnetic interactions within purely organic molecules.

O 51.4 Wed 18:00 P4

Quantifying the interplay between fine structure and geometry of an individual molecule on a surface — •Manuel Steinbrecher[1], Werner M.J. v. Weerdenburg[1], Etienne F. Walraven[1], Niels P.E. v. Mullekom[1], Jan W. Gerritsen[1], Fabian D. Natterer[2], Danis I. Badrtdinov[1], Alexander N. Rudenko[4,1], Vladimir V. Mazurenko[3], Mikhail I. Katsnelson[1], Ad v.d. Avoird[1], Gerrit C. Groenenboom[1], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands — [2]Department of Physics, University of Zurich, Switzerland — [3]Ekaterinburg — [4]School of Physics and Technology, Wuhan University, China

With spin-resolved scanning tunneling microscopy (SP-STM) and electron spin resonance (ESR) we have probed single TiH molecules deposited on a thin insulating MgO layer in a vector magnetic field at mK temperatures down to MHz frequencies. We find that the molecule retains a non-trivial orbital angular momentum resulting in a strongly renormalized and anisotropic g-tensor. As we prove, the latter does not stem from Kondo or Jahn-Teller effects. From quantum chemistry embedded cluster calculations we find an analytical expression for the g-tensor, which solely depends on the splitting of the ground states and the spin-orbit coupling. In a dynamic expansion of the model, the position of the H atom and rotational dynamics of the molecule were investigated. [1] Steinbrecher *et al.*, PRB 103, 155405 (2021) [2] v. Weerdenburg *et al.*, RSI 92, 033906 (2021)

O 51.5 Wed 18:00 P4

Magnetic molecule as a parity sensor in entangled spin and YSR excitation on a superconductor — •Jon Ortuzar[1], Stefano Trivini[1], Katerina Vaxevani[1], Jingchen Li[5], Ane Garro[1], Miguel Angel Cazadilla[3], Sebastian Bergeret[2,3], and José Ignacio Pascual[1,4] — [1]CIC nanoGUNE-BRTA, San Sebastián, Spain — [2]Centro de Física de Materiales (CFM-MPC), San Sebastián, Spain — [3]Donostia International Physics Center (DIPC), San Sebastián, Spain — [4]Ikerbasque, Bilbao, Spain — [5]School of Physics, Sun Yat-sen University, Guangzhou, China

A magnetic molecule coupled to a superconductor induces Yu-Shiba-Rusinov (YSR) bound states, detected by tunneling spectroscopy as long-lived quasiparticle excitations inside the superconducting gap [1]. The degeneracy of the system can be lifted by intrinsic magnetic anisotropy so that entangled spin and YSR excitations are possible [2]. We tune the exchange coupling between a FeTPP-Cl molecule and the proximitized Au(100)/V substrate to go through a Quantum Phase Transition from an unscreened half-integer spin (even parity) system to a screened integer spin (odd parity) system [3]. We use a single site superconductor model [4] to prove the capability to detect the parity of the system, getting fundamental insight into the interplay of YSR and spin excitation.

[1] B. W. Heinrich, et. al., Prog. Surf. Sci.93, 1 (2018) [2] N. Hatter, et. al., Nat. Commun.6, 8988 (2015) [3] S. Kezilebieke, et. al., Nano Lett.19, 4614 (2019) [4] F. von Oppen and K. J. Franke, Phys. Rev. B103, 205424 (2021).

O 51.6 Wed 18:00 P4

Electronic Properties of Dysprosium-based Fe-Porphyrins Metal-Organic Coordination networks on Au(111) and Ag(100) Substrates — •Serim Jeon[1,3], Lukas Spree[1,2], Corina Urdaniz[1,2], Caroline Hommel[1,2], Andreas Heinrich[1,2], Christoph Wolf[1,2], and Luciano Colazzo[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Republic of Korea — [2]Ewha Womans University, Seoul, Republic of Korea — [3]Department of Physics, Ewha Womans University

Dy has become the cornerstone for many investigations on lanthanide-directed molecular-magnetism, thanks to its strong magnetic anisotropy. Iron Tetrapyrrole complexes, on the other hand, have shown promise as materials for quantum information processing. Creating a platform that incorporates both might have a way for the realization of multiqubit architectures. The presented study describes electronic properties within a metal-organic network MOF consisting of Iron-Tetrakis-(4-Cyanophenyl) Porphyrin Fe-TCPP and Dysprosium. Fe-TCPP was deposited on Au(111), Ag(100), and MgO/Ag(100). The deposition of Dy induces coordination between the cyano groups and the lanthanide atoms and gives rise to ordering and the formation of large islands of 2D MOF. The lanthanide-based organic network was examined by scanning tunneling microscopy STM on the substrates. Significant shifts in the energy of LUMO of the MOFs were observed via scanning tunneling spectroscopy STS after deposition of Dy. The results were combined with ab initio calculation to further elucidate the electronic structure of the deposited materials.

O 51.7 Wed 18:00 P4

Long-lived spin states of Fe atomic chains on Cu_2N via Hamiltonian engineering — Robbie J. G. Elbertse[1], •Taehong Ahn[2,3], Jiyoon Hwang[2,3], Jeongmin Oh[2,3], Jorn C. Rietveld[1], Sander Otte[1], Andreas J. Heinrich[2,3], and Yujeong Bae[2,3] — [1]Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology, Lorentzweg 1, Delft 2628 — [2]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Korea — [3]Department of Physics, Ewha Womans University, Seoul, Korea

A spin-polarized scanning tunneling microscope (STM) operating at various magnetic fields allows us to characterize the spin relaxation time of atomic structures on a surface, enabling Hamiltonian engineering. Here, we introduce 1D Fe atomic chains built on Cu_2N/Cu(100) using a home-built STM and the evolution of their spin lifetime depending on the length of chains and the direction and magnitude of magnetic fields. To measure the spin lifetime ranging from 10^{-6} to 10 seconds, we used two different detection schemes; pump-probe [Science 329, 1628-1630 (2010)] and two state switching [Science 335, 196-199 (2012)]. We observed the spin lifetime of Fe chains changes non-monotonically as passing through a diabolic point with varying magnetic fields changes the degree of superposition of the two Neél states in the lowest energy eigenstates. To control the spin lifetime of Fe chains, we use magnetic fields as a control knob. Our work shows a capability of the spin lifetime in a large time scale and the precise control of spin dynamics in engineered atomic structures.

O 51.8 Wed 18:00 P4

Local characterization of Yu-Shiba-Rusinov excitations in magnetic field — •Niels P.E. van Mullekom[1], Benjamin Verlhac[1], Werner M.J. van Weerdenburg[1], Hermann

Osterhage[1], Manuel Steinbrecher[1], Katharina J. Franke[2], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University Nijmegen, the Netherlands — [2]Fachbereich Physik, Freie Universität Berlin, Germany.

An isolated spin, typically derived from an atomic or molecular adsorbate, can interact with a BCS superconductor leading to so-called Yu-Shiba-Rusinov (YSR) excitations. These excitations can be probed with scanning tunneling microscopy/spectroscopy (STM/STS), which are often characterized by resonances that reside in the superconducting gap and are linked to the excitation energies. These spin-based excitations to date have been poorly studied in magnetic fields, due to influence of magnetic fields on typical BCS superconductors. Here, utilizing STM/STS at milliKelvin temperatures, we report on a study of the YSR excitations of individual adsorbates in variable magnetic field.

O 51.9 Wed 18:00 P4

Extending the spin excitation lifetime of a magnetic molecule on a proximitized superconductor — •Katerina Vaxevani[1], Stefano Trivini[1], Jingcheng Li[1], Jon Ortuzar[1], Dongfei Wang[1], Danilo Longo[1], and Jose Ignacio Pascual[1,2] — [1]CIC nanoGUNE-BRTA, 20018 Donostia-San Sebastian, Spain — [2]Ikerbasque, Basque Foundation for Science, 48013 Bilbao, Spain

Magnetic molecules adsorbed on surfaces have been used as a platform to individually address and manipulate spins. Long spin-relaxation times are required in order to be able to use atomic spins in quantum information processing and data storage. Normally, coupling of the spin with the conduction electrons of metallic substrates can quench the excited state lifetime and lead to short relaxation times, but the presence of superconducting paring effects in the metal substrate can protect the excited spin from relaxation[1]. Here, we use a substrate of a few monolayers of gold epitaxially grown ontop of an oxygen reconstructed 1x5-V(100) surface to decouple the molecular spin of an iron-porphyrin-chloride from itinerant electrons. The gold film exhibits a proximitized superconducting gap with in-gap de Gennes-Saint James resonances, which protects molecular spin excited states and results into a lifetime of τ=80ns. The spin lifetime decreases with increasing the film thickness due to the gradual gap-closing by the in-gap states. Our results elucidate the use of proximitized gold electrodes for addressing quantum spins on surfaces, envisioning new routes for tuning the value of their spin lifetime.

[1] B. W. Heinrich et al., Nature Physics 9, 765 (2013).

O 51.10 Wed 18:00 P4

The Emergence of Magnetism in [5]-Aza-Triangulene — •Lorenz Meyer[1], Francisco Romero Lara[1], Niklas Friedrich[1], Alessio Vegliante[1], Manuel Vilas Varela[2], Unai Uriarte Amiano[1], Natalia Koval[1], Emilio Artacho Cortés[1], Diego Peña Gil[2], and Jose Ignacio Pascual[1] — [1]1 CIC nanoGUNE, San Sebastian (Spain) — [2]Centro Singular de Investigación en Química Biológica y Materiales Moleculares (CIQUS), Santiago de Compostela (Spain)

Zigzag-edged Graphene triagulenes are known to host a magnetic ground state due to sublattice imbalance. The magnetic properties of these structures can be tuned by the size of the triangulene or via heteroatom substitution. This makes them intriguing for future spintronic applications paving the way towards organic logical devices. So far, the influence of heteroatoms in larger triangulenes seems to be unknown. In our work, we report the bottom-up synthesis of [5] aza-Triangulene by means of surface-assisted cyclodehydrogenation on a Au(111) surface. Electronic transport measurements carried out with a scanning tunneling microscope reveal the high-spin ground state of this molecule via low energy phenomena. Additionally, we shine light on not only the influence of heteroatoms but also of additional Hydrogen atoms and defects which are able to tune the magnetic ground state. Density functional theory and Mean-Field-Hubbard calculations confirm the spin texture and the electronic structure of the observed molecules.

O 52: Poster Wednesday: Adsorption and Catalysis 2

Time: Wednesday 18:00–20:00 Location: P4

O 52.1 Wed 18:00 P4

Investigation of the influence of the spin in adsorption structures of oxygen on a cerium chloride catalyst surface in the Deacon process using computational methods — •Kathrin Nieswiec and Franziska Hess — Technische Universität Berlin, Institut für Chemie, Straße des 17. Juni 124, 10623 Berlin, Germany

CeO_2-based catalysts in the HCl oxidation undergo reversible deactivation by forming $CeCl_3$. This works examines the interaction of the $CeCl_3$-(110)-surface with O_2 during the reoxidation of $CeCl_3$. $CeCl_3$ is an insulator, where Ce is 9-fold coordinated that exposes different adsorption sites at the surface. Different adsorption structures of atomic and molecular oxygen on the $CeCl_3$-(110)-surface are examined in terms of energy, electronic state and magnetization. Cerium-based catalyst systems are investigated by DFT calculations in the PAW approach with the PBE functional including Hubbard-U correction. Ab-initio thermodynamics are employed to analyze temperature and pressure-dependency of oxygen adsorption on the surface. Upon adsorption of oxygen on the surface, Ce^{3+} can be locally oxidized to Ce^{4+}, resulting in different spin and charge density distributions. Consequently, the adsorption energies of oxygen can variate by over 1 eV, depending on the initial spin state. The ab-initio thermodynamics analysis based on the obtained oxygen adsorption energies suggests that the $CeCl_3$ surface is expected to be covered by oxygen due to the strong Ce-O bond. These results suggest that spin must be explicitly included when examining adsorption on $CeCl_3$ surfaces in order to obtain correct adsorption energies for catalysis applications.

O 52.2 Wed 18:00 P4

Influence of the Substrate Orientation Dependent Reactivity on the On-surface Ullmann Coupling Reaction of 2,2-Dibromobiphenyl Studied by XPS — •Paul Schweer and Karina Morgenstern — Chair of Physical Chemistry I, Ruhr-University of Bochum, Germany

The Ullmann coupling reaction is considered the leading approach for accessing low dimensional covalent organic structures in the context of on-surface synthesis. Despite extensive studies on the influence of the halogen substitution pattern of the halogen-aryl precursor on the reaction mechanism and product formation, the impact of ortho-substitution is barely investigated. Hence, this work aims to reveal the temperature-induced reaction steps for 2,2-dibromobiphenyl on low-index Cu surfaces. For this purpose, the precursor is deposited in ultra-high vacuum at 86 K and stepwise annealed to 600 K. By using X-ray photoelectron spectroscopy, chemical changes of the organic species and the bromine atoms are identified and assigned to the typical Ullmann reaction steps: dissociation of the halogen-carbon bond, surface bonding, and C-C coupling. Here, we further discuss different precursor reactivities on Cu(111) and Cu(110) and an additional binding energy shift on Cu(110), indicating a structural change of the surface bound biphenyl before C-C coupling at a higher temperature.

O 52.3 Wed 18:00 P4

Surface Chemistry of the MOST Energy Storage System 2-Carbethoxy-3-Phenyl-Norbornadiene/Quadricyclane — •Felix Hemauer, Cornelius Weiss, Johann Steinhauer, Valentin Schwaab, Hans-Peter Steinrück, and Christian Papp — Friedrich-Alexander-Universität, Erlangen, Germany

The intermittent character of renewable energy sources gives the necessity for novel energy storage technologies. So-called molecular solar thermal (MOST) energy systems directly combine the light-harvesting process with storing the gained energy as molecular strain. In a photoconversion reaction, the energy-lean norbornadiene (NBD) is converted to its energy-rich valence isomer quadricyclane (QC). By derivatization of the molecular framework, the light-harvesting properties of the molecules are optimized.

The pair 2-carbethoxy-3-phenyl-NBD/QC was investigated as model system for heterogeneously catalyzed energy release. X-ray photoelectron spectroscopy was employed to study the adsorption and thermal evolution on a Ni(111) and Pt(111) surface, respectively. An unambiguous identification of the QC and NBD derivatives at low temperature was possible on both surfaces. In case of nickel, no cycloreversion of the QC derivative to NBD was found, but individual decomposition routes setting in at about 170 K. For platinum, the back conversion under energy release was found to start at 150 K and being completed at 230 K. Above 300 K, a fragmentation into carbonaceous species occurred. The work was supported by the DFG (Project No. 392607742) and the HZB for allocation of synchrotron radiation beamtime.

O 52.4 Wed 18:00 P4

Temperature-induced surface reactions of 1-cyclohexylethanol and acetophenone on Pt(111) — •Valentin Schwaab, Felix Hemauer, Eva Marie Freiberger, Natalie Waleska, Hans-Peter Steinrück, and Christian Papp — Friedrich-Alexander-Universität, Erlangen, Germany

1-Cyclohexylethanol (1-CHE) is an interesting candidate when it comes to molecular energy storage systems. Such secondary alcohols can function as electrofuel, whereby their oxidation over a platinum electrode yields the respective ketone together with two electrons and two protons. Additionally, 1-CHE and acetophenone (APH) represent an attractive liquid organic hydrogen carrier (LOHC) pair, as the complete dehydrogenation of the former leads to the release of four equivalents of H_2.

To gain fundamental insights into the surface reaction of both molecules, high-resolution temperature-programmed X-ray photoelectron spectroscopy experiments (HR-TPXPS) were carried out on a Pt(111) model catalyst. Based on the obtained C 1s and O 1s data, the dehydrogenation reaction of the alcohol and the formation of potential catalyst poisoning decomposition products are discussed.

We acknowledge financial support by the Bavarian Ministry of Economic Affairs, Regional Development and Energy, and by the DFG (Project No. 419654270). We thank HZB for the allocation of synchrotron radiation beamtime.

O 52.5 Wed 18:00 P4

Force-induced single-molecular switch of graphene-nanoribbon-fused helicene by atomic force microscopy — •Akitoshi Shiotari[1,2], Ayumu Ishii[1], and Yoshiaki Sugimoto[1] — [1]The University of Tokyo, Kashiwa, Japan — [2]Fritz-Haber Institute of the Max-Planck Society, Berlin, Germany

On-surface synthesis is an effective bottom-up method to fabricate nanocarbon materials, such as graphene nanoribbons (GNRs), in an atomically precise manner. In this study, using the on-surface method with multiple precursors, we successfully synthesized new helicene-derivative molecule fused by seven-atom-wide armchair GNRs on an Au(111) surface. Atomic resolution imaging with non-contact atomic force microscopy (AFM) identified the molecular structure and the helicity (i.e., chirality of the twisted terminal of the GNR). Although the helicities of individual GNRs on the surface remained stable during observation, the approach of the AFM tip to the helicene-type terminal caused repulsive interactions, leading to the inversion of the twisted structure in a selective and reversible manner. This finding would contribute to the advanced design of switchable molecules and control of chiral-molecule-based devices and machines.

O 52.6 Wed 18:00 P4

On-Surface Synthesis of Kekulene and Isokekulene — •Tim Naumann, Qitang Fan, Lukas Heuplick, Lukas Ruppenthal, Simon Werner, Tobias Vollgraff, Jörg Sundermeier, and J. Michael Gottfried — Fachbereich Chemie, Philipps-Universität Marburg, Marburg (Germany)

The concept of aromaticity explains the exceptional stability of monocyclic, planar, conjugated molecules. When extending this concept on cycloarenes, however, the question arises whether the π-electron system is best described by the Clar model or rather by annulenoid aromaticity, i.e. delocalization in the inner and outer annulene ring. This question has been discussed especially in the context of kekulene, an alternant benzenoid cycloarene. On-Surface techniques offer the possibility to synthesize kekulene and to investivate its resonance stabilisation. To create complete monolayers of kekulene for further spectroscopic studies, we developed a high-yield on-surface synthesis of kekulene on Cu(111) from vapor-deposited 1,4,7(2,7)-triphenanthrenacyclononaphane-2,5,8-triene, which undergoes cyclodehydrogenation upon annealing, resulting in extended (up to 100 nm) well-ordered domains of kekulene. While the reaction is highly selective towards kekulene on Cu(111), reaction on Cu(110) leads to an isomer of kekulene with a lower symmetry, named isokekulene. The precursor and the products were analyzed with scanning tunneling microscopy (STM), X-ray photoelectron spectroscopy (XPS) and other methods. Further annealing leads to peripheral C-H bond activation and linking towards chains and islands.

O 52.7 Wed 18:00 P4

2D arrays of reaction centers for CO oxidation over Pt/SiO2/Si interface — •Artur Böttcher[1], David Rettinger[1], Jakob Hauns[1], Manfred Kappes[1], Daniela Exner[2], Rahul Parmer[3], Matteo Amati[3], and Luca Gregoratti[3] — [1]Institute of Physical Chemistry, KIT, 76131 Karlsruhe, Germany — [2]Institute for Applied Materials, KIT, 76344 Karlsruhe, Germany — [3]Elettra - Sincrotrone Trieste ScPA, Area Science Park, 34149 Basovizza-Trieste, Italy

We created arrays of tailored reaction centers on the SiO2/Si-wafer by annealing thin Pt layers deposited on He+-beam patterned Graphene/SiO2/Si substrates.[1] The resulting spots have been identified as amorphous areas consisting of silicon carbides and silicon oxy-carbides. These defected surface regions are expected to act as pinning sites for migrating Pt atoms. By thermally activating the surface diffusion of Pt adatoms (1h, 600°C, UHV), the migrating atoms become immobilized thus forming chemical bonds with compounds created in the defected surface regions, resulting in 2D arrays of tailored Pt-based islands. The chemical state of individual islands has been studied by monitoring core level states (ESCA microscopy). The islands consist of Pt, PtSix and PtOx compounds and appear to be chemically resistant under exposure to molecular oxygen and carbon monoxide at T=550°C, p= 0.4 mbar, 0.5 h. The CO2-light-off curves enable to estimate the contribution of the artificial reaction centers to the total CO->CO2 conversion yield.

[1] A. Böttcher et al. 2020 Nanotechnology 31 505302

O 52.8 Wed 18:00 P4

$CO \rightarrow CO_2$ conversion over Pt/SiO_2/Si model catalyst — •Artur Böttcher[1], Pascal Weisenburger[1], David Rettinger[1], Jakob Hauns[1], Rahul Parmer[2], Matteo Amati[2], Luca Gregoratti[2], and Manfred Kappes[1] — [1]Institute of Physical Chemistry, KIT, 76131 Karlsruhe, Germany — [2]Elettra - Sincrotrone Trieste ScPA, Area Science Park, 34149 Basovizza-Trieste, Italy

We studied the thermal stability of the Pt/SiO_2/Si interface as the model catalyst for CO oxidation reaction. The 2D catalysts were prepared under UHV conditions by growing very thin Pt films on the SiO_2/Si wafer and subsequently performing a long-term annealing of the interface at elevated temperatures, T*(700-900K). Such a treatment results in the formation of the submicron-sized islands. The size/shape distribution of the islands scales primarily with the thickness of the Pt film. The chemical state of the islands as probed by ESCA microscopy (Elettra) evidences the formation of PtSinOm alloys as the dominating component of the islands. The CO_2 light-off curves taken for Pt/SiO_2/Si interfaces fabricated at room temperature reveal the highest CO/CO_2 conversion yield, Y, and the lowest activation temperature, T_A ∼400 K. Increasing alloying degree considerably quenches the conversion yield and shifts up the CO/CO_2 conversion onset.

O 52.9 Wed 18:00 P4

Adsorption and photocatalytic inactivation of Corona-virus like particles by anatase TiO2(101) — •Mona Kohantorabi, Michael Wagstaffe, Heshmat Noei, and Andreas Stierle — Centre for X-ray and Nano Science (CXNS), Deutsches Elektronen-Synchrotron (DESY), 22607 Hamburg, Germany

The adsorption of corona-virus like particles (VLPs), on the surface of the model catalyst TiO2(101) was investigated using different spectroscopic and microscopic techniques.

Two different methods were employed to inactivate the virus after it was loaded on the surface of TiO2: 1) UV light and 2) thermal treatment. The adsorbed virus morphology and virus particle arrangement on the surface of TiO2 were investigated using grazing-incidence small-angle scattering. Microscopic studies demonstrate that the denatured spike proteins and other virus proteins dissociate from VLPs and adsorb on the surface of TiO2.

Clarification of the interaction of the virus with the surface of semiconductor oxides will aid in obtaining a deeper understanding of the chemical processes involved in photo-inactivation of microorganisms which is important for the design of effective photocatalysts for air purification and self-cleaning materials.

O 52.10 Wed 18:00 P4

Calculation of XPS Core-Level Fingerprints of Intermediates

in Methanol Synthesis on Cu Surfaces — •Mattis Gossler, Azad Kirsan, and Bernd Meyer — Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany

$Cu/ZnO/Al_2O_3$ is used as a highly efficient catalyst in industry for the synthesis of methanol from syn gas, i.e. H_2, CO and CO_2. Despite intensive research efforts over the past decades, the precise reaction pathway and the relevant intermediates in the synthesis process are still under debate. New insights can be provided by recently performed ambient pressure x-ray photoelectron spectroscopy (AP-XPS) measurements on ZnO-supported Cu clusters with well-defined crystal terminations [1]. In order to assist the interpretation of the measured AP-XP spectra, we determined the C-1s and O-1s core level shifts for a series of intermediates in the methanol synthesis process on a variety of Cu surfaces using DFT calculations. First, a careful search for the most stable structure of all intermediates on each surface was performed. Core-level shifts were then calculated by an ΔSCF approach after removing a core electron. This fingerprint data allows us to identify the most relevant intermediates present in the AP-XPS experiments and to assess the importance of different Cu surfaces for the overall reactivity.

[1] R. Gleissner, H. Noei, S. Chung, G.D.L. Semione, E.E. Beck, A.-C. Dippel, O. Gutowski, G. Gizer, V. Vonk, A. Stierle, J. Phys. Chem. C **125** (2021) 23561–23569

O 52.11 Wed 18:00 P4

The relation between structure sensitivity and doping of ceria(111) vs. ceria(100) — Emilia Pozarowska[1], Linus Pleines[2], Mauricio J. Prieto[3], Liviu C. Tănase[3], Lucas de Souza Caldas[3], Aarti Tiwari[3], Thomas Schmidt[3], Jens Falta[2], Carlos Morales[1], and •Jan Ingo Flege[1] — [1]Applied Physics and Semiconductor Spectroscopy, BTU Cottbus-Senftenberg, Cottbus, Germany — [2]Institute of Solid State Physics, University of Bremen, Bremen, Germany — [3]Department of Interface Science, Fritz-Haber Institute, Berlin, Germany

CeO_x-Cu inverse catalysts have been shown to convert CO_2 into valuable chemicals through catalytic hydrogenation. The catalytic activity may further be enhanced by alloying ceria with trivalent, catalytically active metals, such as Sm, promoting the formation of Ce^{3+} active sites. In this work, the structural and chemical properties of (111)- and (100)- oriented CeO_x islands alloyed with samarium were explored by low-energy electron microscopy and X-ray photoemission electron microscopy. After Sm deposition on the as-grown CeO_x islands, the near-surface region of (100)-oriented CeO_x is reduced after exposure to H_2 at 470 °C, whereas the deeper layers as well as the whole (111)-oriented islands retain the Ce^{4+} state. Subsequent reoxidation with O_2 leads to the complete Ce^{4+} state recovery, suggesting the healing of oxygen vacancies. Additional annealing at 470 °C induces samarium diffusion into the ceria matrix. Yet, subsequent exposure to H_2 reduces neither the (111)- nor the (100)-oriented $CeSmO_x$ islands, suggesting a quite unexpected stability of this system.

O 52.12 Wed 18:00 P4

Operando studies of CuZn catalysts for methanol synthesis — •David Kordus[1,2], Nuria Jimenez Divins[2], Janis Timoshenko[1], and Beatriz Roldan Cuenya[1] — [1]Departement of Interface Science, Fritz Haber Institute of the Max Planck Society — [2]Departement of Physics, Ruhr University Bochum

Methanol is an essential feedstock for the chemical industry and may be a key component when switching to renewable resources. Industrially methanol production uses a ternary $Cu/ZnO/Al_2O_3$ catalyst. Especially the interaction between Cu and ZnO is critical for a high activity of the catalyst. Multiple model catalysts were synthesized to target different aspects of this system, but particularly the Cu-ZnO interactions. In-situ and operando spectroscopy methods (XAS, XPS, Raman) together with microscopy and measurements of the catalytic performance allow us to follow the chemical and structural changes of the catalysts under reaction conditions. We investigated size-selected Cu and CuZn nanoparticles on multiple support materials (SiO_2, Al_2O_3 and ZnO/Al_2O_3) to obtain a better insight into particle-support interactions. These studies show that the formation of metallic Zn for CuZn particles on SiO_2 is detrimental for the catalysts activity, as it coincides with a deactivation. On Al_2O_3 in contrast, ZnO is incorporated into the support and forms a spinel structure similar to $ZnAl_2O_4$. In another catalyst shaped Cu particles are used to create preferential facets on the surface of our catalysts. These unalike surface structures will then lead to different catalytic behavior. Here, a higher activity was observed for cubic particles.

O 53: Poster Wednesday: Spins and Magnetism

Time: Wednesday 18:00–20:00 Location: P4

O 53.1 Wed 18:00 P4

Ferroelectric BaTiO3 (001): Atomic-level characterization and polarization by combined STM/AFM — •Llorenç Albons Caldentey[1], Dominik Wrana[1], Igor Sokolović[2], Aji Alexander[1], and Martin Setvin[1] — [1]Department of Surface and Plasma Science, Faculty of Mathematics and Physics, Charles University, 180 00 Prague 8, Czech Republic — [2]Institute of Applied Physics, TU Wien, 1040 Vienna, Austria

Perovskites attract strong interest thanks to their catalytic properties, efficient electron-hole separation in light harvesting, or the frequent occurrence of ferroelectricity. In this work we show that BaTiO3 single crystals can be cleaved along the (001) plane to obtain flat surfaces with domains of either BaO or TiO2 termination. Using low temperature (4K) qPlus nc-AFM we have characterized the surface atomic structure and studied the tip-induced ferroelectric polarization of the material. The impact of ferroelectric poling on adsorbed species is discussed.

O 53.2 Wed 18:00 P4

DFT based analysis of surface reactions of stainless steels through degradation in aqueous media — •Vahid Jamebozorgi — Bielefeld University and Bielefeld University of applied science

Stainless steels (SSs) are widely used in industry due to their outstanding mechanical and physical properties plus their durability in corrosive media. The corrosion resistance of SSs is caused by the formation of a thin passive layer which mainly consists of chromium oxides. However, the passive layer makes SSs more vulnerable to localized corrosion including pitting corrosion. Pitting corrosion occurs when the passive layer interacts locally with bond-forming atoms and molecules dissolved in the aqueous media, e.g. halides or hydroxide. However, the process of bond formation, erosion of the passive layer, and consequently the degradation of SSs is still not completely understood. Amongst other parameters, the adsorption energy and work function seem to play a key role in pitting corrosion. The experimental determination of the work function is challenging since the surface texture, crystal defects and impurities can increase or decrease work function value significantly. Here, we show how density-functional theory (DFT) computation techniques can be used to obtain adsorption energies and work function values for different crystallographic orientations on the surfaces of Fe, Cr and Ni. As a result we will provide insight into the degradation mechanisms of SSs surfaces in aqueous media.

O 53.3 Wed 18:00 P4

Thermally-induced magnetic order from glassiness in elemental neodymium — •Benjamin Verlhac[1], Lorena Niggli[1], Anders Bergman[2], Umut Kamber[1], Andrey Bagrov[1,2], Diana Iuşan[2], Lars Nordström[2], Mikhail I. Katsnelson[1], Daniel Wegner[1], Olle Eriksson[2,3], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands — [2]Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden — [3]School of Science and Technology, Örebro University, SE-701 82 Örebro, Sweden

In thermodynamic systems, temperature is synonymous with disorder as phase transitions between order to disorder occur when temperature is increased. Recently, the first example of a spin-Q glass was found in elemental neodymium between 30mK and 4K(1). This phase originated from magnetic frustration within the dhcp lattice of neodymium. In this study, we show by means of spin-polarized scanning tunneling microscopy that neodymium undergoes an unusual magnetic transi-

tion, where long range multi-Q order emerges from the spin-Q glass phase as temperature is increased from 5 K to 15 K(2). We also developed a new analysis method, which analyzes the experimental data and extracts the phase transition temperature. These findings are supported by atomistic spin dynamics simulations, in which the phase transition is qualitatively explained by destroying spin frustration due to various exchange contributions of the different sublattices.
(1) Kamber, U. et al., *Science* **368** (2020)
(2) Verlhac, B. et al., arXiv:2109.04815

O 53.4 Wed 18:00 P4

Yu-Shiba-Rusinov states of Mn on Pb(110) — •Bharti Mahendru, Martina Trahms, Gaël Reecht, and Katharina J. Franke — Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany

Unpaired electron spins exchange coupled to a superconductor give rise to bound states inside the superconducting energy gap and are called Yu-Shiba-Rusinov (YSR) states. Previously, it has been shown that the crystal-field splits singly-occupied d levels of Mn atoms on Pb surfaces, which leads to distinct YSR states inheriting the symmetry of the spin-carrying orbital [1,2].
Here we investigate single atoms and chains of Mn on a Pb(110) surface using scanning tunneling microscopy and spectroscopy. We find different adsorption sites of the individual Mn atoms and chains, which can be distinguished by different YSR states. Some of the Mn sites show a bistability in the presence of the STM tip, while some of the YSR states are also influenced by the tip position.

[1] Michael Ruby, et al., Phys. Rev. Lett. 117, 186801, 2016
[2] Michael Ruby, et al., Phys. Rev. Lett. 120, 156803, 2018

O 53.5 Wed 18:00 P4

Imaging fast magnetization dynamics by Lorentz microscopy with event-based electron detectors — •Alexander Schröder, Christopher Rathje, Xinlai Xing, and Sascha Schäfer — Institute of Physics, University of Oldenburg, Germany

In recent years, ultrafast transmission electron microscopy (UTEM) [1] has successfully enabled the imaging of ultrafast dynamics on the nanoscale by utilizing femtosecond electron pulses in an optical-pump/electron-probe approach. In a potentially more flexible approach, instead of femtosecond electron pulses, a continuous electron beam could be used to map the optically induced dynamics provided sufficiently fast, event-based electron detectors are employed.

Here, we present Lorenz microscopy of nanoscale magnetic dynamics observed with a Timepix3 hybrid pixel electron detector. In particular, we study the distortion of magnetic vortices triggered by femtosecond optical pulses, accessing reversible processes on time scales ranging from nanoseconds to milliseconds.

[1] A. Feist et al., Ultramicroscopy 176, 63 (2017).

O 54: Poster Wednesday: 2D Materials 2

Time: Wednesday 18:00–20:00 Location: P4

O 54.1 Wed 18:00 P4

Mapping angle- and doping-dependent dispersion of bending graphene — •Zhihao Jiang[1], Paulina Majchrzak[1], Bjarke Jessen[2], Maëlle Kapfer[2], Deepnarayan Biswas[1], Jose Avila[3], Pavel Dudin[3], Cory Dean[2], and Søren Ulstrup[1] — [1]Aarhus University — [2]Columbia University — [3]Synchrotron SOLEIL

The possibility to systematically engineer the interlayer rotation angle (θ) between two-dimensional (2D) materials stacked in heterostructures offers an intriguing means to tailor superlattices, electronic band structures and interactions. Here, we set out to perform a proof-of-principle nanoARPES experiment demonstrating the ability to continuously twist a graphene flake stacked on hexagonal boron nitride by tuning θ using a nano-rotation device engaged by the tip of an atomic force microscope (AFM). The overarching objective is thereby to capture the evolution of the θ-dependent electronic dispersion of graphene. Ultimately, these experiments are expected to pave the way for band structure measurements of systematically twisted heterostructures composed of bilayer graphene and monolayer dichalcogenides.

O 54.2 Wed 18:00 P4

Growth and characterization of WSe$_2$ on epitaxial graphene on SiC(0001) — •Adrian Schütze, Philip Schädlich, and Thomas Seyller — Institute of Physics, TU Chemnitz, Chemnitz, Germany

2D materials such as, for example, graphene, hexagonal boron nitride or transition metal dichalcogenides have recently received much interest as building blocks for electronic devices. For a successful integration of these materials, scalable growth methods are essential. In this work we investigate the growth of WSe$_2$ by metal organic molecular beam epitaxy (MOMBE) [1] on epitaxial graphene on SiC(0001). In that process W(CO)$_6$ is used as a precursor in conjunction with selenium vapor produced by decomposition of SnSe$_2$ in a thermal evaporator. Using MOMBE we were able to grow ultra-thin films of WSe$_2$ on epitaxial graphene which were characterized by a combination of X-ray photoelectron spectroscopy (XPS), angle resolved photoemission spectroscopy (ARPES), low-energy electron diffraction and microscopy (LEED, LEEM) and atomic force microscopy (AFM). The films were observed to consist of triangular domains. We discuss the influence of the growth parameters such as the substrate temperature on the structural and electronic properties of the layers.

[1] S. Tiefenbacher et al., Surf. Sci. 318 (1994) L1161

O 54.3 Wed 18:00 P4

Vanadyl phthalocyanine: study of the formation of a characteristic molecular spin pattern on a diamagnetic template — •Corina Urdaniz, Kyungju Noh, Luciano Colazzo, Jaehyun Lee, Andreas Heinrich, Christoph Wolf, Fabio Donati, and Yujeong Bae — Center for Quantum Nanoscience (QNS), IBS, Ewha Womans University, Seoul, Republic of korea

Achieving quantum coherent control of spins on surfaces at the atomic scale is the goal for quantum coherent nanoscience. A good surface spin system requires two components: a localized spin and a buffer layer to isolate that spin from the metallic substrate. The use of magnetic molecules as hosts for spin qubits is a promising pathway towards quantum information processing.

In this work, we used Vanadyl phthalocyanine (VOPc), a well-known spin = 1/2 molecule with long coherence times up to one microsecond in its crystalline form, and the non-magnetic Titanyl phthalocyanine (TiOPc) as a buffer layer from the metal. We found that TiOPc is an effective buffer layer, preserving vacuum-like electronic structure of VOPc. We present compelling arguments for using TiOPc as self-assembling templates with long-range order for adsorbed VOPc molecules. Our results suggest that the TiOPc/VOPc system is a potential candidate for long-lived molecular spin states on surfaces for the study of quantum information processing.

O 54.4 Wed 18:00 P4

Competing Processes as Quality Limitation: New Insights into Microscopic Growth Mechanism of hexagonal Boron Nitride on Ir(111) — •Marko Kriegel[1], Karim Omambac[1], Marin Petrovic[2], Birk Finke[1], Frank Meyer zu Heringdorf[1,3], and Michael Horn-von Hoegen[1] — [1]Faculty of Physics, University of Duisburg-Essen, D-47057 Duisburg, Germany — [2]Center of Cxcellence for Advanced Materials and Sensing Devices, Institude of Physics, HR-10000 Zagreb, Croatia — [3]Interdisciplinary Center for Analytics on the Nanoscale (ICAN), D-47057 Duisburg, Germany

Despite the tremendous research effort targeting industrial growth of two-dimensional hexagonal boron nitride (hBN), especially on transition metal surfaces, until today no superior growth recipe was developed, promising large domain sizes, homogeneous lattice constants and a matching orientation of layer and substrate. Here we study single layer hBN grown on Ir(111) via conventional chemical vapor deposition (CVD) using borazine as precursor. We found two competing processes governing the density of hBN domains on a microscopic scale: Kinetic limitations at low T and the disintegration of B$_3$N$_3$ rings as well as the grown layer at high T. The interplay of both processes sets a fundamental limit to the achievable quality in this and other 2D material systems. Our study combines data from high-resolution reciprocal space mapping, using Spot Profile Analyzing Low Energy Electron Diffraction (SPA-LEED), allowing us to determine the complete distribution of domain orientations, and Low Energy Electron Microscopy (LEEM), for the measure of domain densities.

O 54.5 Wed 18:00 P4

Tuning MoS$_2$ doping by switching its support material — •Marco Bianchi[1], Daniel Lizzit[4], Paolo Lacovig[2], Charlotte Sanders[3], Davide Curcio[1], Ezequiel Tosi[2], Monika T. Schied[2], Jill Miwa[1], Silvano Lizzit[2], and Philip Hofmann[1] — [1]Dep. of Physics and Astronomy, ASTRID2, iNANO, Aarhus University, DK. — [2]Elettra Sincrotrone Trieste S.C.p.A., Trieste, IT — [3]Artemis Program, UK Central Laser Facility, Harwell, STFC, UK — [4]DPIA - University of Udine, IT

Tuning the electronic properties of a 2D crystal by the interaction with its support is the key to design well-controlled nanoelectronic devices based on transition metal dichalcogenides (TMDCs). In particular, the establishing of a low resistance between a metallic contact and the TMDC has been challenging and different strategies for this have been introduced. It was suggested that a low Schottky barrier could be achieved not only by choosing contact materials with the suitable work function but also by introducing interface defects that can contribute independent of the metal contact work function.

Here we present a combined ARPES, STM, LEED and XPS study of MoS$_2$ grown on Au(111) using well established methods. After intercalation of Bi, which is semimetallic, and its further treatment we observe a doping consistent with what was inferred from recent transport measurements. The results shown here sheds light on a potential way for tuning the effects of contacts of a 2D layer and their influence on the TMDC electronic structure.

O 54.6 Wed 18:00 P4

Subnanoscale Engineering of 2D Magnetism in van der Waals Heterostructures — •Keda Jin[1,2], Jose Martinez-Castro[1,2], Stefan F. Tautz[1,3], and Markus Ternes[1,2] — [1]Institute of Physics II B, RWTH Aachen University, 52074 Aachen, Germany — [2]Peter Grünberg Institute (PGI-3), Forschungszentrum Jülich and Jülich-Aachen Research Alliance (JARA), Fundamentals of Future Information Technology, 52425 Jülich, Germany — [3]Institute of Physics IV A, RWTH Aachen University, 52074 Aachen, Germany

The dry transferred method paves a way to investigate exotic properties and emerging new phenomena in van der Waals heterostructures. Polycarbonate (PC) is commonly used as a polymer for the dry transfer of 2D materials. A limitation of PC is the contamination of chemical residues on the surface and the difficulty to fabricate complex heterostructures. Here, we show the study of different polymers, including polyvinyl chloride (PVC) and nitrocellulose for an effective and clean way to assemble 2D heterostructures. In addition, based on our previous method of studying encapsulated air-sensitive 2D materials (Nano Lett. 18, 6696 (2018)), we show our current development in a new technique to study air-sensitive materials in heterostructures compatible with ultra-high vacuum: in-situ de-encapsulation. This technique aims to provide the required cleanliness of mechanical assembled air-sensitive van der Waals heterostructures for their study by scanning tunneling microscopy at ultra-high vacuum and low temperature.

O 54.7 Wed 18:00 P4

Growth and characterization of monolayer MnSe$_2$ on Au(111) — •Sebastien Elie Hadjadj[1], Evangelos Golias[2], Jack Hayes[1], Marcel Walter[1], Christian Lotze[1], Sangeeta Thakur[1], Ivar Kumberg[1], Ismet Gelen[1], Jorge Torres[1], and Wolfgang Kuch[1] — [1]Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany — [2]MAX IV Laboratory, Lund University, Fotongatan 2, Lund, Sweden

During the last couple of years, there has been a rising interest in novel two-dimensional magnetic materials. Most recently, several groups have shown that magnetic order in two-dimensional materials can be stable. Among them, MnSe$_2$ is interesting, since it has shown magnetic properties at room temperature [1], making it an ideal candidate for applications. However, magnetic order so far has been reported for MnSe$_2$ grown on GaSe or SnSe$_2$, and the role of the interface in the stabilization of magnetic order is not yet clear. We use molecular beam epitaxy to co-deposit Se and Mn on Au(111) to ascertain if the magnetic ordering of monolayer MnSe$_2$ is an intrinsic effect of the material or an interface-induced phenomenon. We examine the films chemically by X-ray photoelectron spectroscopy (XPS) and X-ray absorption spectroscopy (XAS) as well as structurally by scanning tunneling microscopy (STM) and low-energy electron diffraction (LEED). The latter shows a near - 3×3 pattern. The XPS measurements show chemical shifts in the Mn and Se binding energies for the MnSe$_2$ sample in comparison to pure Mn and Se that confirm that the Mn has bonded to Se. [1] D. J. O'Hara et al., Nano Letters 18, 3125 (2018).

O 54.8 Wed 18:00 P4

Towards Shot-Noise Spectroscopy of Majorana Modes in 2D Systems — •Jan Cuperus, Floris Kooij, and Ingmar Swart — Condensed Matter & Interfaces, Utrecht University, Utrecht, The Netherlands

The quasiparticles known as Majorana zero modes (MZMs) are particles that have the peculiar properties to be their own antiparticle and to obey non-Abelian statistics. Because of the latter, MZMs are predicted to be the building blocks of topological quantum computing. Over the past years, many signatures of what could be MZMs have been reported. Examples are the end modes of 1D atomic line defects in ML FeSe [1], or the edge modes found on CrBr3 island grown on NbSe2 [2]. Conclusive observations are however, still to be made. One reason for this is the similarity of MZMs to other (close-to-)zero-energy states, e.g. Yu-Shiba-Rusinov states or Andreev bound states. Scanning tunnelling microscopy (STM) has already shown to be a key tool in the field of condensed matter physics because of its extreme spatial resolution and spectroscopic abilities. Via a new STM-based tool, namely shot-noise spectroscopy, we aim to provide a new point of view onto the matter of MZMs.

References: [1]Chen et al., Nat. Phys., 16, 536-540 (2018) [2]Kezilebieke et al., Nature, 588, 424-428 (2020)

O 54.9 Wed 18:00 P4

The tellurization of Cu(111): From incorporated Te atoms via 1D-like surface reconstructions to closed films — •Andreas Raabgrund, Tilman Kisslinger, Maximilian Ammon, Lutz Hammer, and M. Alexander Schneider — Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

We investigated the tellurization of Cu(111) from low Te coverages up to thick films both structurally and electronically by STM, STS, LEED-IV structural analysis, and DFT. For $\Theta < 0.14\,\mathrm{ML}$ Te forms a substitutional surface alloy with short-range ordered patches of a $(\sqrt{7} \times \sqrt{7})\mathrm{R}19.1^\circ$ structure. Increasing the Te coverage leads to coexisting islands of a $(3 \times \sqrt{3})_{\mathrm{rect}}$ superstructure which is well-ordered and fully developed at $\Theta = 0.33\,\mathrm{ML}$ [1]. In the range $0.33\,\mathrm{ML} < \Theta < 0.40\,\mathrm{ML}$, the $(3 \times \sqrt{3})_{\mathrm{rect}}$ structure coexists next to a well-ordered $(5 \times \sqrt{3})_{\mathrm{rect}}$ phase which is completely developed at $\Theta = 0.40\,\mathrm{ML}$ [2]. Our LEED-IV analyses reveal arrangements of Cu$_2$Te$_2$ chains, whereby for the $(5 \times \sqrt{3})_{\mathrm{rect}}$ phase a substantial reorganization of the surface into troughs allows to incorporate more Te. STS finds an unoccupied chain state, that has lower energy on the $(3 \times \sqrt{3})_{\mathrm{rect}}$ which we interpret as an indication of delocalization with decreasing interchain distance. By further tellurization beyond $\Theta = 1.1\,\mathrm{ML}$ closed Cu$_{2\text{-}x}$Te films develop. For $\Theta > 4\,\mathrm{ML}$, a phase transition takes place below 300 K that results in a wrinkled appearance at low temperatures. On the surface two different reconstructions with distinct electronic fingerprint occur side-by-side. [1] T. Kißlinger et al., Phys. Rev. B **102**, 155422 (2020)

[2] T. Kißlinger et al., Phys. Rev. B **104**, 155426 (2021)

O 54.10 Wed 18:00 P4

Fabrication of nanostructured van der Waals heterostructures — •Khairi F. Elyas[1], Johanna Richter[3], Kirill Bolotin[3], Hannah C. Nerl[2], and Katja Höflich[1] — [1]Ferdinand Braun Institut gGmbH, Berlin, Germany — [2]Humboldt Universität zu Berlin, Institute of Physics, Berlin, Germany — [3]Freie Universität Berlin, Institute of Experimental Physics, Berlin, Germany

Two-dimensional (2D) materials can exhibit a significantly enhanced light-matter interaction making them interesting for highly-confined and low-loss light transport. When combining different 2D materials, the different polaritonic modes may hybridize to combine the strong localization of plasmonic excitations with the long propagation distances of phonon modes. Here we report on the fabrication of heterostructures of the (semi)metallic graphene and the wide-bandgap material hexagonal boron (hBN) nitride. The dry-release transfer of graphene and hBN makes use of polydimethylsiloxane(PDMS) and poly(propylene) carbonate (PPC) films. Due to the strong adhesion between PPC and 2D materials at room temperature, we show that single-layer to few-layer graphene as well as few-layer hBN can be produced on a spin coated PPC film/SiO2/Si substrates by mechanical exfoliation. Using He ion beam patterning we further modify the geometry of the heterostructures on the nanoscale with the specific aim to tune hybrid polaritonic modes. The optical properties of the fabricated heterostructures are then mapped using monochromated low-loss scanning transmission electron microscopy (STEM) electron energy-loss spectroscopy (EELS)and outcomes compared to optical methods.

O 54.11 Wed 18:00 P4

Interlayer charge transport anomalies in 1T-TaS$_2$ and 2H-NbS$_2$ — Edoardo Martino, László Forró, and •Konstantin Semeniuk — Institute of Physics, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland

Layered van der Waals materials, such as transition metal dichalcogenides, have emerged as promising platforms for realising functional materials for practical applications. Tuning interlayer coupling in these systems via twist, intercalation, exfoliation, etc. opens up a vast parameter space for inducing exploitable properties. Probing of the out-of-plane conduction is a useful tool for gauging the subtle interactions between the atomic planes, particularly in bulk crystals of TMDs.

We present comprehensive studies of charge transport anisotropies of bulk 1T-TaS$_2$ and 2H-NbS$_2$. The samples were tailored using focused ion beam in order to ensure homogeneous current flow across the principal crystallographic directions. We find that resistivity anisotropy of 1T-TaS$_2$ is, paradoxically, of the order of unity, with the out-of-plane conduction becoming more preferred at lower temperatures due to a formation of c-axis-oriented quasi-one-dimensional orbital chains. In 2H-NbS$_2$, we observe a pronounced upturn of the out-of-plane resistivity upon cooling. We attribute the anomaly to a unidirectional Kondo scattering, caused by inherent inclusions of 1T-NbS$_2$ layers, which host a lattice of singly occupied orbitals.

[1] E. Martino et al., npj 2D Mater. Appl., **4**, 7 (2020).

[1] E. Martino et al., npj 2D Mater. Appl., **5**, 86 (2021).

O 54.12 Wed 18:00 P4

STM studies of graphene encapsulated Fe3GeTe2 — •Tobias Wichmann[1,2,3], Felix Lüpke[1,2], and F. Stefan Tautz[1,2,3] — [1]Peter-Grünberg-Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]Jülich Aachen Research Alliance (JARA) - Fundamentals of Future Information Technology, Germany — [3]Institut für Experimentalphysik IV, RWTH Aachen, Germany

Fe$_3$GeTe$_2$ is a metallic 2D ferromagnet with a Curie Temperature of 130K, in which Kondo effect and skyrmions have been observed. These properties make it a promising candidate as a source of magnetic proximity effect in van der Waals heterostructures.

To examine its potential use in in van der Waals heterostructures we investigated the influence on adjacent layers of 2D materials through encapsulation with graphene. The resulting structure was characterized in a low temperature STM.

O 54.13 Wed 18:00 P4

Fabrication of nanostructured van der Waals heterostructures — •Khairi F. Elyas[1], Johanna Richter[3], Kirill Bolotin[3], Hannah C. Nerl[2], and Katja Höflich[1] — [1]Ferdinand Braun Institut gGmbH, Berlin, Germany — [2]Humboldt Universität zu Berlin, Institute of Physics, Berlin, Germany — [3]Freie Universität Berlin, Institute of Experimental Physics, Berlin, Germany

Two-dimensional (2D) materials can exhibit a significantly enhanced light-matter interaction making them interesting for highly-confined and low-loss light transport. When combining different 2D materials, the different polaritonic modes may hybridize to combine the strong localization of plasmonic excitations with the long propagation distances of phonon modes. Here we report on the fabrication of heterostructures of the singlecrystalline gold flakes or graphene and the wide-bandgap material hexagonal boron (hBN) nitride. Polydimethylsiloxane(PDMS) and poly(propylene) carbonate (PPC) films were used for the dry transfer of gold and hBN due to the strong adhesion between PPC and 2D materials at room temperature. Using this method, single-layer to few-layer hBN were successfully transferred. Using He ion beam patterning we further modify the geometry of the heterostructures on the nanoscale with the specific aim to tune hybrid polaritonic modes. The optical properties of the fabricated heterostructures are then mapped using monochromated low-loss scanning transmission electron microscopy (STEM) electron energy-loss spectroscopy (EELS)and outcomes compared to optical methods.

O 54.14 Wed 18:00 P4

Carbon Embedding of Pt Cluster Superlattices Templated by Hexagonal Boron Nitride on Ir(111) — •Tobias Hartl[1], Moritz Will[1], Pantelis Bampoulis[1,2], Virginia Boix de la Cruz[3], Paolo Lacovig[4], Vedran Vonk[5], Simon Chung[5], Andreas Stierle[5], Jan Knudsen[3], Silvano Lizzit[4], and Thomas Michely[1] — [1]Universität zu Köln — [2]University of Twente — [3]MAX IV Laboratory and Division of Synchrotron Radiation Research — [4]Elettra-Sincrotrone Trieste S.C.p.A — [5]DESY Hamburg

With the goal to delevop the fabrication of a new type of Pt-nanoparticle carbon support electrocatalyst, we investigate the carbon embedding of Pt cluster superlattices grown on the moiré of a monolayer of h-BN on Ir(111). Using STM and XPS we find that carbon embedding is conformal and does not deteriorate the excellent order of the clusters. The thermal and mechanical stability of the embedded clusters is greatly enhanced by the C forming a strong binding to the Pt clusters. Sintering as well as single cluster pick-up by the STM tip, are both suppressed. (Hartl, T. et al., J. Phys. Chem. C, 2021)

The only cluster decay path left takes place at an elevated temperature above 850 K. Cluster material penetrates through the h-BN sheet, whereby it becomes bound to the underlying metal. There are indications that while the a-C matrix and the Pt clusters bind strongly to each other, upon annealing both weaken their binding to h-BN.

We discuss how the binding between the membrane and the substrate can be weakened, such that it is possible to be split-off via a combination of hydrogen bubbling and dry transfer approaches.

O 55: Poster Wednesday: Organic Molecules at Surfaces 2

Time: Wednesday 18:00–20:00 Location: P4

O 55.1 Wed 18:00 P4

Self-assembly and debromination of a functionalized borazine on Ag(111) — •Birce Sena Tömekce[1], Marc G. Cuxart[1], Martina Crosta[2], Marco Franceschini[2], Daniele Poletto[2], Davide Bonifazi[2], and Willi Auwärter[1] — [1]Physics Department E20, Technical University of Munich, Germany — [2]Institute of Organic Chemistry, University of Vienna, Austria

On-surface synthesis is a promising route towards the generation of doped graphene nanoarchitectures with tunable electronic properties [1]. To fabricate atomically precise hybrid BNC materials, distinct precursors incorporating BN units can be used [2]. In this study, we employed a precursor with a borazine core and Br and OH functionalization. We report on a low-temperature scanning tunneling microscopy/spectroscopy and X-ray photoelectron spectroscopy characterization of well-ordered phases of this precursor on Ag(111). At low temperatures, the molecules adsorb intact. At moderate temperatures, debromination is activated and the self-assembly of a chiral kagomé lattice coexisting with a hexagonal packing is observed. At elevated temperatures, completion of cyclodehydrogenation leads to full planarization of the molecules, followed by covalent intermolecular coupling, thereby forming random BNC-based networks. We further investigated the electronic structure of the kagomé lattice on the single molecule level. Our findings constitute a step towards exploiting the structural and electronic properties of BNC architectures.

[1] R. Pawlak et al., Angew. Chem. Int. Ed. 2021, 60, 8370-8375

[2] C. Sánchez-Sánchez et al., ACS Nano. 2015, 9, 9228-9235

O 55.2 Wed 18:00 P4

The investigation of self-assembly molecule on metal substrate — •Yong-He Pan and GERMAR Hoffmann — Department of Physics, National Tsing Hua University

Field-effect transistors (FETs) made out of organic materials are lighter, mechanically more flexible, have lower costs and have higher field-effect mobility. For picene-$(C_{14}H_{29})_2$ the highest field-effect mobility in all FETs was recorded. Here, phenacene-$(C_{14}H_{29})_2$ is deposited on Au(111) by thermal evaporation and is investigated by STM and Scanning Tunneling Spectroscopy (STS) at 70 K under UHV conditions. The self-assembly molecule forms a rhombohedral unit cell with a two molecular basis, and the alkyl chain, flexible up to 15 degrees, is nearly parallel with the nearby molecules. Scanning close to the bandgap region of the phenacene$(C_{14}H_{29})_2$ leads to imaging of the LUMO-state, which also shows the pronounced electronic state on the phenacene core and energetically broad state on the alkyl chain.

O 55.3 Wed 18:00 P4

adsorption structure and mechanical properties of single nonahelicene molecules on Ag(110) — •Max Halbauer[1,2], Akitoshi Shiotari[1], Takashi Kumagai[3], Kyoko Nozaki[2], and Martin Wolf[1] — [1]Department of Physical Chemistry, Fritz-Haber-Institute of the Max-Planck-Society, Faradayweg 4-6, 14195 Berlin, Germany — [2]Department of Chemistry and Biotechnology, School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, 113-8656 Tokyo, Japan — [3]Center for Mesoscopic Sciences, Institute for Molecular Science, 38 Nishigo-Naka, Myodaiji, 444-8585 Okazaki, Japan

Helicenes are a class of compounds that has received great attention due to their chiroptical properties, while the single-molecule mechanical behavior has often been overlooked. Scanning tunneling microscopy (STM) and atomic force microscopy (AFM) measurements of nonahelicene ([9]H) molecules on Ag(110) were performed in order to address this issue. The adsorption structure of isolated and aggregated [9]H molecules on the surface is revealed by high-resolution imaging. Interactions of the molecule with a probe tip are quantified by force curve measurements and a comparison with co-adsorbed coronene gives insights into the impact of the helical backbone on the mechanical properties.

O 55.4 Wed 18:00 P4

Changes in the coupling of a single-molecule magnetic moment to a Cooper pair condensate by the sequential removal of molecular moieties — •Stefan Schulte[1], Nicolas Néel[1], Krisztian Palotas[2], and Jörg Kröger[1] — [1]Institut für Physik, Technische Universität Ilmenau, Ilmenau, Germany — [2]Institute for Solid State Physics and Optics, Wigner Research Center for Physics, Budapest, Hungary

Using the tip of a STM chemical reactions are induced in organic 5, 10, 15, 20-tetrakis(4-bromophenyl)-porphyrin-cobalt molecules adsorbed on superconducting Pb(111). Two chemical reactions, the dehalogenation and dephenylation of the molecules are presented. Yu-Shiba-Rusinov bound states occur in the superconductor energy gap only after the entire dephenylation of the molecule. The presence of the intragap resonances is related to an unoccupied molecular orbital that is confined to the atomic magnetic center (Co) of the porphyrin. From the energy position and spectral weight of these intragap resonances, a weak interaction between the molecular magnetic moment and the Cooper pair condensate is inferred. The electron-hole asymmetry of the bound states exhibits a spatial oscillation with a wavelength that evidences the exchange coupling of the molecular magnetic moment to the sp-sheet of the Fermi surface. The very low binding energy of the Yu-Shiba-Rusinov levels is consistent with the observed absence of the Kondo effect. Funding by the DFG through KR 2912/10-1 and KR 2912/10-3 is acknowledged.

O 55.5 Wed 18:00 P4

Pentacene on prototypical antiferromagnets: A photoemission study — •Valentin Mischke, David Janas, Giovanni Zamborlini, Jonah E. Nitschke, and Mirko Cinchetti — Department of Physics, TU Dortmund University, Otto-Hahn-Straße 4, 44227 Dortmund, Germany

In the last years, so-called molecular spinterfaces have been intensively studied because of the novel spin properties raising from the molecule-metal hybridization [1]. Recently, antiferromagnets have been considered as an alternative to their ferromagnetic counterparts for the engineering of spinterfaces with novel spin properties. Here, we present the preliminary characterization of a single layer of pentacene atop two prototypical antiferromagnets, namely nickel oxide NiO(111) and iron oxide FeO(100). Both thin films have been grown in-situ, under UHV conditions, by depositing Ni and Fe in a O_2 background atmosphere onto Au(111) and oxygen-passivate Fe(100), respectively. The resulting interfaces have been characterized with low energy electron diffraction and momentum microscopy. In addition, photoemission tomography was employed to determine the energy level alignment of the molecular orbitals at the interface.

[1] Cinchetti, Dediu, Hueso, Nature Materials 16, 507 (2017)

O 55.6 Wed 18:00 P4

Systematically mapping the distance-dependent tip-sample interaction for the PTCDA/Ag(111) system — •Tim Dierker and Philipp Rahe — Fachbereich Physik, Universität Osnabrück, Barbarastraße 7, 49076 Osnabrück, Germany

Scanning probe microscopy (SPM) has been continually improved by establishing a number of controlled tip functionalizations [1]. In particular, the attachment of a single 3,4,9,10-perylene-tetracarboxylic-dianhydride (PTCDA) molecule to the apex of a metallic tip enables mapping of the electrostatic potential near sample surfaces by so called scanning quantum dot microscopy [2,3]. In order to reliably apply this technique, the expedient pick-up of a single molecule is required [4]. Here, the tip-molecule interaction between a metallic tip and surface-adsorbed molecules is investigated by means of systematic scanning tunneling (STM) and atomic force (AFM) microscopy measurements. In particular the vertical dependencies of the STM tunneling current and the AFM frequency shift are mapped along different axes of single PTCDA molecules embedded in the molecular film on Ag(111). Fingerprints for the dynamic behavior of the molecules are clearly revealed and guide the vertical manipulation for molecular pick-up.

[1] L. Gross, Nat. Chem. 3, 273 (2011)
[2] C. Wagner et. al., PRL 115, 026101 (2015)
[3] M. F. B. Green et. al., JJAP 55, 08NA04 (2016)
[4] M. F. B. Green et. al., Beilstein J. Nanotech. 5, 1926 (2014)

O 55.7 Wed 18:00 P4

Photoemission and Raman Spectroscopic Studies of n-GaAs(100) Surface Passivation with Thioglycolic Acid — •Alexander Ehm, Oleksandr Selyshchev, and Dietrich R. T. Zahn — Semiconductor Physics, TU Chemnitz, Chemnitz D-09107, Germany

Gallium arsenide is one of the most investigated inorganic semiconductors and used in a vast variety of applications and prospective for new high-performance devices. A challenge in constructing such devices is a surface of native oxides causing a high surface density of states, which leads to the mid-gap pinning of the surface Fermi level, band bending, and the formation of a surface depletion layer. Sulphur passivation yields a significant reduction of the depletion layer and related effects but requires several treatment steps [1].

We report a new simple approach utilizing thioglycolic acid (TGA) to provide a one-step effective removal of surface oxides and protection from reoxidation compared to etching with inorganic acids. This effect is confirmed by X-ray photoemission spectroscopy and the reduction of the depletion layer is confirmed by Raman spectroscopy results. n-GaAs(100)/poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) solar cells show improved performance for such passivated n-GaAs(100) surfaces.

[1] V. N. Bessolov, M.V. Lebedev, D. R. T. Zahn: J. Appl. Phys. 82 (5) (1997)

O 55.8 Wed 18:00 P4

Vibrational quanta of single melamine on Cu(100) — •Rebecca Cizek, Nicolas Néel, and Jörg Kröger — TU Ilmenau

Melamine adopts an up-standing adsorption geometry on Cu(100) and can be tautomerized by the local injection of electron from the tip of a scanning tunneling microscope [1]. Here, we compare inelastic electron tunneling spectroscopy (IETS) of intact and tautomerized melamine. Two low-energy vibrational quanta are observed for both molecules. The tautomer exhibits an energy shift and enhancement of one of the excitations. These findings are moreover complemented by IETS and tautomerization of the deuterated molecule.

[1] R.-P. Wang et al., J. Phys. Chem. Lett. 12, 1961 (2021)

O 55.9 Wed 18:00 P4

Photoemission orbital tomography of NiTPP molecules deposited on the passivated Fe(100)-p(1x1)O surface — •Michael Gutnikov[1], David Janas[1], Jonah Nitschke[1], Mira Arndt[1], Vitaliy Feyer[2], Giovanni Zamborlini[1], and Mirko Cinchetti[1] — [1]Department of Physics, TU Dortmund University, Otto-Hahn-Straße 4, 44227 Dortmund, Germany — [2]Peter Grünberg Institut (PGI-6), Forschungszentrum Jülich, Leo-Brandt-Straße, 52425 Jülich, Germany

Recently, it was shown that a single layer of oxygen is sufficient to decouple nickel porphyrins from a ferromagnetic surface, thus, preserving most of the electronic features of the pristine molecules [1]. However, distortions of the molecular structure, e.g. upward-bent phenyl-rings, can make the interpretation of surface imaging techniques like STM extremely challenging.

Additional information can be gained by photoemission tomography (PT), which combines angle-resolved photoelectron spectroscopy (ARPES) with ab-initio calculations of the gas phase molecules to interpret the orbital arrangement of thin molecular layers.

This work provides a characterization of NiTPP molecules on an oxygen passivated Fe(100)-p(1x1)O surface using PT, focusing not only on the energy level alignment but also on their azimuthal orientation with respect to the substrate.
[1] G. Albani et al. Micromachines **12**, 191 (2021)

O 55.10 Wed 18:00 P4

Surface Chemical Bond of Alternant vs. Non-Alternant Aromatic Isomers — •Florian Münster[1], Lukas Ruppenthal[1], Leonard Neuhaus[1], Jan Herritsch[1], Jon Henrick Both[1], Pengcai Liu[2], Xing-Yu Chen[2], Jiawen Cao[2], Xiao-Ye Wang[2], and J. Michael Gottfried[1] — [1]Fachbereich Chemie, Philipps-Universität Marburg, Germany — [2]College of Chemistry, Nankai University, Tianjin, China

The different effects of the topology of pyrene and cyclohepta[*fg*]acenaphthylene (acepleiadylene) on the occupied and the unoccupied electronic states as well as on the desorption process from the Cu(111) surface are studied using PES, NEXAFS and TPD. Both molecules are aromatic but differ in their topology. While pyrene has an alternating structure, its constitutional isomer acepleiadylene has a non-alternating one. With TPD, we showed that the desorption of acepleiadylene begins at about 340 K, 40 K higher than for pyrene, indicating a stronger bond to the Cu(111) surface. Using the modified leading edge analysis we find a desorption energy of 152 kJ/mol for acepleiadylene compared to 108 kJ/mol for pyrene, each at monolayer coverage. Furthermore, combining PES and NEXAFS, we were able to assess the energy needed in the excitation process of electrons originating from occupied valance orbitals into unoccupied ones. Here we find a 0.5 eV smaller energy difference for the non-alternating species in comparison to the alternating species.

O 55.11 Wed 18:00 P4

New Photon Scanning Tunnelling Microscope for studying electrically driven single photon emitters in the GHz range — •Andreas Reutter[1,2], Mike Stummvoll[1,2], Neda Noei[1], Markus Etzkorn[1,2], and Uta Schlickum[1,2] — [1]Institut für Angewandte Physik, Technische Universität Braunschweig, Mendelssohntraße 2, 38106 Braunschweig — [2]Laboratory for Emerging Nanometrology, Langer Kamp 6a/b, 38106 Braunschweig

Single atoms and molecules have always been an interesting research area. An important tool for investigating such is scanning tunnelling microscopy (STM) which has become a widely used method to characterize not only the surface density states and vibrational excitations but also electrical excitations and recombination processes of individual atoms and molecules by STM-induced luminescence (STML).

We would like to present a state-of-the-art, self-build, low-temperature, ultra-high vacuum STM with the possibility for STML measurements and for the time-resolved probing of electrically driven single photon emitters with a band width of up to around 20 GHz. This allows for instance the investigation of charge transfer in molecules below the nanosecond range and further insight into its exciton decay.

O 55.12 Wed 18:00 P4

Anchoring *p*-terphenyl-based thiols to top-layer sulphur defects on MoS_2/Au(111) — J. Rika Simon[1], Dmitrii Maksimov[2], •Juan Pablo Guerrero Felipe[1], Paul Wiechers[1], Christian Lotze[1], Ana M. Valencia[3,4], Caterina Cocchi[3,4], Björn Kobin[4], Stefan Hecht[4], Mariana Rossi[2], and Katharina J. Franke[1] — [1]Freie Universität Berlin, Germany — [2]MPI for the Structure and Dynamics of Matter, Hamburg, Germany — [3]Carl von Ossietzky Universität Oldenburg, Germany — [4]Humboldt-Universität zu Berlin, Germany

Systems consisting of transition metal dichalcogenides (TMDCs) and organic molecules is a field gathering much interest in recent years. By using a scanning tunneling microscope (STM), it is possible to explore these systems in detail, where the TMDC layer works as a decoupling layer between the adsorbate and metal substrate. In our experiments, we investigate a monolayer of MoS_2 on Au(111) where interesting results occur once a sulphur defect on the top-layer is created. It gives rise to a localized resonance around the Fermi energy, not present in pristine MoS_2. Here we show experimental and theoretical comparisons where the thiol-based molecule CF3-3P-SH (4”-(trifluoromethyl)-[1,1':4',1”-terphenyl]-4-thiol) is anchored to a sulphur top-layer defect, creating a chemical bond between the molecule and MoS_2. We observe a characteristic Kondo resonance due to the interaction between molecule and substrate. Additionally, we have mapped out the spatial distribution of the electronic states as well as explored the vibronic states of the molecule.

O 55.13 Wed 18:00 P4

On-Surface Transmetalation of a Lead-Porphyrin on the Cu(111) surface — •Jan Herritsch, Stefan R. Kachel, Qitang Fan, Mark Hutter, Lukas J. Heuplick, Florian Münster, and J. Michael Gottfried — Philipps-Universität Marburg

Starting from porphyrin complexes, highly ordered nanostructures can be assembled on surfaces in which reactive metal centers are firmly anchored in a defined environment. Such structures have enormous potential in various areas of modern technology. A reaction in which there is an exchange of the central atom by another element leads to a drastic change in the properties of the functionalized surface. Here, we report on a thermally induced Pb/Cu metal exchange of lead(II)-tetraphenylporphyrin (Pb(TPP)) on the Cu(111) surface. Using temperature-dependent XPS, we were able to track this exchange reaction and the accompanying change of the Pb oxidation state by probing the Pb 4f level. The reaction starts already below 380 K and is completed at 600 K. In parallel, partial desorption of a monolayer occurs above 430 K. In a temperature-programmed reaction experiment (TPR), the desorbing species are unambiguously identified as the product, Cu(TPP), of the metal exchange by mass spectrometry. By STM, the adsorbate structure of Pb(TPP) on Cu(111) were revealed and individual free Pb atoms, which are formed in the course of the Pb/Cu metal exchange, were observed. Moreover, side-reactions of the peripheral phenyl substituents occur due to dehydrogenative coupling reactions.

O 55.14 Wed 18:00 P4

Tip-enhanced Raman Spectroscopy of a lifted single PTCDA molecule — •Rodrigo Cezar de Campos Ferreira[1], Jiří Doležal[1,2], Sofia Canola[1], Prokop Hapala[1], Pablo Merino[3], and Martin Švec[1] — [1]Institute of Physics,Czech Academy of Science;, Czech Republic — [2]Faculty of Mathematics and Physics, Charles University; Czech Republic — [3]Instituto de Ciencia de Materiales de Madrid; CSIC, Madrid, Spain

Advanced scanning probe techniques in the research of molecular adsorbates on surfaces have been instrumental in unveiling fundamental quantum phenomena with high sensitivity and spatial resolution at the nanoscale level. Such advances impact areas of molecular electronics, nanophotonics, and near-field spectroscopies. Tip-enhanced Raman spectroscopy (TERS) is a technique that provides access to vibrational modes of individual molecules via the extremely confined plasmonic field at the STM tip apex. This can be also used as a probe to chemically scrutinize single bonds with subnanometer precision. In this work, the STM-controlled TERS technique in a UHV environment at low temperature was performed for the perylene tetracarboxylic dianhydride molecule (PTCDA), in which we measured the Raman fingerprint for an array of PTCDAs on Ag(111) as well as for a single molecule. Moreover, we were able to simultaneously follow the conductance spectrum of the system while lifting a single PTCDA from the surface. The observed transition in the Raman spectra correlates with the differential conductance showing the transition of the system from S=0 neutral to S=1/2 anion state.

O 55.15 Wed 18:00 P4

Electronic properties of CuPc/TiSe2 heterostructures — •Hibiki Orio[1,2], Kiana Baumgärtner[1,2], Christian Metzger[1,2], Markus Scholz[3,4], Kai Rossnagel[4,5], and Friedrich Reinert[1,2] — [1]Universität Würzburg Experimentelle Physik VII, Würzburg, Germany — [2]Würzburg-Dresden Cluster of Excellence ct.qmat, Würzburg Dresden, Germany — [3]European XFEL Facility, Schenefeld, Germany — [4]Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [5]KiNSIS, Universität Kiel, Kiel, Germany

Transition metal dichalcogenides (TMDC) are a class of quasi-2D materials that exhibit a variety of electronic properties including superconductive, excitonic insulator, and charge-density wave (CDW) phases. TiSe2 is one of the most prominent TMDC because it has intriguing CDW properties. The physical process of the CDW is not fully understood, however, and is attributed to either be purely electron-electron or jointly electron-electron and electron-phonon mediated. To gain more insight into the CDW buildup, modulating the intrinsic physical properties is useful [1]. For this purpose, we have used organic molecules as several interactions such as charge transfer or a rearrangement of the electron density can modulate the electronic properties at the organic/TMDC interface. We have evaporated copper phthalocyanine (CuPc) on TiSe2 single crystals. We report the perturbed electronic characteristics of the resulting CuPc/TiSe2 heterostructure examined with angle-resolved and X-ray photoelectron spectroscopy.

[1] K. Baumgärtner et al., submitted.

O 55.16 Wed 18:00 P4

Chirality-induced electron spin filtering in chiral helicene mono-, double- and multilayers — •Ruwen Quenter[1], Paul Valerian Möllers[1], Karl-Heinz Ernst[2], and Helmut Zacharias[1] — [1]Center for Soft Nanoscience, WWU Münster, Germany — [2]Empa, Dübendorf, Switzerland

The transmission yield of electrons through molecules with chiral structure can depend on the electron spin. This phenomenon is established as chirality-induced spin selectivity (CISS).[1] While initial demonstrations of CISS were conducted with, e.g., short DNA strands,[2] studies with simpler molecules such as helicene potentially allow for a better insight into the mechanism. Previous work[3] on CISS in ordered monolayers of heptahelicene ([7]H) demonstrated that while the preferentially transmitted spin orientation depends on the helicity of the molecules, no major influence of the substrate is evident. Since the CISS increases with the number of helical turns, we investigate how it evolves for double and multilayers of [7]H adsorbed on a Cu(332) surface. Photoelectrons were excited from the substrate by deep-UV laser pulses and transmitted through [7]H mono-, double or multilayers. Subsequently, the average spin polarization of the photoelectrons was measured via Mott scattering. With a [7]H monolayer a spin polarization of $P \approx 8\%$ was shown earlier[3] and reproduced now. For the double and multilayers no increased spin polarization magnitude was found. [1] D. H. Waldeck et al., APL Mater. **9**, 040902 (2021) [2] B. Göhler et al., Science **331**, 894 (2011) [3] M. Kettner et al., J. Phys. Chem. Lett. **9**, 2025 (2018)

O 56: Poster Wednesday: Nanostructures 2

Time: Wednesday 18:00–20:00 Location: P4

O 56.1 Wed 18:00 P4

Pt Wedge on h-BN/Rh(111): Structural Analysis by HR-XPS — •Natalie J. Waleska, Fabian Düll, Florian Späth, Udo Bauer, Philipp Bachmann, Johann Steinhauer, and Christian Papp — Friedrich-Alexander-Universität, Erlangen, Germany

An approach to overcome the material gap to commercial catalysts is the investigation of metal nanoclusters on 2D materials (e.g. h-BN). To date, mostly 3D metal clusters with a narrow size distribution on such support materials were studied. However, the clusters' structure and thus the available adsorption sites, as well as the catalytic activity varies strongly from 3D to monolayer clusters or even single atoms.

To investigate the structural differences of clusters with varying sizes, a Pt wedge was prepared in a single preparation step on the h-BN/Rh(111) substrate, ranging from 0.21 to 0.001 ML Pt coverage. The analysis was performed using HR-XPS and CO as a probe molecule. From the data, we were able to determine the transition from monolayer to 3D cluster formation for the as-prepared Pt wedge. Upon heating to 550 K, structural changes of the Pt clusters were observed as a result of cluster ripening and sintering.

We thank Helmholtz-Zentrum Berlin for allocation of synchrotron-radiation beamtime and BESSY II staff for support during beamtime. This work was funded by the DFG within SFB 953 "Synthetic Carbon Allotropes" (Project #182849149).

O 56.2 Wed 18:00 P4

Charge effects on the (de-)Hydrogenation activity of Pt$_n$-clusters on a-SiO$_2$ thin films — •Tobias Hinke[1], Andrew Crampton[1], Marian Rötzer[1], Maximilian Krause[1], Florian Schweinberger[1], Bokwoon Yoon[1], Uzi Landman[2], and Ueli Heiz[1] — [1]Physical Chemistry, Department of Chemistry & Catalysis Research Center, Technical University of Munich, 85748 Garching, Germany — [2]School of Physics Georgia Institute of Technology, Atlanta GA, U.S.A.

Catalytic model systems facilitate the gain of fundamental insights on molecular mechanisms and enable atom-precise manipulation of catalytic processes. Besides the size-effects of clusters with less than 100 atoms, the influence of the supporting material and the substrate are of special interest when steering reactivity. The catalyst's local electron density can be altered by varying the underlying metal. A high local work function of the substrate, Mo(112), yields clusters of positive charge while a low local work function , Pt(111), results in a negative charge. This allows for specifically altering the reactivity as well as the coking stability of the catalytic system.

In order to elucidate the impact of different support materials as well as cluster size-effects the change in activity of a-SiO$_2$ thin-film supported Pt$_n$-clusters towards ethylene (de-)hydrogenation was investigated.

The catalytic samples were characterized in a UHV setup with base pressures below $5 * 10^{-10}$ mbar through IRRAS and electron spectroscopy (XPS, MIES, UPS), while reactivity was monitored with TPD and pulsed valve experiments (p-MBRS).

O 56.3 Wed 18:00 P4

Design of an Enhanced Ce-Evaporator for Ce$_x$O$_y$ Thin Film Synthesis — •Flora Siegele, Kevin Bertrang, Tobias Hinke, and Ueli Heiz — Physical Chemistry, Department of Chemistry and Catalysis Research Center, Technical University of Munich

In today's industry, the importance of sustainability regarding resource consumption and waste management is strongly increasing and hence, the contribution of catalysis-based processes is becoming more and more important. Substantial advances can be achieved by investigating catalytic model systems to facilitate the understanding of basic mechanisms on a molecular level. These model catalysts comprise a metal single crystal substrate with metal clusters (1-100 atoms), deposited on a metal oxide thin film (2-20 ML). Especially the lewis acidic and basic properties of metal oxidic compounds showed promising results in terms of controlled tuning of the catalytic performance. However, the reducibility of thin film materials like CeO$_2$ offers an additional possibility of adjusting catalytic properties. Due to the lack of experimental results focusing on the influence of CeO$_2$ thin films on the reactivity and selectivity of clusters, the poster presents the implementation of an optimized CeO$_2$ thin film generation procedure with a home-built ribbon evaporator. With this evaporator design, including a self-built quartz crystal microbalance, deposition rates of multiple monolayers per minute could be achieved and tracked with high accuracy. Electron emission spectroscopy (x-ray photoelectron spectroscopy and Auger electron spectroscopy) was performed to investigate the composition of the deposited film.

O 56.4 Wed 18:00 P4

CO Adsorption on PdPt Alloy Nanoparticles — •Daniel Silvan Dolling[1,2], Jan-Christian Schober[1,2], Marcus Creutzburg[1], Heshmat Noei[1,2], and Andreas Stierle[1,2] — [1]Universität Hamburg — [2]DESY

Platinum palladium alloy nanoparticles are of high interest because of their role as catalysts for different processes, including exhaust control and methane oxidation. Catalyst behavior is determined by the shape, structure and alloy composition of the particles. To enhance catalyst efficiency, it is thus necessary to improve our understanding of the structure of the nanoparticles and the active adsorption sites. In order to access detailed structural and morphological information, we employ the use of a model catalyst. In this work, platinum and palladium are co-deposited via molecular beam evaporation on α-Al2O3 (0001). The nanoparticles are grown epitaxially and have a well defined height to diameter ratio and alloy composition. By investigating the particle surface with the probe molecule CO using polarized infrared reflection absorption spectroscopy (IRRAS), we determined the surface species on top and side facets of the particles. The morphology was further identified with X-ray diffraction and X-ray reflectivity. The effect of the Pd/Pt alloy composition on the adsorption was investigated by comparing the IRRAS reflectivity. Moreover, the difference in the adsorption of CO was studied for samples annealed in hydrogen and oxygen.

O 56.5 Wed 18:00 P4

Methane activation with small Ta clusters — •Kevin Bertrang, Tobias Hinke, Nikita Levin, Martin Tschurl, and Ueli Heiz — Physical Chemistry, Department of Chemistry and Catalysis Research Center, Technical University of Munich

With the exhaustion of petroleum reserves, methane will become an important feedstock for the synthesis of fuels and fine chemicals. The challenge is to find an efficient way to activate the highly inert molecule under mild conditions and steer the reaction towards the formation of chemically precious products while preventing coking.

Studies of small cationic Ta-clusters in the gas phase and their oxides were found to exhibit high activity towards non-oxidative C-C-coupling of methane, yielding dehydrogenated carbohydrate species and ethane. The cluster charge was identified as a key parameter for activity. These studies are extended to their supported analogues. To replenish the cluster charge acidic (SiO_2) and reducible (CeO_2) thin metal-oxide films are employed and cluster oxygen content is tuned.

Characterization is performed by means of vibrational (IRRAS) and electron spectroscopy (XPS) and reactivity is studied via TPD and pulsed valves experiments.

[1] N. Levin et al. *J. Am. Chem. Soc.* **2020**, 142, 12, 5862-5869

O 56.6 Wed 18:00 P4

3d-Nanoparticles on Graphene: Influence of Temperature — •Kai Besocke, Mahboobeh Ravankhah, and Mathias Getzlaff — Institut für Angewandte Physik, Heinrich-Heine-Universität Düsseldorf

With its unique properties, such as high quality crystal structure, excellent electrical conductivity and high tensile strength, graphene is a promising substrate for fabricating nanocomposites. In this context we are investigating the influence of graphene as substrate for the deposition of metallic nanoparticles and the influence of susequent heating. In this contribution we present our results concerning the mobility and distribution of $Fe_{0.5}Ni_{0.5}$-Nanoparticles on graphene surfaces.

A W (110) single crystal serves as the substrate being coated with a Co thin film of about 20 ML and annealed at temperatures up to 500 °C. Surfaces prepared in such a way exhibit elongated Co-islands with a width of several hundred nm and height up to 10 nm, which are coated with graphene subsequently. For graphene synthesis the samples are heated in a Propene atmosphere of 10^{-6} mbar for several minutes. The nanoparticles are produced by means of Ar magnetron sputtering in a Haberland source and aggregation takes place in a He atmosphere. The spherical particles under investigation have diameters of several nm. Particle distributions are analyzed via STM, both as-prepared as well as after heating. It will be discussed, whether the nanoparticles are more mobile on graphene compared to other surfaces.

O 57: Poster Wednesday: Electronic Structure

Time: Wednesday 18:00–20:00 Location: P4

O 57.1 Wed 18:00 P4

Josephson effect in the two-band superconductor Niobium Diselenide — •Xianzhe Zeng[1], Haonan Huang[1], Sujoy Karan[1], Klaus Kern[1,2], and Christian Ast[1] — [1]Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany — [2]Institut de Physique, Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

Niobium Diselenide is a two-dimensional (2D) van der Waals s-wave superconductor that can be described by a two-band model. The Josephson effect between Niobium Diselenide and another known BCS superconductor is non-trivial since it involves Copper pair tunneling of two bands with different order parameters. Here, we measure quasiparticle tunneling as well as the Josephson current between a clean superconducting Niobium Diselenide surface and a superconducting Vanadium tip using low-temperature scanning tunneling microscopy and spectroscopy. We analyze our results with the McMillan formula for a two-band superconductor. Our observations present a first step towards understanding more complicated scenarios, such as the predicted p-wave triplet superconductivity in Chromium Tribromide islands on Niobium Diselenide.

O 57.2 Wed 18:00 P4

Bulk Ion Conductivity and Near Surface Composition of Ionic Liquid and Zwitterionic Salt Based Electrolytes for Lithium Battery Applications — •Fabian Ullmann, Joel Tayo, Anna Dimitrova, and Stefan Krischok — Institut für Mikro und Nanotechnologien, Institut für Physik, TU Ilmenau

In this contribution we focus on the bulk conductivity and the near surface composition of several tertiary electrolytes which consist of Ionic Liquid (IL), Zwitterionic salt (ZwS) and lithium salt. Two ILs and ZwSs are chosen: IL: 1-ethyl-3-methylimidazolium bis-(trifluoromethylsulfonyl)imide - [EMIm][Tf2N] and 1-butyl-1-methylpyrrolidinium bis-(trifluoromethylsulfonyl)imide - [BMP][Tf2N]; ZwS: 3-(3-methylimidazolium-1-yl)Propane-1- sulfonate and 3-(3-vinylimidazolium-1-yl)Propane-1-sulfonate. As lithium precursor - bis(trifluoromethylsulfonyl)imide - Li[Tf2N] was used. Different Li[Tf2N]/ZwS ratios are considered and studied. Electrochemical Impedance Spectroscopy (EIS) enables us to determine the ion conductivity of the electrolytes. The results reveal that the presence of ZwSs enhances the ion conductivity, although they itself are not ion conductive. Further, by using X-Ray Photoelectron Spectroscopy (XPS) we analyze the near surface chemical composition at UHV-conditions. The XPS analysis displays a cation/anion/ZwS distribution as depended on the concentration of Li[Tf2N] and the type of IL and ZwS used. The spectroscopic results revealed solute-solvent interactions which modify the ion mobility.

O 57.3 Wed 18:00 P4

Line shape analysis of the resonant eigenstates in a quantum corral by means of tunneling spectroscopy and non contact AFM — •Marco Weiss, Michael Roessner, Fabian Stilp, and Franz J. Giessibl — Institute of Experimental and Applied Physics, University of Regensburg, Germany

Back in 1993 Crommie et al. [1] arranged 48 Fe adatoms on a Cu(111) surface in a circle with a diameter of 14.26 nm. This quantum corral confines surface electrons in a circular potential well. Past investigations with scanning tunneling microscopy revealed energetically discrete eigenstates that spatially appear as Bessel functions. But these studies on the Fe quantum corral showed unwanted movement of the corral walls during spectroscopic measurements. [1]

Instead of Fe, we used CO molecules to provide the corral with more stable walls. This permitted us to access a larger voltage window and allowed for a detailed line shape analysis of the corrals eigenenergy levels. Surprisingly, we discovered a large Gaussian broadening of the eigenstates.

We also acquired nc-AFM data of the corral. As we have previously done, we determined the occupation of electronic states that cross the Fermi level [2]. Similar to our previous work, we observed that the presence of the probe tip shifts the energy of the electronic states under study. In this work, we show that we can also use the AFM data to estimate the lifetime (i.e. the spectral width) of these states.

[1] M. F. Crommie et al., Science 262, 218-220 (1993)

[2] F. Stilp et al., Science 372, 1196-1200 (2021)

O 57.4 Wed 18:00 P4

Interaction between an artificial and a natural atom — •Fabian Stilp, Marco Weiss, and Franz J. Giessibl — University of Regensburg, Regensburg, Germany

The surface state of Cu (111), a quasi-2-dimensional electron gas, is trapped to a small surface area of about 15 x 15 nm^2 by placing 48 CO-molecules in a circular shape on the surface via atomic manipulation. By doing so, one creates a quantum corral with discrete electronic states forming an artificial atom. This structure can be described reasonably well by an infinitely high circular potential well leading to corral states with Bessel-type radial functions and an angular momentum normal to the surface. To investigate the interaction between this artificial atom and a natural atom we bring Fe atoms inside the corral and measure the response of the corral states.

Thanks to the large size, one can study the structure of the wave functions within that artificial atom by nc-AFM showing an angular dependence of the corral states after placing the atoms inside the corral. This change of the wave functions leads to an energy shift of a few meV confirmed by tunneling spectroscopy measurements. By investigating the change of the wave functions and the energies of the corral states when placing the adatom inside, one can draw conclusion about the interaction between this artificial atom and a natural atom. Here we expand the interpretation of the adatom acting repulsively on the corral states as stated by Stilp et al. [1].

[1] F. Stilp, A. Bereczuk, J. Berwanger, N. Mundigl, K. Richter, F.J. Giessibl, Science 372, 1196-1200 (2021)

O 57.5 Wed 18:00 P4

Spontaneous Charge Localization on Polar Surfaces — •Michele Reticcioli[1], Zhichang Wang[2,3], Zdenek Jakub[2], Igor Sokolovic[2], Matthias Meier[1,2], Gareth S. Parkinson[2], Michael Schmid[2], Dominik Wrana[4], Lynn A. Boatner[5], Ulrike Diebold[2], Martin Setvin[2,4], and Cesare Franchini[1,6] — [1]Faculty of Physics and Center for Computational Materials Science, University of Vienna, Austria — [2]Institute of Applied Physics, TU Wien, Vienna, Austria — [3]State Key Laboratory for Physical Chemistry of Solid Surfaces, Xiamen University, China — [4]Department of Surface and Plasma Science, Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic — [5]Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, USA — [6]University of Bologna, Italy

Excess charge on polar surfaces of ionic compounds is commonly described by the two-dimensional electron gas (2DEG) model, a homogeneous distribution of charge confined in a few atomic layers. Conversely, our density functional theory calculations and scanning-probe microscopy measurements on $KTaO_3$(001) show spontaneous localization of the excess charge in the form of polarons, bipolarons and charge density waves. These electronic reconstructions form on the defect-free surface, and alter the material properties and functionalities to different degrees. Controlling the degree of charge ordering could be of great benefit for a wide range of applications: in our study, we analyze the impact on the surface reactivity by considering the interaction with CO molecules.

O 57.6 Wed 18:00 P4

Evolution of Property and Bonding Maps — •Carl-Friedrich Schön and Matthias Wuttig — RWTH Aachen University, Aachen, Germany

Since picking up the first tool, it has been the goal of mankind to create materials that best suit human needs. While for the longest time any development in this field was driven by an empirical approach, modern means of physics and chemistry gave rise to the concept of material and property maps based on chemical bonding. We have composed a database of elemental and binary compounds. For all compounds, the corresponding values of a set of properties were included, containing the conductivity, the Born Effective Charge, the Effective Coordination Number (ECoN) and the Bandgap, the melting point, the density and the atomic density. A Gaussian mixture algorithm was utilized to separate the compounds in the database into n clusters. With the number of allowed clusters set to 4, the algorithm nicely separates materials which employ metallic, ionic and covalent bonding. This implies that properties can be used to distinguish bonding mechanisms. Interestingly, a fourth class of materials is identified, characterized by a property portfolio neither found in metals, or materials employing ionic or covalent bonding. This is further support for the concept of metavalent bonding, a novel bond type characterized by the competition between localization and delocalization. This classification provides a close link between chemical bonding mechanisms and properties enabling novel routes to material's design with material maps.

O 57.7 Wed 18:00 P4

A theoretical investigation into gallic acid pyrolysis — •Jakob Kraus and Jens Kortus — Institute of Theoretical Physics, TU Bergakademie Freiberg, Leipziger Str. 23, D-09599 Freiberg, Germany

Thermodynamical and kinetic information on the first two steps of gallic acid pyrolysis is calculated based on density functional theory and quantum chemistry. For the kinetics, transition states are identified with the help of the climbing image nudged elastic band method. Both reactions exhibit two transition states. One of them is related to the rotation of OH groups, and the other one is related to the breaking and forming of bonds. The gallic acid pyrolysis as a whole is judged to be endothermal, and it changes from endergonic to exergonic between 500 K and 750 K. The second reaction step, the dehydrogenation of pyrogallol, is identified as the limiting step of gallic acid pyrolysis, with reaction rate constants below $1\ s^{-1}$ for temperatures below 1250 K.

O 58: Poster Wednesday: New Methods and Developments, Frontiers of Electronic Structure Theory

Time: Wednesday 18:00–20:00 Location: P4

O 58.1 Wed 18:00 P4

Fully Atomistic Modelling of Tip-enhanced Raman Spectra from First Principles — •Yair Litman, Franco Bonafe, Alaa Akkoush, Heiko Appel, and Mariana Rossi — MPI for the Structure and Dynamics of Matter, Hamburg, Germany.

Tip-enhanced Raman scattering (TERS) has emerged as a powerful tool to study surfaces with subnanometer spatial resolution [1]. In particular, single-molecule TERS studies have shown the capability to visualize high-resolution images of individual molecular normal modes in real space [2]. Theoretical simulations that can provide an unambiguous interpretation and atomic description of obtained TERS images often rely on crude approximations of the local electric field [3]. In this work, we present a novel method to compute TERS images by combining Time Dependent Density Functional Theory (TDDFT) and Density Functional Perturbation Theory (DFPT) to calculate Raman cross sections with realistic local fields. The new approach allows for a fully *ab initio* atomistic description of the tip-molecule-surface system, and naturally incorporates chemical effects arising from the molecule-surface interaction. We show results for benzene and pyridine and discuss the importance of a realistic description of the local field, and self consistent evaluation of the electronic density response. Finally, we evaluate the use of 2D TERS imaging as an identification tool for defects in 2D materials. [1] M. Richard-Lacroix, *et al.*, Chem. Rev. **56**, 3922 (2017) [2] L. Joonhee, *et al.*, Nature **568**, 78 (2019) [3] P. Liu, X. Chen, H, Ye, and L. Jensen, ACS Nano **13**, 9342 (2019)

O 58.2 Wed 18:00 P4

Deep learning based signal processing for touch-sensitive surfaces — •Jakob Elsner, Viktor Fairuschin, and Thorsten Uphues — Institute for Sensor and Actuator Technology, Coburg, Germany

Touch-sensitive surfaces enable intuitive and efficient operation of electronic devices and eliminate the need for external peripherals and mechanical components, making touch technology increasingly important in the modern society. However, conventional touch technologies, i.e. capacitive, resistive or optical, are usually limited to non-metallic materials that hardly meet the stringent requirements for robustness and hygiene in a medical environment. Stainless steel is one of the most commonly used materials in medical fields due to its high strength, chemical resistance and excellent hygienic properties. In this work, we present a novel approach based on Lamb wave technology and deep learning analytics, and apply this new principle to design a stainless steel touch-sensitive surface. Compared to Rayleigh wave-based touch technology, our approach requires no additional reflective structures and involves only a single piezoelectric transducer used to monitor the entire surface, while position-sensitive information is extracted from raw Lamb wave signals using a trained deep neural network.

O 58.3 Wed 18:00 P4

Enhanced Sampling of Surface Reactions Using Boltzmann Generators — •David Hering, Johannes T. Margraf, and Karsten Reuter — FHI Theory Department, Berlin, DE

Computational surface science and catalysis research is still mainly conducted with static density functional theory (DFT) calculations. This approach is computationally convenient, but misses important aspects of surface chemistry, such as anharmonic free energy contributions. In principle, DFT-based molecular dynamics (MD) simulations (ideally combined with enhanced sampling algorithms) would allow a much more accurate description of these processes. Unfortunately, these are far too expensive to be routinely applied to complex surface/adsorbate systems. This is due to the fact that configurations in MD are generated sequentially. As a consequence, MD configurations are not statistically independent so that a very large number of samples is required to obtain converged ensemble properties. To overcome this limitation, Noé and co-workers recently proposed a generative machine learning model called the Boltzmann Generator, which was used to generate independent configurations of biomolecules. In this contribution, we explore how Boltzmann Generators can also be used to sample the free energy surface of surface/adsorbate systems relevant for heterogeneous catalysis. In particular, training protocols and validation metrics of generated ensembles will be discussed.

O 58.4 Wed 18:00 P4

theoretical and experimental investigation of Fe and Ni-TCNQ on graphene — •azin shahsavar, zdeněk jakub, anna kurowská, jakub planer, ondrej herich, lenka černá, lukáš kormoš, pavel procházka, and jan čechal — CEITEC Brno University of Technology Purkyňova 656/123 612 00 Brno, Czech Republic.

Due to the outstanding properties of the 2D metal-organic frameworks (MOF), intensive computational and experimental studies have been done. However, the lack of fundamental studies of MOFs on the graphene backbone is observed. This work studies Fe and Ni as metal and tetracyanoquinodimethane (TCNQ) with a high electron affinity as an organic linker functionalized on graphene. Here we present DFT calculations results to unveil the electronic and magnetic properties of iron and nickel-TCNQ physisorbed on graphene. Adsorption and Fermi energies, structural, and magnetic properties will be reported. Our experimental observations prove Fe- and NiTCNQ@Gr/Ir(111) are thermally highly stable up to 500 and 250 °C, respectively, making them promising materials for single-atom catalysts or high-density storage media [1]. [1] Z. Jakub et al., Nanoscale., 1-9 (2022). DOI: 10.1039/d2nr02017c

O 58.5 Wed 18:00 P4

Assessment of Structural Descriptors for the Construction of High-Dimensional Neural Network Potentials — •Moritz R. Schäfer[1], Jonas Finkler[2], Stefan Goedecker[2], and Jörg Behler[1] — [1]Georg-August-Universität Göttingen, Institut für Physikalische Chemie, Theoretische Chemie, Tammannstraße 6, 37077 Göttingen, Germany — [2]Basel University, Department of Physics and Astronomy, Klingelbergstrasse 82, 4056 Basel, Switzerland

High-dimensional neural network potentials (HDNNPs) can be used to efficiently compute close-to ab initio quality energies and forces for performing large-scale molecular dynamics simulations of complex systems. In this method, the total energy is constructed as a sum of environment-dependent atomic energy contributions. Also electrostatic interactions based on environment-dependent charges can be included. Hence, a set of reliable structural descriptors for the atomic local environments is crucial to develop accurate potentials. Often, atom-centered symmetry functions (ACSFs) are used for this purpose in HDNNPs. In this work, we benchmark the accuracy and transferability of HDNNPs with respect to alternative descriptors like the recently proposed overlap matrix descriptor.

O 58.6 Wed 18:00 P4

Fortnet, a software package for training Behler-Parrinello neural networks — •Tammo van der Heide[1], Jolla Kullgren[2], Peter Broqvist[2], Vladimir Bačić[3], Thomas Frauenheim[4,5,1], and Bálint Aradi[1] — [1]BCCMS, University of Bremen, Bremen, Germany — [2]Dept. of Chemistry - Ångström Laboratory, Uppsala University, Uppsala, Sweden — [3]Dept. of Physics and Earth Sciences, Jacobs University Bremen, Bremen, Germany — [4]Beijing CSRC, 100193 Beijing, P. R. China — [5]Shenzhen JL CSAR Institute, Shenzhen 518110, P. R. China

A new, open source, parallel, stand-alone software package (Fortnet) has been developed, which implements Behler-Parrinello neural networks. It covers the entire workflow from feature generation to the evaluation of generated potentials, coupled with higher-level analysis such as the analytic calculation of atomic forces. The functionality is demonstrated by driving the training for the fitted correction functions of the density functional tight binding (DFTB) method, which are commonly used to compensate the inaccuracies resulting from the DFTB approximations to the Kohn-Sham Hamiltonian. Their usual two-body form limits the transferability of parameterizations between very different structural environments. After investigating various approaches, we have found the combination of DFTB with a near-sighted artificial neural network, acting on-top of baseline correction functions, the most promising one. It allows to introduce many-body corrections on top of two-body parametrizations, while excellent transferability to deviating chemical environments could be demonstrated.

O 58.7 Wed 18:00 P4

Machine learning enhanced DFTB method for periodic systems — •Wenbo Sun, Guozheng Fan, Tammo van der Heide, Adam McSloy, Thomas Frauenheim, and Bálint Aradi — Bremen Center for Computational Materials Science, University of Bremen, Am Fallturm 1, Bremen 28359, Germany.

The Density Functional based Tight Binding (DFTB) is an approximative density functional based quantum chemical simulation method with low computational costs. In order to increase its accuracy, we have introduced a machine learning algorithm to optimize several parameters of the DFTB method, concentrating on solids with defects. The backpropagation algorithm was used to reduce the error between DFTB and DFT results w.r.t. the training dataset and to obtain adjusted DFTB Hamiltonian and overlap matrix elements. Afterwards, the generalization capability of the trained model was tested for geometries not being part of the training set. In the current work, we have focused on defective periodic silicon and silicon carbide systems as target materials and the density of states (DOS) as target property to demonstrate the feasibility of our approach. The trained model was able to reduce the differences between the DFTB and the DFT DOS significantly, while other derived properties (e.g. charge distribution, partial DOS) remained physically sound. Also, the transferability of the obtained model could be verified. Our method allows to carry out relatively fast simulations with high accuracy and only moderate training efforts, and represents a good compromise for cases, where long range effects make direct machine learning predictions difficult.

O 58.8 Wed 18:00 P4

Electronic properties of Density Functional Tight Binding by Machine Learning — •Guozheng Fan[1], Adam McSloy[1], Balint Aradi[1], Chi-Yung Yam[2], and Thomas Frauenheim[1,2] — [1]Bremen Center for Computational Materials Science (BCCMS), University of Bremen, Bremen, Germany — [2]Beijing Computational Science Research Center (CSRC), Beijing, China

We have introduced a machine learning workflow, which could optimize electronic properties in density functional tight binding method. With this workflow, we can train and predict electronic properties in a cheap, accurate and transferable way. The implementation features of batch calculations greatly improve the calculation efficiency, especially for high throughput calculations. This workflow could optimize electronic properties by train basis functions or train a spline model to generate two center integrals for off-diagonal and onsite for diagonal Hamiltonian and overlap. The results show that compared with previous Slater-Koster parameters, the dipole moments, charges, the ratios of the on-site populations and the atomic numbers in charge population analysis method can be improved by both tuning basis function parameters or optimizing integrals in spline model directly. The training on basis functions could prevent the two center integrals go randomly and keep Hamiltonian and overlap in reasonable range. Besides, the multiple electronic properties could be improved simultaneously.

O 58.9 Wed 18:00 P4

Unsupervised regression-based measures for applications on atomistic features — •Alexander Goscinski[1], Guillaume Fraux[1], Giulio Imbalzano[1], Félix Musil[1,2], Sergey Pozdnyakow[1], and Michele Ceriotti[1] — [1]Laboratory of Computational Science and Modeling, Institute of Materials, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland — [2]National Center for Computational Design and Discovery of Novel Materials (MARVEL), Lausanne, Switzerland

The quality of the features as input for a machine learning model is a crucial factor for the prediction quality and the computational efficiency. Commonly, to assess the quality of features, they are compared by benchmarking the regression performance on several properties. Complementary to such a quality assessment, this work presents certain measures for direct feature-to-feature comparisons without the need of a target property. These measures are used to quantify the capacity of features representing geometrical space in atomistic applications and derive an understanding of the information encoded in features.

O 58.10 Wed 18:00 P4

Accelerating plane-wave-based *ab initio* molecular dynamics by optimization of Fast-Fourier transforms for modern HPC architectures — •Christian Ritterhoff, Tobias Klöffel, Sagarmoy Mandal, and Bernd Meyer — Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany

The most important advantage of plane-wave basis sets is that wave functions can be transformed efficiently from reciprocal to real space and back by using the Fast-Fourier transform (FFT) algorithm. This allows to evaluate the kinetic and potential energy in reciprocal and real space, respectively, where both operators are diagonal. This reduces the computational cost for applying the Hamilton operator from N^2 to $N \log N$. However, the scalability of current FFT libraries is rather limited on today's HPC systems, which offer large numbers of compute nodes, each of them with many cores. Here we present our optimization of the FFTX library of the Quantum Espresso software package. Data distribution and communication patterns have been revised to make optimal use of combined MPI and OpenMP parallelization. Scalability is further increased by combining FFTs into batches and by introducing overlapping computation and communication. We implemented the revised FFTX library in our optimized version of the CPMD code [1], and we demonstrate the achieved acceleration by a series of benchmark simulations.

[1] T. Klöffel, G. Mathias, B. Meyer, Comput. Phys. Commun. **260** (2021) 107745

O 58.11 Wed 18:00 P4

Surface tension measurement of pure water in vacuum — •Paul T. P. Ryan, Jiri Pavelec, Jan Balajka, Michael Schmid, and Ulrike Diebold — Institute of Applied Physics, TU Wien, Austria

Very little is known about the surface tension of pure liquids in contact with their pure gaseous phases, i.e. without the presence of other gases or liquid phase contaminants. This is surprising given that contaminants are known to greatly affect surface tensions values[1]. Recently we have developed a method to dose liquid water onto pristine surfaces in UHV using a small cryostat [2,3]. We combine this approach with the pedant drop method [4] to measure the surface tension of ultra-clean liquids in contact with their pure gaseous phases. The upgraded version of the small cryostat, replaces the syringe typically used in the pendant-drop method. The ultra-clean liquid is condensed onto a small cryostat placed in a vacuum chamber. A pendant drop is formed and carefully photograph allowing the surface tension of the liquid to be directly determined. The design of the apparatus will be discussed and preliminary measurements of ultra-clean water will be presented. [1] Yuki Uematsu, et. al., Current Opinion in Electrochemistry, Volume 13, (2019) [2] Jan Balajka, et. al. , Review of Scientific Instruments 89, (2018) [3] Jan Balajka, et. al., Science, 361, (2018) [4] Berry, J. D. et. al., J. Coll. Interface Sci. 454, 226*237, (2015).

O 58.12 Wed 18:00 P4

Home-Built UHV Suitcase — •Luca Lezuo[1], Lucie Dockalová[2], Gareth Parkinson[1], Ulrike Diebold[1], and Jiri Pavelec[1] — [1]Institute of Applied Physics, TU Wien, Wiedner Hauptstrasse 8-10/134, Vienna, Austria — [2]Institute of Physical Engineering, Brno University of Technology

Due to their extreme sensitivity to adsorbing molecules, most surface experiments have to be carried out in ultra-high vacuum (UHV). Ideally, multiple different techniques are used to explain and understand the phenomena happening on an atomic scale. To this end, it is often necessary to transfer a sample from one chamber to another.

As a showcase, we discuss the analysis of perovskite oxides, produced by pulsed laser deposition (PLD) as thin films, in another UHV chamber that allows atomically resolved STM/AFM imaging with a Q+-sensor at liquid N_2/He - temperatures. The home built UHV suitcase consists of three stages divided by gate valves. It has a scroll pump for rough vacuum, a turbo pump to reach high vacuum, a NEG pump and a cryopump to ensure a clean transfer and an ION/NEG combination to provide UHV conditions long term in the storage stage, where the sample is transported.

O 58.13 Wed 18:00 P4

Wettability investigation of microscale water droplets on silicon substrate using atomic force microscopy — •Mohammadali Hormozi[1], Marvin Hoffer[1,2], Pauline Brumm[2], and Regine von Klitzing[1] — [1]Soft Matter at Interfaces, Department of Physics, Technical University of Darmstadt, 64289 Darmstadt, Germany — [2]Institute of Printing Science and Technology (IDD), Technische Universität Darmstadt, Magdalenenstraße 2, 64289 Darmstadt, Germany

The wettability of a particular substrate by a liquid drop is of interest in many scientific fields. This phenomenon is often described by the contact angle between the considered liquid and substrate. This parameter is an important boundary condition, especially for the wetting of mixtures and solutions. This study shows a methodology for evaluating the contact angle of different droplets with a base diameter down to 0.5 micrometers. 3D topography of water droplets -generated through condensation and Inkjet printing on a silicon substrate- has been determined using Atomic Force Microscopy. Also, the topography of printed mixture droplets including water-glycerol and water-glycerol-isopropanol has been measured. Different curves have been fitted to a 2D cross-section of each droplet, which provides information about their contact angle. The contact angle of the mixtures deviates from the macroscale contact angle at the vicinity of the three-phase contact line; however, this phenomenon cannot be seen in pure water droplets. While no effect of droplet diameter could be detected for diameters, ranging from 0.5 to 30 micrometers, the macroscopic contact angle of droplets is several degrees higher.

O 58.14 Wed 18:00 P4

High-frequency shot-noise STM to study correlated electron systems — •Maialen Ortego Larrazabal[1], Jiasen Niu[2], Koen M Bastiaans[3], Jianfeng Ge[2], Tjerk Benschop[2], Milan P Allan[2], and Ingmar Swart[1] — [1]Debye Institute for Nanomaterials Science, Utrecht University, PO Box 80000, 3508 TA Utrecht, The Netherlands — [2]Leiden Institute of Physics, Leiden University, Niels Bohrweg 2, 2333 CA Leiden, The Netherlands — [3]Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, Netherlands

The fluctuations in time of a measured signal provide information that is not present in the time averaged value. For example, the discreteness of the electric charge leads to fluctuations in the tunneling current in an STM, known as shot-noise. Shot-noise measurements convey information about the correlations among the electrons in condensed matter systems, such as the effective charge of the carrier or their distribution in the tunneling process. However, other contributions to the measured noise, such as 1/f and thermal noise, make it difficult to isolate the shot-noise component. For this reason, we use custom-built electronics that allows us to read out the noise signal of the STM at high frequencies and cryogenic temperatures and that does not interfere with conventional STM measurements.

O 58.15 Wed 18:00 P4

Optimization of a Simple Electrospray Deposition Device — •Ken Kolar[1], Miriam Meyer[2], Henrik Siboni[1], Christophe Nacci[1], Grant Simpson[1], and Leonhard Grill[1] — [1]Institute for Chemistry, Department of Physical Chemistry, University of Graz, Austria — [2]Institute for Ion Physics and Applied Physics, University of Innsbruck, Austria

A commercially available design of an electrospray deposition apparatus was optimized for cleaner and more controllable deposition in a high vacuum. The apparatus consists of a series of 5 differentially pumped chambers separated by skimmer cones or apertures. An angle-adjustable bellow was introduced between the second and third pumping chamber for better control over the alignment and, consequently, the ion beam flux reaching the final stage. Monitoring the flux/alignment was done by current measurements around the apertures and behind the last one with a conductive probe. Also, heating of the transfer capillary and enclosing the emitter-transfer capillary interface in a small transparent chamber (to allow different ambient gas environments) were introduced. The first depositions were done to test the performance of the improved design with some well-studied molecules.

O 59: Poster Wednesday: Plasmonics and Nanooptics 2

Time: Wednesday 18:00–20:00 Location: P4

O 59.1 Wed 18:00 P4

Quantum description of the optical response in metal nanoparticles — •Jonas Grumm, Robert Salzwedel, Malte Selig, and Andreas Knorr — Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Berlin, Germany

The optical response of metal nanoparticles is dominated by the formation of collective electronic resonances, so-called plasmons.

Here, we present a microscopic approach for their temporal dynamics based on the self-consistent treatment of microscopic Boltzmann transport equations and macroscopic Maxwell equations for the electromagnetic fields. Numerical simulations describe the thermalization of the phonons and the relaxation of the electrons upon optical excitation and allow to include nonlinear optical processes in the description.

O 59.2 Wed 18:00 P4

Access to hot electron dynamics in a nanotip via ultrafast THz-streaking — •Dominik Weber, Felix Sommer, Moritz Heindl, and Georg Herink — Experimental Physics VIII - Ultrafast Dynamics, University of Bayreuth, Germany

Field enhancement and localization of single-cycle Terahertz radiation at metallic nanotips forms the basis for a broad range of emerging ultrafast interactions - ranging from ultrafast nearfield and tunnelling microscopy, transient carrier dynamics to strong-field phenomena. We present an experimental access to ultrafast hot electron dynamics at the apex of a free-standing nanotip based on THz-nearfield streaking spectroscopy [1,2]. Using tunable fs-pulses, we induce non-equilibrium electron excitations and transient hot electron distributions. Access to local hot electron dynamics confined to the apex is provided via nonlinear sub-cycle field emission at the peak of the enhanced single-cycle THz-waveform. Based on experimental streaking waveforms, we discuss the impact of excitation conditions on hot electron relaxation and possibilities for external control of ultrafast carrier dynamics.

L. Wimmer et al., "Terahertz control of nanotip photoemission", Nature Physics 10 (2014)

G.Herink et al., "Field emission at terahertz frequencies: AC-tunneling and ultrafast carrier dynamics", New Journal of Physics 16 (2014)

O 59.3 Wed 18:00 P4

Near-field optical investigation of bandgap effects in SnTe — •Christian Justus, Konstantin G. Wirth, Dario Siebenkotten, Lukas Conrads, Sophia Wahl, Matthias Wuttig, and Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

Phase change materials (PCMs) are prime candidates for non-volatile memory solutions[1]. Storage mediums with high memory density require a method for determining material properties with nanometer precision. One such tool to investigate the optical properties of nanostructures is scattering-type scanning near-field optical microscopy (s-SNOM). Thus far, bandgap effects in semiconductors have eluded experimental observation in s-SNOM. Specifically metavalently bonded materials may exhibit a bandgap shift by over a factor of three in reduced dimensions, such as in the case of Tin Telluride (SnTe) with a shift from 0.2 to 0.7 eV[2]. The primary goal of this work is to use s-SNOM to spectoscopically characterize bandgap effects in SnTe thin films. We find good agreement between Fourier-transform infrared spectroscopy (FTIR) and s-SNOM measurements over a spectral range spanning 0.3 to 0.9 eV and are able to identify a bandgap contribution to the spectral s-SNOM data. Our results may help to get a deeper insight into confinement effects on the optical properties of nano-scale PCM structures, such as PCM memory cells, and may provide a valuable tool for the analysis and characterization of PCM memory devices with high storage density.

O 59.4 Wed 18:00 P4

Energy and momentum distribution of surface plasmon-induced hot carriers — •Christopher Weiss[1], Eva Prinz[1], Michael Hartelt[1], Benjamin Stadtmüller[1,2], and Martin Aeschlimann[1] — [1]Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Germany

Investigating the energy and momentum space signature of plasmon-induced hot electrons is essential for understanding novel plasmonic energy conversion schemes. The question remains, if plasmon-induced and photon-induced hot carriers are fundamentally different. For the bulk plasmon resonance, a fundamental difference is known, yet for the technologically important surface plasmons this is far from being settled. Just recently, we identified a similar characteristic signature in the surface plasmon polariton (SPP) emission that distinguishes them from photon-induced electrons [1].

To separate the energy and momentum distribution of the plasmon-induced hot electrons from those of the photoexcited electrons, we employ a two-colour femtosecond time-resolved 2-photon photoemission (2PPE) experiment. The spatial evolution of the photoemitted electrons was observed with energy-resolved photoemission electron microscopy (PEEM) and momentum microscopy during the propagation of an SPP pulse along a gold surface. Building on these findings, we extend this concept to SPPs of single crystalline silver surfaces to investigate the influence of the band structure and material properties.

[1] Hartelt et al., ACS Nano 15, 12 (2021), 19559–19569

O 59.5 Wed 18:00 P4

Near-field optical investigation of few layer graphene — •Lina Jäckering, Konstantin Wirth, Christian Justus, and Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

Few layer graphene (FLG) samples usually consist of various stacking orders, which show different optical and electronical properties. Direct imaging and characterization of stacking domains in FLG samples can be done with scattering scanning near-field optical microscopy (s-SNOM) [1]. S-SNOM is outstanding for its nanoscale resolution of about 20 nm and its capability to image buried structures [2]. Thus, it allows to characterize encapsulated FLG samples as they are used in transport devices. Previous s-SNOM investigations focussed on the excitation of free charge carriers in FLG at about 0.1 eV [3,4]. Recently, FLG stackings have been identified by exciting interband transitions (0.27 − 0.56 eV) [1,5]. Interband resonances of FLG have only been measured at punctual energies or small energy regimes. Here, we investigate trilayer graphene and bilayer graphene in the spectral range of 0.27−0.9 eV, providing a deeper understanding of their near-field optical response, particularly of their interband resonance. Furthermore, we identify different stacking domains in FLG samples encapsulated in hexagonal boron nitride. Our findings provide a promising technique to select stacking domains for fabrication of transport devices.

1. Kim et al., ACS Nano, **9**, 7 (2005); 2. Jeong et al., Nanoscale, **9**, 12 (2017); 3. Fei et al., Nature **487**, 82 (2012); 4. Jiang et al., Nat. Mater. **15**, 840 (2016); 5. Wirth et al., arXiv: 2203.07971v1 (2022)

O 59.6 Wed 18:00 P4

Photoemission electron microscopy of Ag nanostructures on silicon substrates — •Muhamed Sewidan, Katharina Engster, Kevin Oldenburg, Sylvia Speller, and Ingo Barke — Rostock University, Rostock, Germany

We study electronic and optical coupling phenomena between plasmonic nano-objects, organic molecules, and the substrate. Localized plasmons created in metal nanostructures can lead to enhanced multiphoton photoelectron emission under pulsed light excitation. In a photoemission electron microscope (PEEM) individual particles of sub-15 nm size can be easily distinguished, providing access to the single-particle plasmon properties for a large number of species simultaneously [1]. We present results on the spatially resolved photoemission dependence on wavelength, polarization, and the surface composition for size-selected Ag particles and nanostructure arrays prepared by nanosphere lithography on silicon. [1] K. Oldenburg et al., J. Phys. Chem. C 123, 1379 (2019)

O 59.7 Wed 18:00 P4

Inelastic electron-photon scattering with broadband optical pulses — •Niklas Müller, Rasmus Lampe, Gerrit Vosse, Christopher Rathje, and Sascha Schäfer — Institute of Physics, University of Oldenburg, 26129 Oldenburg, Germany

The inelastic scattering of fast electrons at spatially confined light fields has recently enabled new techniques in ultrafast transmission electron microscopy (UTEM) [1,2,3] but is typically performed with narrow bandwidth light pulses. Here, we study the interaction of fast electrons with broadband, strongly chirped light fields. Optical pulses are generated in a home-build noncollinear optical parametric amplifier (NOPA) [4] with a spectral width of up to 200 nm in the visible range. Light reflection at an aluminum coated silicon nitride membrane leads to an intense multicolor near-field at which we inelastically scatter femtosecond electron pulses forming photon sidebands in the electron energy spectrum. The spectral sideband position changes with the relative timing between electron and light pulse according to the instantaneous frequency of the driving laser field. Different parameter regimes of electron and light chirp and their impact on inelastic scattering patterns are discussed.

[1] G. De Abajo and M. Kociak, New J. Phys. 10, 073035 (2008) [2] Barwick et al., Nature 462, 902-906 (2009) [3] Feist et al., Nature 521, 200-203 (2015) [4] G. Cerullo and S. De Silvestri, Rev. Sci. Instrum. 74, 1 (2003)

O 59.8 Wed 18:00 P4

Probing Excitons-Photon interaction in WSe2 beyond the Non-Recoil Approximation — •Fatemeh Chahshouri, Masoud Taleb, and Nahid Talebi — Institute of Experimental and Applied Physics, Kiel University, 24118 Kiel, Germany

Cherenkov radiation from electrons propagating in materials with a high refractive index have applications in particle-detection mechanisms. However, the theory of the Cherenkov radiation has been treated up to now using the non-recoil approximation, which neglects the effect of electron deceleration in materials. Here, we report on the effect of the electron-beam deceleration on the radiated spectrum and exciton-photon interactions in nm-thick WSe2 crystals beyond the non-recoil approximation. The calculation of the Cherenkov radiation is performed by simulating the energy distribution of an electron beam propagating inside a thick WSe2 using the Monte Carlo method, and ascertaining the radiating power from electron beams with Liénard-Wiechert retarded potentials. Using this approach, we numerically demonstrate that in thick flakes the radiation due to the electron-beam deceleration is the dominating radiation mechanism. Our numerical findings agrees well with the experimental cathodoluminescence spectra. We further demonstrated that the captured CR in thick slabs could cause Fabry-Perot resonances that emerge as fine structures in the acquired CL spectra and demonstrate the fully coherent process of CR emission, happening due to its phase-matched excitation nature. Our findings pave the way for an accurate design of particle scintillators and detectors, based on the strong-coupling phenomenon.

O 60: Overview Talk Thorsten Deilmann

Time: Thursday 9:30–10:15 Location: S054

Invited Talk O 60.1 Thu 9:30 S054

Exciting states in atomically thin layers — •Thorsten Deilmann — Institute of Solid State Theory, University of Münster, Germany

Monolayers of two-dimensional materials and its stacking unite the fascinating characteristics of the confined in-plane physics with novel features due to the interlayer interaction. Especially in semiconducting materials largely tunable band gaps and optical responses have been observed in various materials. Unraveling the corresponding excited states is a crucial challenge for basic research as well as for possible applications in opto-electronic devices. Several external stimuli, e.g. doping, electric, or magnetic fields, can be applied to probe the response of many-particle states like excitons or trions, and thus also of its fundamental properties. Applying first-principles methods allows to predict, e.g., the quantum mechanical nature when an exciton is doped and becomes a trion, or its g factor due to the Zeeman effect. These calculations facilitate a connection of the experimental observations with the physical properties of the excited states. In the talk, several features in mono- and multilayer transition metal dichalcogenides and similar materials will be demonstrated.

O 61: Plasmonics and Nanooptics 2

Time: Thursday 10:30–12:45 Location: H2

Topical Talk O 61.1 Thu 10:30 H2

Single Molecule Nonlinearity in a Plasmonic Waveguide — •Markus Lippitz — Experimental Physics III, University of Bayreuth

Plasmonic waveguides offer the unique possibility to confine light far below the diffraction limit. Past room temperature experiments focused on efficient generation of single waveguide plasmons by a quantum emitter. However, only the simultaneous interaction of the emitter with multiple plasmonic fields would lead to functionality in a plasmonic circuit. Here, we demonstrate the nonlinear optical interaction of a single molecule and propagating plasmons. An individual terrylene diimide (TDI) molecule is placed in the nanogap between two single-crystalline silver nanowires. A visible wavelength pump pulse and a red-shifted depletion pulse travel along the waveguide, leading to stimulated emission depletion (STED) in the observed fluorescence. The efficiency increases by up to a factor of 50 compared to far-field excitation. Our study thus demonstrates remote nonlinear four-wave mixing at a single molecule with propagating plasmons. It paves the way toward functional quantum plasmonic circuits and improved nonlinear single-molecule spectroscopy.

O 61.2 Thu 11:00 H2

Excitation of coherent phonon modes in plasmonic gold nanoparticles — •Dominik Höing[1,2], Robert Salzwedel[4], Malte Selig[4], Kartik Ayyer[2,3], Jochen Küpper[2,3], Andreas Knorr[4], and Holger Lange[1,2] — [1]Institute of Physical Chemistry, Universität Hamburg, Hamburg, Germany — [2]The Hamburg Centre for Ultrafast Imaging, Hamburg, Germany — [3]Center for Free-Electron Laser Science, Hamburg, Germany — [4]Institut für Theoretische Physik, Technische Universität Berlin, Berlin, Germany

The plasmonic properties of gold nanoparticles (AuNP) have been widely studied, because of their exhibition of strong light-matter coupling and hot carrier generation. The carrier dynamics that follow the optical excitation are presently understood as a series of events with increasing time-scales. Among them, the excitation of coherent acoustic phonon modes, also called breathing modes, is thought to result from the ultrafast heating of the lattice due to electron-phonon coupling. However, previous experimental studies were insufficient in detecting the onset of the breathing oscillation. Here, we use a combination of transient absorption spectroscopy, time-resolved X-ray scattering and a model combining quantum coherent and hydrodynamic theory to develop a different picture: We show that the gradient in electron density induced by the optical excitation instantaneously couples to coherent phonons, resulting in an immediate onset of the breathing oscillation with the optical excitation. Thus, the processes involved in the plasmon decay are more intertwined as previously assumed.

O 61.3 Thu 11:15 H2

Probing a plasmon-polariton quantum wave packet — Sebastian Pres[1], Bernhard Huber[1], Daniel Fersch[1], Enno Schatz[2], Daniel Friedrich[2], Victor Lisinetskii[1], Ruben Pompe[3], •Matthias Hensen[1], Bert Hecht[2], Walter Pfeiffer[3], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]NanoOptics & Biophotonics Group, Experimental Physics 5, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [3]Fakultät für Physik, Universität Bielefeld, Universitätsstr. 25, 33615 Bielefeld, Germany

Beyond the classical picture, plasmon-polariton modes are treated as quasi-particles in quantum physics and they are considered essential for the realization of future nanoscale quantum functionality. Implementing and demonstrating such functionality requires local access to the quasi-particle's quantum state to monitor its corresponding quantum wave-packet dynamics. Here, we report the local probing of such an eigenstate superposition, linked to a nanoslit resonator, using plasmon-polariton-assisted electron emission as signal in coherent two-dimensional nanoscopy^1. We observe a quantum coherence oscillating at the third harmonic of the plasmon-polariton frequency and identify it, using quantum dynamical simulations, to arise from the superposition of energetically non-adjacent plasmon-polariton occupation number states. The simulations also contain an improved model for plasmon-polariton-assisted electron emission processes.

[1] M. Aeschlimann et al., *Science* 333, 1723-1726 (2011)

O 61.4 Thu 11:30 H2

Modelling Plasmon-Exciton Interaction using a Coupled-Oscillator Approach — •Simon Durst, Christoph Schnupfhagn, and Markus Lippitz — Experimental Physics III, University of Bayreuth

Coupling effects between plasmonic and excitonic systems have attracted increasing interest in the last decade. In particular, Surface Lattice Resonances, diffractive modes supported by periodic arrays of metallic nanoparticles, allow for long-range coherent energy transfer and have been shown to strongly couple to interspersed excitonic emitters. Here, we will present experimental results showing strong coupling between Surface Lattice Resonances and the excitonic mode of a J-aggregate dye. The angle-resolved extinction spectra of this coupled system are modelled using a coupled oscillator model, which describes observed effects, such as a loss of contrast of the upper polariton branch or an asymptotic behaviour of the lower polariton branch well.

O 61.5 Thu 11:45 H2

Light emission from single self-decoupled molecules — •Vibhuti Rai[1], Lukas Gerhard[1], Gabriel Derenbach[1], Nico Balzer[2], Michal Valášek[2], Marcel Mayor[2], and Wulf Wulfhekel[1] — [1]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology (KIT), D-76344 Eggenstein-Leopoldshafen, Germany — [2]Institute of Nanotechnology, Karlsruhe Institute of Technology (KIT), D-76344 Eggenstein-Leopoldshafen, Germany

Realization of single molecules as electric light sources faces a dilemma: In order to emit light, they need to be decoupled from the metallic electrodes to prevent fluorescence quenching. To conduct, however, the molecular orbitals need to hybridize with the electrodes. This can be achieved by decoupling the molecular chromophore to a certain extent from the electrode via anchoring groups. In the past, such self-decoupled molecules often lacked reproducibility of the decoupling and energy of the emitted light. Here, we show reproducible and well defined electroluminescence of two different NDI chromophores linked to a gold substrate via a molecular tripod and via a vacuum barrier to the tip of a scanning tunnelling microscope [1]. The stability of the system allows to perform systematic experiments so far only possible for molecules decoupled via insulating layers.

[1] Edelmann, K. et al. Rev. Sci. Instrum. 89, 123107 (2018).

O 61.6 Thu 12:00 H2

Investigation of chemical interface damping on electrochemically functionalized flat gold and nanoporous gold surfaces — •Maurice Pfeiffer[1], Xinyan Wu[1], Alexander Petrov[1,2], and Manfred Eich[1,2] — [1]Institute of Optical and Electronic Materials, Hamburg University of Technology, Germany — [2]Institute of Photoelectrochemistry, Helmholtz-Zentrum Hereon, Geesthacht, Germany

The chemical interface damping (CID) effect occurs at metal surfaces when applying surface modifications, such as oxidation. It results in an increase of the electron collision frequency of the metal, thus altering its optical properties. Previous research showed that this effect may also enhance the efficiency of photocatalytic water splitting by visible light [1]. We investigate the CID effect on flat gold as well as nanoporous gold samples with the latter having the advantage of a very high surface to volume ratio, making surface effects (like CID) more pronounced. Our experimental setup consists of a spectroscopic ellipsometer to investigate the optical properties. This device is equipped with an electrochemical cell in which we can reversibly oxidize the sample surface, allowing an in-situ study of the CID effect. Previous research investigated the CID effect on gold nanoparticles and observed an increase of the damping rate at the plasmon resonance [2]. Our results on flat gold reveal a similar increase of the damping, but additionally enable the investigation of broadband properties from which we expect further insights on the nature of the damping mechanism.

[1] Graf et al., ACS Nano 2021, 15, 2, 3188-3200 [2] Foerster et al., Nano Lett. 2020, 20, 5, 3338-3343

O 61.7 Thu 12:15 H2

Switching on the electroluminescence of single molecules adsorbed directly on a metal surface — Vibhuti Rai[1], •Lukas Gerhard[1], Nico Balzer[2], Michal Valášek[2], Christof Holzer[3], Liang Yang[2], Martin Wegener[2], Carsten Rockstuhl[3], Marcel Mayor[2], and Wulf Wulfhekel[1] — [1]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology (KIT), D-76344 Eggenstein-Leopoldshafen, Germany — [2]Institute of Nanotechnology, Karlsruhe Institute of Technology (KIT), D-76344 Eggenstein-Leopoldshafen, Germany — [3]Institute of Theoretical Solid State Physics, Karlsruhe Institute of Technology (KIT), D-76131 Karlsruhe, Germany

Exciting single molecules to emit light into the far-field requires them to be electronically decoupled from the contacting metallic leads. In the scanning tunnelling microscope (STM) geometry, typically this has been achieved by inserting insulating layers between the molecules and the metal substrate [1]. Here, we report our finding that it is possible to activate the electroluminescence (EL) of individual 2,6- core-substituted naphthalene diimide derivatives (Tpd-sNDI), adsorbed directly on a metal surface with the help of the STM tip. We observe that to emit light, both orbitals involved in the optical transition need to be electronically decoupled. The STM measurements were performed with a home-built, low temperature, ultra-high vacuum scanning tunnelling microscope (STM) with optical access [2].

[1] Zhang, Y. et al. Nature 531, 623 (2016).

[2] Edelmann, K. et al. Rev. Sci. Instrum. 89, 123107 (2018).

O 61.8 Thu 12:30 H2

First principles investigation of plasmonic hydrogen catalysis on metallic magnesium nanoparticles — •Oscar A. Douglas-Gallardo, Connor L. Box, and Reinhard J. Maurer — University of Warwick, Coventry, United Kingdom

Plasmon-induced hot-carrier photochemistry is currently a promising avenue to achieve highly selective and efficient chemical transformation on plasmonic metal surfaces. A new class of materials made up of Earth-abundant-elements has gained increasing interest in the plasmonic area, as an alternative to the oft studied late transition metals (Au, Ag and Cu).

Here, we will present our results on the optical and electronic properties of metallic magnesium nanoparticles and their potential use as plasmonic hydrogen catalysts. We explore the optical and electronic properties with time-dependent density functional tight-binding (TD-DFTB) and molecular dynamics with electronic friction (MDEF) simulations. Our results show that Mg nanoclusters can produce highly energetic hot-electrons and they energetically align with electronic states of physisorbed molecular hydrogen, the occupation of which by these hot electrons can promote hydrogen dissociation. The reverse reaction, hydrogen evolution on metallic Mg, may also be promoted by hot electrons, but following a different mechanism.

O 62: Surface Reactions and Heterogeneous Catalysis 1

Time: Thursday 10:30–12:45 Location: H4

O 62.1 Thu 10:30 H4

Gaussian Approximation Potentials for Surface Catalysis — •Sina Stocker[1,2], Gábor Csányi[3], Karsten Reuter[1], and Johannes T. Margraf[1] — [1]Fritz Haber Institut der Max Planck Gesellschaft, Berlin, Germany — [2]Technische Universität München, Germany — [3]University of Cambridge, United Kingdom

Predictive-quality first-principles based microkinetic models are increasingly used to analyze (and subsequently optimize) reaction mechanisms in heterogeneous catalysis. In full rigor, such models require the knowledge of all possible elementary reaction steps and their corresponding reaction barriers. Unfortunately, for complex catalytic processes (such as the generation of ethanol from syngas) the number of possible steps is so large that an exhaustive first-principles calculation of all barriers becomes prohibitively expensive.

To overcome this limitation, we develop machine learned (ML) interatomic potentials to model syngas conversion on Rhodium. These ML potentials can be used to determine free energy reaction barriers for a large number of adsorbates at various CO coverages and at a fraction of the computational cost of the underlying first-principles method. Specifically, we use here the Gaussian Approximation Potential (GAP) framework and explore iterative training in combination with umbrella sampling.

O 62.2 Thu 10:45 H4

Hydrogen adsorption on Pd surfaces and its effect on CO$_2$ activation — •Herzain I. Rivera-Arrieta[1], Igor Kowalec[2], Lucas Foppa[1], Andrew Logsdail[2], David Willock[2], and Matthias Scheffler[1] — [1]The NOMAD Laboratory at the Fritz Haber Institute of the Max Planck Society and the Humboldt-Universität zu Berlin, Germany — [2]Cardiff Catalysis Institute, Cardiff University, UK

Understanding the reactivity of Pd-based catalysts in hydrogenation processes requires an accurate description of its surfaces at realistic temperature (T) and hydrogen pressure (p_{H_2}). Herein, by modeling the surface phase diagram of Pd (111) and (100) as a function of T, p_{H_2} via *ab initio* atomistic thermodynamics [1], we predict 1 monolayer (ML) coverage of H as the most stable configuration under common experimental reaction conditions [2]. Taking a particular interest in the activation of CO$_2$, we performed simulations concerning the interaction of the molecule with the surfaces as the H-coverage increases from 0 to 1ML. Our results show how 1 ML of H inhibits the formation of chemisorbed $\mathrm{CO}_2^{\delta-}$, the initial intermediate in the CO$_2$ hydrogenation reaction. The generated data will be combined with information from other metal surfaces, and the subgroup-discovery artificial-intelligence approach [3] will be applied to identify which basic materials parameters correlate with indicators of CO$_2$ activation, e.g. the adsorption energy or the C-O bond elongation.

[1] C. Stampfl, *et al.*, *Appl. Phys. A*, **69**, 471 (1999).
[2] H. Bahruji, *et al.*, *J. Catal.*, **343**, 133 (2016).
[3] B. R. Goldsmith, *et al.*, *New J. Phys.*, **19**, 013031 (2017).

O 62.3 Thu 11:00 H4

Atomic steps as active sites in the Co-catalyzed Fischer-Tropsch synthesis: Evidence from an operando STM study on a stepped model catalyst — •Katharina Golder and Joost Wintterlin — Ludwig-Maximilians-Universität München, Germany

Atomic steps are often assumed to represent the active sites on the surface of a heterogeneous catalyst. This assumption is based on turnover data that display a certain scaling of the activity with the particle sizes of the catalysts. However, this evidence is indirect and often controversial. We have recently shown that the activity of a Co(0001) single crystal surface in the Fischer-Tropsch synthesis of hydrocarbons is proportional to the density of atomic steps. This is a quite direct evidence for the activity of atomic steps, but it was obtained for a flat model system. Here we present results of investigations on a Co(10$\bar{1}$5) surface that has a similar step density as the Co particles of the industrial Fischer-Tropsch catalyst. The experiments were performed with a scanning tunneling microscope (STM) at a syngas (2H$_2$+CO) pressure of $\sim$1 bar and a temperature of 503 K. Hydrocarbon products were detected by a special gas chromatograph (GC). The STM data show that the morphology of the stepped surface is stable under operando conditions. The activity measured by GC is considerably higher than that of the Co(0001) sample. It is close to the activities reported for supported Co catalysts, a result that bridges the materials gap between model systems and the industrial Fischer-Tropsch catalysts.

O 62.4 Thu 11:15 H4

Predicting Binding Motifs of Complex Adsorbates Using Machine Learning with a Physics-inspired Graph Representation — •Wenbin Xu[1], Karsten Reuter[1], and Mie Andersen[2] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]Aarhus Institute of Advanced Studies and Department of Physics and Astronomy, Aarhus University, Denmark

Complex adsorbates are involved in many surface catalytic reactions such as Fischer-Tropsch, methanol, or higher oxygenate synthesis. The modeling of these species at transition metal catalysts must account for their ability to exhibit a wide range of adsorption motifs, including mono- and multi-dentate adsorption modes. Given the combinatorial explosion of possible adsorption motifs and the computational cost of density functional theory, it is desirable to develop machine learning (ML) models for predicting the binding motifs and their associated adsorption enthalpies. Most ML models to date are only applicable to simple adsorbates. In this work, we overcome this limitation and propose a kernelized ML model with a physics-inspired graph representation for the prediction of complex species. The model is data-efficient and its good extrapolation ability makes it promising for comprehensively exploring complex reaction networks on novel catalysts. Furthermore, we show that the outliers with large prediction errors can be reliably captured from an ensemble uncertainty prediction approach.

O 62.5 Thu 11:30 H4

Machine-Learning Driven Global Optimization of Surface Adsorbate Geometries — •Hyunwook Jung, Sina Stocker, Karsten Reuter, and Johannes T. Margraf — Fritz-Haber-Institut der MPG, Berlin, Germany

The adsorption energy is an essential descriptor for predicting catalytic activity in theoretical models of heterogeneous catalysis. Although established scaling relations facilitate the prediction of adsorption energies for small adsorbates like OH, they are not applicable to larger adsorbates that are frequently encountered in syngas chemistry. Such systems often feature complex potential energy surfaces due to their flexibility and the possibility of multidentate binding to the surface. Consequently, computing adsorption energies for such adsorbates implies a complex global optimization to find the ground state geometry. This is prohibitively expensive at the density functional theory (DFT) level for routine applications. To tackle this issue, we present a global optimization protocol for adsorbate geometries which trains a surrogate Gaussian Approximation Potential on-the-fly. The approach is applicable to generic surface models (i.e. without defining surface sites) and minimizes both user intervention and the number of DFT calculations by iteratively updating the training set with configurations explored by the algorithm. We demonstrate this approach for diverse adsorbates on the Rh (111) and (211) surfaces.

O 62.6 Thu 11:45 H4

New catalysts for oxidative-coupling of methane: theoretical search and experimental validation — •Aliaksei Mazheika[1], Michael Geske[1], Matthias Mueller[2], Stephan Schunk[2], Frank Rosowski[1,3], and Ralph Kraehnert[1] — [1]Technische Universitaet, Berlin, DE — [2]hte GmbH, Heidelberg, DE — [3]BASF SE, Ludwigshafen, DE

Oxidative-coupling of methane (OCM) is a direct way for conversion of methane to higher hydrocarbons - ethane, ethylene. Despite many years spent for the search of an efficient catalyst, still a material which would satisfy industrial rentability has not been found. In our study we proceed from an experimentally observed volcano-like dependence of C$_2$-yields on formation energies of carbonates on oxide catalysts [1]. We developed a new method which allows to calculate carbonates formation energies from CO$_2$ adsorption energies on the surfaces of corresponding oxides. In combination with artificial intelligence methods (data mining and symbolic regression) this was used in the high-throughput screening. The latter is done in a way of active-learning, and we demonstrate its advantages compared to traditional scheme. Several catalysts obtained from the screening have been synthesized

and experimentally tested together with less promising materials. Obtained C_2-yields follow the same volcano-type dependence that formed initial basis for the employed strategy. The best catalyst candidates reach maximum C_2-yields comparable to the well-known OCM catalysts and outperform them at lower temperatures.
[1] H. Wang *et al.*, https://doi.org/10.26434/chemrxiv-2022-gxt5n

O 62.7 Thu 12:00 H4

Finding catalyst *genes* with subgroup discovery — •Aliaksei Mazheika[1], Yanggang Wang[2], Rosendo Valero[3], Francesc Vines[3], Francesc Illas[3], Luca M. Ghiringhelli[4], Sergey V. Levchenko[5], and Matthias Scheffler[4] — [1]Technische Universitaet, Berlin, DE — [2]University of Science and Technology, Shenzhen, CN — [3]Universitat de Barcelona, Barcelona, ES — [4]The NOMAD Laboratory at the Fritz Haber Institute and Humboldt University, Berlin, DE — [5]Moscow

Catalytic-materials design requires predictive modeling of the interaction between catalyst and reactants. This is challenging due to the complexity and diversity of structure-property relationships across the chemical space. Here, we report a strategy for a rational design of catalytic materials using the artificial intelligence approach (AI) subgroup discovery. We identify catalyst *genes* (features) that correlate with mechanisms that trigger, facilitate, or hinder the activation of carbon dioxide (CO_2) towards a chemical conversion. The AI model is trained on first-principles data for a broad family of oxides. We demonstrate that surfaces of experimentally identified good catalysts consistently exhibit combinations of *genes* resulting in a strong elongation of a C-O bond. The same combinations of *genes* also minimize the OCO-angle, the previously proposed indicator of activation, albeit under the constraint that the Sabatier principle is satisfied. Based on these findings, we propose a set of new promising catalyst materials for CO_2 conversion.—A. Mazheika *et. al.* Nature Comm. 2022, 13, 419.

O 62.8 Thu 12:15 H4

Selectivity in single-molecule reactions by tip-induced redox chemistry — •Florian Albrecht[1], Shadi Fatayer[1,2], Iago Pozo[3], Ivano Tavernelli[1], Jascha Repp[4], Diego Pena[3], and Leo Gross[1] — [1]IBM Research - Zurich, 8803 Rüschlikon (Switzerland) — [2]Physical Science and Engineering Division, King Abdullah University of Science and Technology (KAUST), 23955-6900 Thuwal (Saudi Arabia) — [3]Centro Singular de Investigación en Química Biolóxica e MateriaisMoleculares (CiQUS) and Departamento de Química Orgμnica, Universidade de Santiago de Compostela, 15782 Santiago de Compostela (Spain) — [4]Institute of Experimental and Applied Physics, University of Regensburg, 93053 Regensburg (Germany)

Since the first experiments by S.-W. Hla in 2000 [1], tip-induced on-surface synthesis is an active field. Not only in on-surface chemistry, selective control over the outcome of a reaction is a major quest. Here, we activate a molecule adsorbed on ultrathin insulating films by dehalogenation and perform selective constitutional isomerization reactions in a low temperature UHV combined STM and AFM. The selectivity is controlled by the polarity and amplitude of applied voltage pulses. The insulating films stabilize the isomers in different charge states and allow for their characterization. The importance of molecular charge state on the reaction is supported by DFT-derived isomerization energy landscape. [1] S.-W. Hla et al., Phys. Rev. Lett. 85, 2777 (2000).

O 62.9 Thu 12:30 H4

Abiotic Formation of an Amide Bond via Surface-Supported Direct Carboxyl-Amine Coupling — •Biao Yang[1,2], Kaifeng Niu[1,3], Felix Haag[2], Nan Cao[1,2], Junjie Zhang[1], Haiming Zhang[1], Qing Li[1], Francesco Allegretti[2], Jonas Björk[3], Johannes Barth[2], and Lifeng Chi[1] — [1]Institute of Functional Nano and Soft Materials (FUNSOM), Soochow University, Suzhou 215123 (P. R. China) — [2]Physics Department E20, Technical University of Munich, D-85748 Garching, (Germany) — [3]Department of Physics, Chemistry and Biology, IFM, Linkçping University, 58183 Linköping (Sweden)

Amide bond formation is one of the most important reactions in biochemistry, notably being of crucial importance for the origin of life. Herein, we combine scanning tunneling microscopy and X-ray photoelectron spectroscopy studies to provide evidence for thermally activated abiotic formation of amide bonds between adsorbed precursors through direct carboxyl-amine coupling under ultrahigh-vacuum conditions by means of on-surface synthesis. Complementary insights from temperature-programmed desorption measurements and density functional theory calculations reveal the competition between cross-coupling amide formation and decarboxylation reactions on the Au(111) surface. Furthermore, we demonstrate the critical influence of the employed metal support: whereas on Au(111) the coupling readily occurs, different reaction scenarios prevail on Ag(111) and Cu(111).[1] [1] Biao Yang+*, Kaifeng Niu+, et al. Angew. Chem. Int. Ed. anie.202113590 (2021)

O 63: New Methods and Developments 4: Spectroscopies, Diffraction and Others (joint session O/KFM)

Time: Thursday 10:30–12:30 Location: H6

Topical Talk O 63.1 Thu 10:30 H6

Element and Structure Analysis of Surfaces Using Positrons — •Christoph Hugenschmidt — Forschungs-Neutronenquelle Heinz Maier-Leibnitz (MLZ), Technische Universität München, Lichtenbergstr. 1, 85748 Garching, Germany

With the advent of bright low-energy positron beams novel analysis tools have been developed exploiting the unique properties of positron matter interaction such as repulsive crystal potential or positron trapping in surface states [1]. Positron annihilation is established for defect spectroscopy and the characterization of the free volume in amorphous matter. By applying a slow positron beam, however, defects near the surface can be specifically addressed, e.g. for the determination of the oxygen vacancy concentration in $YBa_2Cu_3O_{7-x}$ [2]. The positron counterparts of reflection high-energy electron diffraction (RHEED) and electron induced Auger-electron spectroscopy (AES) intrinsically exhibit superior surface sensitivity. In contrast to electrons, positrons show total reflection for small glancing angles. It was demonstrated that with reflection high-energy positron diffraction the structure of the topmost and the immediate subsurface atomic layer of surfaces are revealed with outstanding accuracy. The main advantages of positron annihilation induced AES are the missing secondary electron background and its topmost layer sensitivity for element analysis allowing, e.g. the *in-situ* observation of the Ni adatom migration from the Pd surface into the bulk [3]. [1] C. H.; Surf. Sci. Reports 71 (2016) 547; [2] M. Reiner et. al.; Phys. Rev. B 97 (2018) 144503; [3] S. Zimnik et. al.; Surf. Sci. 664 (2017) 61

O 63.2 Thu 11:00 H6

Active sample manipulation with electrostatic beams: a different way of bridging the high-voltage gap — •Francesco Guatieri, Kilian Brenner, and Christoph Hugenschmidt — Heinz Maier-Leibnitz Zentrum (MLZ), Technical University of Munich, Lichtenbergstr. 1, 85748 Garching, Germany

Most electrostatic positron beams used to perform surface studies are accelerated to the desired implantation energy by floating the target to a high electrostatic potential relative to ground. The presence of tens on kilovolts of electric potential makes the use of instrumentation attached directly to the sample inconvenient. The conventional solution to this hurdle consists in wiring insulated connections from the sample to a high-voltage galvanic decoupling placed outside of the experimental chamber far away from the sample holder. This solution carries limitations on the kind and amount of electrical connections employed. We will present, instead, a novel approach to in-operando sample manipulation that we have developed to be used at the Doppler broadening spectrometer installed on the NEPOMUC positron source, which implements the galvanic insulation in situ and removes many of the limitations imposed by conventional solutions.

O 63.3 Thu 11:15 H6

Advanced Kernel-Based NMR Cryoporometry Characterization of Mesoporous Solids — •Henry R.N.B. Enninful, Daniel Schneider, Richard Kohns, Dirk Enke, and Rustem Valiullin — Leipzig University, Leipzig, Germany

NMR cryoporometry is a pore space characterization technique for industrial and natural materials such as catalysts, gas storage materials, cartilage, bones, rocks and many more. While gaining wide use, the fundamental phenomena underlying solid-liquid phase transitions in geometrically disordered porous materials is still not fully understood. This may lead to inaccurate pore size distributions from the NMR cryoporometry technique.

In this work, we have developed a new approach to NMR cryoporometry. Herein, it takes account of cooperativity effects in pores, the existence of a variable non-frozen layer (NFL) thickness between the frozen core and pore wall and the effect of curvature on thermal fluctuations in pores which hitherto are missing in the current approach. In the first place, we compile a family of transition curves characterizing the phase state in pores with different pore sizes, so called kernels. Thereafter, we apply a general framework for predicting phase equilibria in a collection of pores. Specifically, the proposed kernel-based approach is coupled with the serially-connected pore model (SCPM) to be able to predict phase behavior in independent pore systems as well as in pore networks. We demonstrate the new approach by applying it to ordered porous materials such as MCM-41 and SBA-15. Consequently, a more accurate pore size distribution (PSD) is obtained.

O 63.4 Thu 11:30 H6

Development of an electron spin resonance spectrometer in ultra-high vacuum for surface spins — •Juyoung Park[1,2], Franklin H. Cho[1,2], Jisoo Yu[1,2], Luciano Colazzo[1,2], Yejin Jeong[1,2], Junjie Liu[3], Arzhang Ardavan[3], Giovanni Boero[4], Andreas Heinrich[1,2], and Fabio Donati[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Republic of Korea — [2]Ewha Womans University, Seoul, Republic of Korea — [3]The Clarendon Laboratory, Department of Physics, University of Oxford, Oxford, UK — [4]Ecole Polytechnique Fédérale de Lausanne, Laboratory for Microsystems, Lausanne, Switzerland

We present the development of an electron spin resonance (ESR) spectrometer operating in ultra-high vacuum (UHV) for studying surface-adsorbed molecular and atomic spin systems. Such surface spin systems are promising platforms for potential applications in quantum computing and information processing [Science 366, 509 (2019)]. Our spectrometer is capable of both continuous-wave and pulsed ESR measurement in the temperature range of 2.5 K to 300 K. The surface-sensitivity is attained using a surface-type microwave resonator with its resonance frequency in the X-band. The spectrometer is connected to a home-built in-situ preparation chamber which allows us to prepare and characterize surfaces with low-energy electron diffraction and Auger electron spectroscopy. We demonstrate that we are sensitive down to a monolayer of molecular film using organic radicals such as α,γ-Bisdiphenylene-β-phenylallyl, and metal phthalocyanine complexes such as vanadyl phthalocyanine.

O 63.5 Thu 11:45 H6

Unsupervised machine learning-assisted analysis of multidimensional ARPES data — •Steinn Ymir Agustsson[1], Mohammad Ahsanul Haque[2], Fatemeh Zardbani[2], Davide Mottin[2], Panagiotis Karras[2], and Philip Hofmann[1] — [1]Institute of Physics and Astronomy, Aarhus University, Denmark — [2]Institute of Computer Science, Aarhus University, Denmark

In recent years, the size and complexity of experimental data sets has been dramatically growing in many fields of science. For photoemission spectroscopy, the development of novel detectors and multi-dimensional measurement modes (e.g., including a time dependence or spatial dependence), has lead to orders of magnitude more data being produced. Even after a necessary upgrade of the data management system, it remains highly challenging to visualize and superficially interpret the data fast enough to feed back into decisions about what to measure in an ongoing experimental run. A promising approach to address this is the application of machine learning tools. These have shown promising results when applied to data reduction and feature detection tasks in many fields of science. We have developed an unsupervised clustering method which is able to distinguish differences between ARPES spectra obtained from different spatial locations in nanoARPES measurements. This enables quick and automatic identification and classification of regions with different spectral features, allowing to invest more time in the collection of significant data.

O 63.6 Thu 12:00 H6

ViPErLEED: A modern all-in-one LEED I(V) package — •Alexander M. Imre[1], Florian Kraushofer[1,2], Florian Doerr[1], Tilman Kisslinger[3], Michael Schmid[1], Ulrike Diebold[1], Lutz Hammer[3], and Michele Riva[1] — [1]TU Wien, Vienna, Austria — [2]TU Munich, Munich, Germany — [3]FAU Erlangen-Nürnberg, Erlangen, Germany

Many surface science groups use Low-Energy Electron Diffraction (LEED) for quick, qualitative analysis of surface periodicity. Analysis of the beam intensities as a function of electron energy [LEED $I(V)$] is sensitive to surface atom positions at the picometer scale. Thus, comparison with calculated intensities can verify or reject structural models. Despite this, LEED $I(V)$ is currently rather unpopular, largely because the available software solutions are not sufficiently user-friendly. To greatly lower the barrier of entry into the field, we present the Vienna Package for TensErLEED (ViPErLEED) which provides a truly all-in-one package for LEED $I(V)$. ViPErLEED includes a freely available design for electronics that enable upgrading existing LEED setups for LEED $I(V)$ use. With sophisticated image acquisition and processing methods, as well as an automated spot-tracking tool for curve extraction, we greatly simplify the most tedious parts of the experiment. For the calculation of intensities, the package includes a user-friendly front-end and an extensive overhaul to the established TensErLEED package that only requires a few standardized input files. We further describe automated symmetry detection, improvements to the structure search algorithm, and a Python API.

O 63.7 Thu 12:15 H6

On-surface GNR fabrication via electrospray deposition of monomers and polymers from solution — •Felix Baier[1], Christoph Dobner[1], Michael Beckstein[1], Mamun Sarker[2], Alexander Sinitskii[2], and Axel Enders[1] — [1]Universität Bayreuth — [2]University of Nebraska - Lincoln, USA

Strategies for depositing large organic molecules such as proteins, DNA or graphene nanoribbons (GNRs) are urgently needed because the conventional method of evaporation is impossible due to the size of the molecules. GNRs prepared in solution are of particular interest because they are longer compared to those synthesized on the surface and can be produced in large quantities. Since GNRs form crystallites, they cannot be brought onto the surface by direct contact printing and characterized using STM. Therefore, a new electrospray setup was developed for the deposition of GNR precursor molecules, large precursor polymers, and GNR from a solution. The instrument consists of a heatable stainless steel capillary to which a high voltage in the range -8 to 8 kV, with respect to the sample can be applied. The assembly is placed in a glovebox which ensures the cleanliness of the working process. The characterization of the deposits was done with STM under ultra high vacuum after sample transfer. The deposition of TPTP monomers from solution onto Au(111) brought comparable results as other, established approaches were cGNR were formed after direct contact printing in UHV. Larger polymers were also deposited and completely cyclized on the surface after deposition, forming promising GNRs that have not been studied anywhere before.

O 64: Gerhard Ertl Young Investigator Award

Time: Thursday 10:30–13:00 Location: S051

O 64.1 Thu 10:30 S051

Gold-colored metallic water solution — •Tillmann Buttersack[1], Philip Mason[2], Christian Schewe[2], Ryan McMullen[3], Florian Trinter[1,4], Daniel Neumark[5], Stephan Thürmer[6], Robert Seidel[7], Bernd Winter[1], Stephen Bradforth[3], and Pavel Jungwirth[2] — [1]Fritz-Haber-Institute, Berlin, GER — [2]Czech Academy of Sciences, Prague, CZ — [3]U Southern California, Los Angeles, USA — [4]DESY, Hamburg, GER — [5]UC Berkeley, USA — [6]Kyoto U, JP — [7]BESSY, Berlin, GER

Whereas tap water and common aqueous solutions conduct some electricity, pure water is an almost perfect insulator. The required pressures to transform water into a metallic state will not be achievable on Earth. Liquid Ammonia undergoes a gradual phase transition from a blue electrolyte solution to a bronze-colored metallic solution if the concentration of solvated alkali metal is enhanced. Water and alkali metals react vigorous, as the valence electrons of the alkali metal are transferred to the aqueous solution almost instantly, leading to a Coulomb explosion. Here, we demonstrate, that water from the gas phase adsorbs onto the surface of sodium-potassium alloy droplet leading to the formation of a thin shiny and golden layer. Characterization of the liquid with photoelectron and UV/Vis spectroscopy revealed a plasmon energy of 2.7 eV and signatures of liquid water. These observations are spectroscopic evidence for an aqueous solution with metallic properties. Its plasmon frequency is in the visible region of the spectrum and thus is the reason for the fascinating golden color.

O 64.2 Thu 11:00 S051

Standing molecules for quantum sensing — •Taner Esat — Peter Grünberg Institute (PGI 3), Forschungszentrum Jülich, 52425 Jülich, Germany — Jülich Aachen Research Alliance (JARA), Fundamentals of Future Information Technology, 52425 Jülich, Germany

Artificial nanostructures, fabricated by placing building blocks like atoms and molecules in well-defined positions, are an almost universal playground where quantum effects can be studied and exploited on the atomic scale. In my talk, I will show that the manipulation capabilities of a scanning tunneling microscope (STM) allow the fabrication of metastable structures which do not form spontaneously in nature and that these structures offer attractive functionalities for quantum information and sensing. Specifically, I will demonstrate that a large planar aromatic molecule can be lifted into an upright standing geometry on a pedestal of two metal adatoms using the STM tip. This atypical and surprisingly stable upright orientation of the single molecule enables the system to function as a quantum dot and a coherent single-electron field emitter. Utilizing a novel homebuilt STM that uses adiabatic demagnetization refrigeration (ADR) to reach millikelvin temperatures, I will show that the standing molecule is weakly coupled to the surface and that it exhibits spin-flip excitations corresponding to a spin-1/2 system. If the standing molecule is fabricated on the tip, it can be used for the measurement of surface potentials and magnetic fields on the atomic scale at the same time - possibly even as an electron spin resonance sensor on the tip.

O 64.3 Thu 11:30 S051

On-surface synthesis and characterization of cyclo[18]carbon — •Katharina Kaiser[1], Lorel Scriven[2], Fabian Schulz[1], Przemyslaw Gawel[2], Leo Gross[1], and Harry Anderson[2] — [1]IBM Research - Zurich, Säumerstrasse 4, 8803 Rüschlikon, CH — [2]Department of Chemistry, Oxford University, Chemistry Research Laboratory, Oxford, OX13TA, UK

Cyclo[n]carbons, purely *sp*-hybridized carbon allotropes with unique structural and electronic properties, have been predicted for decades to exist. Although a synthetic route was already elaborated in the 1980's, and glimpses of cyclocarbons were detected in gas phase, they could never be stabilized long enough for characterization. A distinctive feature of cyclo[n]carbons is their two conjugated π-systems that can lead to the formation of two orthogonal ring currents and so-called double-aromatic stabilization. However, distortions in the geometry can lead to a lowering in ground state energy, and theoretically predicted ground state structures were found to depend on the level of theory. Consequently, cyclo[n]carbon's structures remained unknown.

AFM and STM at low temperatures allow triggering certain on-surface chemical reactions by atom manipulation and can thus facilitate the controlled formation of highly reactive molecules on inert surfaces from more stable precursors. Using this approach, we formed cyclo[18]carbon on a thin layer of NaCl. By comparing high-resolution AFM images with a functionalized tip and AFM simulations of different predicted resonance structures, we identified two possible ground state structures of cyclo[18]carbon adsorbed on bilayer NaCl.

O 64.4 Thu 12:00 S051

Correlative in situ microscopy of hydrogen oxidation on rhodium: from the meso- to the nanoscale — •Johannes Zeininger — Institute of Materials Chemistry, TU Wien, Getreidemarkt 9, 1060 Vienna, Austria

Catalytic surface reactions, such as CO oxidation, may produce a variety of spatio-temporal effects on the catalyst surface. To reveal the mechanisms behind such spatio-temporal effects, it is advantageous to apply multiple techniques to the same samples, particularly at different length scales. In other research areas than catalysis this idea has led to the correlative microscopy approach. In catalysis, this approach is, if at all, used for sample characterization, often in two separate setups. In the present contribution, the correlative microscopy approach has been applied in situ to the catalytic hydrogen oxidation on rhodium to reveal the mechanism of multifrequential kinetic oscillations, a novel effect recently detected in this reaction. Such oscillations were first observed on a polycrystalline Rh foil, where the reaction oscillated in a self-sustained way between the states of high and low activity (a known behavior), but with different frequencies on adjacent domains (a new and unexpected behavior). Later on, such an effect was also observed on the nanofacets of a Rh tip modeling a single catalytic particle. Such behavior contradicts previous observations and expectations based on the known spatial coupling mechanisms and was never observed before, for any surface reaction.

O 64.5 Thu 12:30 S051

Efficient electronic passivation schemes for surface calculations of semiconductors exhibiting spontaneous polarization: Thermodynamic and electronic properties of GaN surfaces — •Su-Hyun Yoo[1], Mira Todorova[1], Liverios Lymperakis[1], Chris Van de Walle[2], and Jörg Neugebauer[1] — [1]Department of Computational Materials Design, MPI für Eisenforschung, Germany — [2]Computational Materials Group, Materials Department, UCSB, USA

Semiconductor surfaces play a central role in modern technology related to catalysis, electronics, and energy applications. The most widespread approach to study surfaces with density-functional theory calculations is to use slab geometries with periodic boundary conditions. A common strategy employed to avoid artificial charge transfer from one side of the slab to another, is to passivate the dangling bonds at its backside. Using the examples of wurtzite polar and semipolar surfaces, we demonstrate that the conventional passivation scheme using pseudo-H atoms fails to describe the electronic structure of low-symmetry semiconductors. We therefore developed an improved passivation method [npj Comp. Mater. 7, 58, 2021/PR Mater. 5, 044605, 2021] that takes the polarization effect and the concept of surface reconstructions into account. It accurately describes surface electronic properties and enables computationally efficient surface energy calculations. Using this novel approach we have studied the orientation-dependent thermodynamic stability and electronic properties of GaN surfaces. The resulting Wulff shape provides insight of how to avoid facet-related defects such as V-pits hampering GaN-based electronics.

O 65: 2D Materials 2: Growth, Structure and Substrate Interaction

Time: Thursday 10:30–12:30 Location: S052

O 65.1 Thu 10:30 S052

Segregation-enhanced epitaxy of borophene on Ir(111) by thermal decomposition of borazine — •Karim Omambac[1], Marko Kriegel[1], Marin Petrovic[1,2], Pantelis Bampoulis[4], Christian Brand[1], Pascal Dreher[1], David Janoschka[1], Ulrich Hagemann[3], Nils Hartmann[3], Philipp Valerius[4], Thomas Michely[4], Frank-J. Meyer zu Heringdorf[1,3], and Michael horn-von Hoegen[1] — [1]Universität Duisburg-Essen, Germany — [2]Institute of Physics, 10000 Zagreb, Croatia — [3]Interdisciplinary Center for the Analytics on the Nanoscale, Germany — [4]Universität zu Köln, Germany

While borophene is typically prepared by molecular beam epitaxy [1], we report here on an alternative way of synthesizing large single-phase borophene domains by segregation enhanced epitaxy. X-ray photoelectron spectroscopy shows that borazine dosing at 1100°C onto Ir(111) yields a boron-rich surface without traces of nitrogen. At high temperatures the borazine thermally decomposes, nitrogen desorbs, and boron diffuses into the substrate. Using time-of-flight secondary ion mass spectroscopy we show that during cooldown the sub-surface boron segregates back to the surface where it forms borophene. In this case electron diffraction reveals a (6x2) reconstructed borophene χ6-polymorph [1], and scanning tunneling spectroscopy suggests a Dirac-like behavior. Studying the kinetics of borophene formation in low energy electron microscopy shows elongated and extended borophene domains with exceptional structural order. [1] ACS Nano 13 (12), 14511-14518 (2019).

O 65.2 Thu 10:45 S052

Investigation of sub-monolayer Sn phases on Au(111) — •Julian Andreas Hochhaus[1,2], Lukas Kesper[1,2], Stefanie Hilgers[1], Ulf Berges[1,2], and Carsten Westphal[1,2] — [1]Technische Universität Dortmund, Fakultät Physik, Otto-Hahn-Str. 4, D-44227, Dortmund, Germany — [2]DELTA, TU Dortmund, Maria-Goeppert-Mayer-Str. 2, D-44227, Dortmund, Germany

In this study we investigate the temperature and layer thickness dependence of sub-monolayer phases of Sn on Au(111).

Stanene, the two-dimensional graphene analog of tin, is predicted to exhibit similar exceptional electronic properties. Since tin is one of the heavier elements in the carbon group, stanene exhibits topological properties even at RT due to its strong spin-orbit coupling. Therefore, stanene is a promising material for application in future two-dimensional topological devices.

Since the surface and interface structure of low-dimensional materials often influences their electronic properties, we focus on the structural and chemical analysis of the Sn/Au interface.

Here, we report a structural and chemical investigation of different sub-monolayer Sn phases on Au(111) by means of low energy electron diffraction (LEED) and photoelectron spectroscopy (XPS). Tin layers of different thicknesses were deposited gradually on the Au(111) surface by physical vapor deposition and subsequently heated. A strong dependence of the structural and chemical configuration of the tin atoms on the layer thickness and the post-deposition annealing temperature was observed.

O 65.3 Thu 11:00 S052

On the transition from MoS_2 single-layer to bilayer growth on the Au(111) surface — •Moritz Ewert[1], Lars Buss[1], Francesca Genuzio[3], Tevfik Onur Menteş[3], Andrea Locatelli[3], Jens Falta[2], and Jan Ingo Flege[1] — [1]Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus-Senftenberg, Germany — [2]Institute of Solid State Physics, University of Bremen, Germany — [3]Elettra-Sincrotrone Trieste S.C.p.A., Basovizza, Trieste, Italy

MoS_2 is well known for changing from an indirect to a direct band-gap semiconductor as a single layer. Here, for the model system MoS_2/Au(111), we present in-situ studies of the continued growth of micron-size single-layer MoS_2 islands including the first formation of bilayer patches.

We have used angle-resolved photoemission spectroscopy from micrometer sized regions to investigate the local band structure of the islands' rims and centers, showing a prevalence for bilayer and single-layer formation at the rims and centers, respectively. The bilayer patches can clearly be identified locally on the few nanometer scale employing intensity-voltage low-energy electron microscopy as a fingerprinting method. Astonishingly, micro-spot low-energy electron diffraction hints toward the nucleation of the second layer of the MoS_2 between the single layer MoS_2 and the Au(111) substrate when the step bunches formed by the single-terrace growth mechanism become sufficiently high.

O 65.4 Thu 11:15 S052

Structural and electronic investigations of CVD-grown TMDs on Au(111) — •Julian Picker[1], Maximilian Schaal[2], Ziyang Gan[1], Marco Gruenewald[2], Christof Neumann[1], Antony George[1], Felix Otto[2], Torsten Fritz[2], and Andrey Turchanin[1] — [1]Institute of Physical Chemistry, Friedrich Schiller University Jena, Lessingstraße 10, 07743 Jena, Germany — [2]Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany

Transition metal dichalcogenides (TMDs) are layered two-dimensional (2D) materials which have come into the focus of research in recent years. Especially, the exciting change of properties when going from bulk to monolayer make them interesting for novel electronic applications. Here, we demonstrate the *ex-situ* growth of MoS_2 and $MoSe_2$ monolayers on Au(111) by chemical vapor deposition (CVD) in a two-zone furnace. Afterwards, the samples were analyzed in ultra-high vacuum (UHV) with the help of surface sensitive methods, including (low-temperature) scanning tunneling microscopy and spectroscopy (STM/STS), low-energy electron diffraction (LEED) as well as X-ray and angle-resolved ultra-violet photoelectron spectroscopy (XPS/ARPES). While we could confirm the growth of high-quality single crystalline TMDs, we also accessed the structural and electronic properties of these samples down to the atomic scale. Finally, the impact of annealing on the structure and properties of the TMDs was studied.

O 65.5 Thu 11:30 S052

Spectroscopic and microscopic study of carborane based 2D materials — •Martha Frey[1], Julian Picker[1], Jakub Višňák[2], Christof Neumann[1], Tomáš Baše[2], and Andrey Turchanin[1] — [1]Friedrich Schiller University Jena, Institute of Physical Chemistry, Lessingstraße 10, 07743 Jena, Germany — [2]The Czech Academy of Sciences, Institute of Inorganic Chemistry, 250 68 Husinec-Rez, c.p. 1001, Czech Republic

Carboranes are electron-delocalized molecular clusters containing boron, carbon and hydrogen. Because of their high stability and structural variability, they are interesting for a wide range of applications including nanoscale engineering, catalysis and boron neutron capture therapy. Here, we present the fabrication of a novel boron-based two-dimensional (2D) material *via* electron irradiation induced cross-linking of carborane self-assembled monolayers (SAMs) on silver or gold substrates. The SAMs, the cross-linking process and the resulting 2D nanosheets have been characterized with different complementary surface sensitive techniques including X-ray photoelectron spectroscopy (XPS), low-energy electron diffraction (LEED), scanning tunneling microscopy (STM) and scanning electron microscopy (SEM).

O 65.6 Thu 11:45 S052

Growth and Structural Properties of 2D Blue Phosphorene on Au(111) and on Au(100) — •Maximilian Schaal[1], Julian Picker[2], Felix Otto[1], Marco Gruenewald[1], Roman Forker[1], and Torsten Fritz[1] — [1]Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany — [2]Institute of Physical Chemistry, Friedrich Schiller University Jena, Lessingstraße 10, 07743 Jena, Germany

Blue phosphorene (BlueP) is a novel two-dimensional material that shares properties with black phosphorene and is potentially even more interesting for optoelectronic applications because of its layer dependent band gap of approx. 2 to 3 eV and superior charge carrier mobility [1]. Despite these promising properties, the growth of quasi-freestanding BlueP is still challenging.

In our contribution we will report on the structural and electronic properties of epitaxial BlueP on Au(111) and on Au(100) by means of scanning tunneling [hydrogen] microscopy (ST[H]M), distortion-corrected

low-energy electron diffraction (LEED) as well as X-ray photoelectron spectroscopy and diffraction (XPS and XPD). It is already known that on Au(111) a network consisting of BlueP islands, which are connected by Au linker atoms, is formed [2]. In contrast, we demonstrate that on Au(100) quasi-free standing BlueP domains grow [3].
[1] Z. Zhu and D. Tománek, Phys. Rev. Lett. 112, 176802 (2014).
[2] H. Tian et al., Matter 2, 111 (2020).
[3] M. Schaal, J. Phys.: Condens. Matter 33, 485002 (2021).

O 65.7 Thu 12:00 S052

How the supple Pt(110) surface paves the way to single-domain h-BN growth. — •Marco Thaler[1], Dominik Steiner[1], Florian Mittendorfer[2], and Erminald Bertel[1] — [1]Department of Physical Chemistry, University of Innsbruck, Austria — [2]Institute of Applied Physics and Center for Computational Materials Science, Vienna University of Technology, Austria

Its structural flexibility renders Pt(110) an extraordinary substrate for hexagonal boron nitride (h-BN) growth. Borazine-exposure at high temperature yields either an extended film or empty terraces depending on dosage. Annealing preadsorbed Borazine, in contrast, yields nuclei with a structure differing from the extended film. DFT modelling of the latter was carried out by starting with a stretched film, which ruptured upon relaxation into smaller h-BN islands. The Pt surface layer below them is severely disordered. Only after high-T annealing the islands relaxe into the final h-BN/Pt(110)-(1xn) missing-row configuration. Thus, this non-classical, two-step nucleation proceeds via a metastable transition structure. Under the conditions of high-T exposure the metastable nuclei are continuously dissolved by desorption and H-induced etching. Stochastic formation of a critical island size with the more stable (1xn)m.r. structure requires a high supersaturation, but once formed, such an island grows explosively into an extended film. Hence, single-domain growth of h-BN on Pt(110) differs significantly from that on vicinal Cu(110), where (1-12) step edges act as nucleation sites giving rise to uniform nuclei capable of seamless coalescence.

O 65.8 Thu 12:15 S052

Development of strain- and gate-controllable STM sample holder — •Jz-Yuan Juo[1], Bong Gyu Shin[1], Soon Jung Jung[1], and Klaus Kern[1,2] — [1]Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, DE-70569 Stuttgart, Germany — [2]Institut de Physique, École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

An atomic-scale understanding of strain effects on electronic properties is essential for implementing two-dimensional materials into flexible electronics. The scanning tunneling microscopy (STM) is an ideal method to advance this understanding. However, the combination of STM and strain-controllable devices remains challenging due to the high demand for compatibilities with limited STM space, ultrahigh vacuum, and mechanical stability. We have developed an indentation-based sample holder for STM measurements. A gearbox and a piezo stack were used to control the distance between the indentor and 2D materials transferred on the suspended polyimide. The gearbox has a travel range of $\sim$120 μm and precision of $\sim$1.4 μm; the piezo stack has a travel range of $\sim$1.8 μm and precision of <1 nm. The combination of gearbox and piezo stack allowed us to tune the distance between indentor and sample continuously with nanometer precision, characterized by atomic force microscopy. Raman spectroscopy was used to measure strain distribution in monolayer MoS_2 at the indented area. The strain controllability is characterized by calculating graphene lattice constant changes in STM atomic-resolution images. Spectroscopic studies confirm the gate tunability by observing Dirac point shifts.

O 66: Oxide Surfaces 2

Time: Thursday 10:30–13:00 Location: S053

Topical Talk O 66.1 Thu 10:30 S053

Charge-ordered states on incipient ferroelectric polar surfaces — •Cesare Franchini — University of Vienna — University of Bologna

The precise atomic-scale knowledge of surface properties is mandatory for interpreting experimental data, optimizing (photo)catalytic processes, or predicting novel electronic phases. This is particularly challenging for perovskite materials, which typically show a variety of surface structural reconstructions that prevent a generally valid and transferable understanding of surface properties. However, recent experiments have shown that a novel cleavage protocol based on controlled exploitation of ferroelectric instabilities can generate well-defined bulk-terminated (001) surfaces, paving the way for a precise characterization of the surface electronic structure. This talk shall report first principles electronic structure calculations on polar KTaO3(001), focusing on the dichotomy between charge localized/delocalized phases and their impact on surface reactivity. Our data, supported by experimental observations, show that polarity-induced uncompensated carriers in KTaO3(001) exhibit charged-order patterns rather than common 2DEGs, manifested by charge-density wave and (bi)polaron states. Eventually, the trapped charge suppresses ferroelectric surface distortions, resulting in a binary CO adsorption process.

[1] M. Setvin et al. Science 359, 572 (2018). [2] M. Reticcioli et al., Nat. Comm. (2022). [3] Z. Wang et al., Sc. Adv. (2022).

O 66.2 Thu 11:00 S053

Surface phonons and their coupling to the 2D electron liquid at the CTO/STO surface — •M. Döttling[1], E. B. Guedes[2,3], T. P. van Waas[4,2], S. Poncé[4,5], M. Caputo[3,6], N. C. Plumb[2], N. Marzari[5], C. Berthod[7], M. Radović[2], J. H. Dil[3,2], and K. Zakeri[1] — [1]PHI, Karlsruhe Institute of Technology — [2]PSD, PSI — [3]IPHYS, EPF Lausanne — [4]IMCN, UC Louvain — [5]THEOS, EPF Lausanne — [6]Elettra, Trieste — [7]DQMP, University of Geneva

Motivated by the recent discovery of a 2D electron liquid (2DEL) formed at the surface of $CaTiO_3$/$SrTiO_3$(001) heterostructure, we probed the phonon spectrum of the system by means of high-resolution electron energy-loss spectroscopy (HREELS). Our main aim was to address any possible interaction of 2DEL with phonons.

The HREELS measurements revealed the existence of the Fuchs-Kliewer (FK) phonon modes at the energies of 20, 60 and 93 meV. Interestingly, the Eliashberg-function $\alpha^2F(\omega)$ constructed based on the quasiparticle band dispersion, probed by angle-resolved photoemission experiments, exhibits peaks at energies of 20, 60 and 80 meV.

The apparent redshift of the highest energy peak in $\alpha^2F(\omega)$, compared to the 93 meV FK mode, can be attributed to the formation of band-gap states (BGS). Such states form upon photon irradiation at the same time as 2DEL is formed. This is shown by the simulated HREEL spectra including BGS. Our results demonstrate the importance of BGS and the necessity of their consideration in describing the properties of oxide heterostructures. Supported by DFG via Heisenberg Programme ZA 902/3-1, ZA 902/6-1 and ZA 902/5-1.

O 66.3 Thu 11:15 S053

Reversible switching of the 2D electron gas of $SrTiO_3$(001) studied by HREELS — •Hannes Herrmann, Florian Schumann, and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg, Institute of Physics, 06120 Halle, Germany

The formation of a 2D electron gas (2DEG) gained high interest in surface science as it became accessible under oxygen-poor conditions at the TiO_2-terminated $SrTiO_3$(001) surface [1]. In this work, we follow the 2DEG formation by studying the $SrTiO_3$ surface plasmon polaritions (SPP) by high-resolution electron energy loss spectroscopy (HREELS). The coupling of the SPP to the 2DEG leads to an asymmetric SPP line shape with extreme broadening. This strong electron-phonon coupling allows to quantify the 2DEG charge carrier density and is used to follow the reversible formation of the 2DEG by heating in UHV/O_2 atmosphere or by adsorbing molecular oxygen at 100 K. The dielectric response will be discussed in terms of a two-layer model with a surface 2DEG which is modelled using a Drude-like response with a strongly frequency-dependent electronic mobility.

[1] A. F. Santander-Syro, O. Copie, A. Barthelemy, and M. J. Rozenberg, Nature 469, 189 (2011).

O 66.4 Thu 11:30 S053

Structure determination of a dodecagonal oxide quasicrystal — •Sebastian Schenk[1], Oliver Krahn[1], Holger L. Meyerheim[2], Marc De Boissieu[3], Stefan Förster[1], and Wolf Widdra[1] —

[1]Institute of Physics, Martin-Luther-Universität Halle-Wittenberg, 06120 Halle, Germany — [2]Max-Planck-Institut für Mikrostrukturphysik, 06120 Halle, Germany — [3]Universite Grenoble Alpes, CNRS, SIMaP, St Martin d Heres, France

The dodecagonal symmetry and the general tiling motive of oxide quasicrystals (OQC) have been proven by electron diffraction (LEED) and scanning tunneling microscopy (STM) from their first observations [1,2]. However, the detailed atomic structure, which includes the decoration of individual tiling elements with metal and oxygen atoms, has been under debate for more than eight years.

In this talk we solve the atomic structure by combination of STM and surface x-ray diffraction (SXRD) of a quasicrystal approximant, which is derived from an ultrathin layer of SrTiO$_3$ on Pt(111). The derived structure model can be generalized for all related ternary oxide quasicrystals and their approximants. The measured diffraction intensities will be compared with calculated ones by lifting the atomic structure of the OQC into its four-dimensional periodic superlattice. These intensities will be compared to SXRD data obtained in synchrotron measurements.

[1] S. Förster et. al., Nature **502**, 215 (2013).
[2] S. Schenk et. al., J. Phys.: Condens. Matter **29**, 134002 (2017).

O 66.5 Thu 11:45 S053

Structural analysis of complex 2D Sr-Ti-O/Pd(111) films. — •Martin Haller, Sebastian Schenk, Stefan Förster, and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg, Institute of Physics, Von-Danckelmann-Platz 3, 06120 Halle, Germany

In 2013 the first 2D oxide quasicrystal (OQC) in thin Ba-Ti-O/Pt(111) films with astonishing structural properties has been discovered [1]. It features a twelvefold rotational symmetry that is incompatible with lattice periodicity. Instead, it is described with a self-similar quasicrystalline structure that involves squares, triangles and rhombuses. In the wake of this discovery combinations of ternary oxide layers and different hexagonal substrates have been investigated to study the epitaxial preconditions for the formation of the OQC. In this contribution, we present a structural analysis for Sr-Ti-O on Pd(111). Globally, as seen from LEED, it forms a periodic structure with unit cell parameters a = 1.32 nm, b = 2.96 nm and α = 93.43°. However, locally a second phase has been identified in atomically-resolved STM data with twelvefold symmetric FFT and long range coherence in the 2D auto-correlation image. The analysis of the tiling statistics reveals a triangle:square:rhomb ratio of 2.84:1:0.39 which deviates slightly from the ideal OQC's ratio of 2.73:1:0.37 [2]. The given structure is discussed in terms of a disordered OQC which is further supported by a 4D hyperspace analyis.

[1] S. Förster et al., Nature 502, 215 (2013)
[2] S. Schenk et al., J. Phys. Condens. Matter, 29 (2017)

O 66.6 Thu 12:00 S053

Transformation from honeycomb binary oxide to quasicrystalline ternary oxide — •Loi V. Tran, Stefan Förster, and Wolf Widdra — Institute of Physics, Martin-Luther-University Halle-Wittenberg, Halle, Germany

Oxide quasicrystals (OQCs) are examples of aperiodically-ordered 2D materials. They have been found for 2D layers derived from BaTiO$_3$ on Pt(111) [1]. Their dodecagonal structure results from Ba decoration of a Ti$_n$O$_n$ network where n = 4 (decorated with no Ba atom), 7 (decorated with one Ba atom) and 10 (decorated with two Ba and two addional O atoms). In contrast, periodic Ti$_2$O$_3$ honeycomb structures with Ti$_6$O$_6$ rings are well known in literature [2,3]. Here, we report on the transition of such honeycomb Ti$_2$O$_3$ structures to dodecagonal OQCs by adsorption of Ba and subsequent annealing. The structural transition is monitored with LEED and workfunction measurements. Upon room temperature Ba deposition, Ti$_2$O$_3$ honeycomb structure remains and Ti$_6$O$_6$ rings are decorated with Ba, reducing the workfunction from 5.12 to 2.97 eV. Annealing to 873 K in oxygen converts the honeycomb structure into a dodecagonal OQC, for which the workfunction increases to 3.85 eV. The workfunction changes associated with the transformation will be discussed in context of the height differences of Ba ions above the surface and the resulting dipole strength.

O 66.7 Thu 12:15 S053

Barium decorated Ti$_2$O$_3$ monolayers: a case study for Pd(111) substrates — •Friederike Wührl, Sebastian Schenk, Oliver Krahn, Stefan Förster, and Wolf Widdra — Institute of Physics, Martin-Luther-Universität Halle-Wittenberg, 06120 Halle, Germany

Oxides at the two-dimensional limit show a high flexibility in their structural properties ranging from hexagonal honeycomb structures to dodecagonal oxide quasicrystals (OQC) [1]. Recent work based on STM, surface x-ray diffraction (SXRD) and DFT calculations confirmed a structure model for oxide quasicrystals that explains this extraordinary structure as an aperiodically ordered Ti$_2$O$_3$ network with rings of different sizes on the metal support Pt(111), in which the largest rings are decorated with Ba ions [2]. The Ba ions form a self-similar tiling, which can be described through triangles, squares and rhombuses. In this contribution we report on a sequence of long-range ordered structures observed in Ba$_x$Ti$_2$O$_{3+y}$ on Pd(111) that are closely related to the OQC. We present a series of structures ranging from a Ba$_{0.66}$Ti$_2$O$_3$ to a BaTi$_2$O$_{3.5}$ stoichometry. By varying the Ba content, we observe a transition from a triangle-square tiling via triangle-square-rhombus tilings to one consisting of triangles and rhombuses only [3]. It will be discussed how the Ba density sets the ratio of four to seven to ten membered rings in the Ti$_2$O$_3$ backbone.

[1] S. Förster *et al.*, Nature **502**, 215 (2013).
[2] E. Cockayne *et al.*, Phys. Rev. B **93**, 020101(R) (2016).
[2] F. E. Wührl *et al.*, Phys. Status Solidi B, 2100389 (2021).

O 66.8 Thu 12:30 S053

The (2x1) reconstruction of calcite(104) — Jonas Heggemann[1], Yashasvi Ranawat[2], Ondřej Krejčí[2], Adam S. Foster[2], and •Philipp Rahe[1] — [1]Fachbereich Physik, Universität Osnabrück, Barbarastraße 7, 49076 Osnabrück, Germany — [2]Department of Applied Physics, Aalto University, Helsinki FI-00076, Finland

Calcite is an abundant material in the Earth's crust, a central constituent of biominerals in living organisms [1], and currently investigated as a capture material for CO2 [2]. Despite intensive studies, however, there is still serious ambiguity regarding the properties of this surface due to conflictive conclusions for the so-called row-pairing [3] and (2x1) reconstruction [4].

Here, we use a combination of non-contact atomic force microcopy (NC-AFM) with CO-functionalized tips at 5K, density functional theory with state-of-the-art dispersion corrections, and NC-AFM image simulations to clarify the microscopic geometry of calcite(104). A (2x1) reconstruction and a glide plane symmetry is consistently found in the NC-AFM data and DFT results. Most importantly, we identify two different adsorption positions for CO molecules within the (2x1) unit cell. These findings are most critical for future studies where processes on calcite(104)-(2x1) are influenced by the surface geometry.

[1] L. Addadi, S. Weiner, Angew. Chem. Int. Ed. Engl. 31, 153 (1992). [2] P. A. E. Pogge von Strandmann, et al., Nat. Commun. 10, 1983 (2019). [3] A. L. Rachlin, et al., Am. Mineral. 77, 904 (1992). [4] S. L. S. Stipp, et al., Geochim. Cosmochim. Acta 58, 3023 (1994).

O 66.9 Thu 12:45 S053

Growth of ultra thin Eu$_x$O$_y$ films on Pt(001): A HREELS, XPS and LEED study — •Anne Oelschläger, Hannes Herrmann, Stefan Förster, and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg, Institut of Physics, 06120 Halle, Germany

The gradually decreasing ionic radii along the series of lanthanides allow an exceptional fine tuning of lattice parameters, which makes this material class interesting for applications in epitaxial growth. In this contribution we report on the growth of thin Eu$_x$O$_y$ films on Pt(001) and characterize the thin film oxidation upon annealing in various environments.

A 0.8 nm thin Eu layer has been deposited onto Pt(001) at room temperature in UHV conditions at a background pressure below $3 \cdot 10^{-10}$ mbar. By monitoring the surface phonons with HREELS and the valence state of Eu with XPS a continuous oxidation of Eu at room temperature by water from the residual gas as well as by molecular oxygen is observed as a transition from metallic Eu to Eu$_2$O$_3$. The transition to Eu^{3+} can be followed by increasing (decreasing) intensities for the surface phonon polariton at 364 cm^{-1} (525 cm^{-1}) in HREELS and the high binding energy component of the Eu3d core level at 1135 eV (1125 eV) in XPS. The long-range order starts to develop in the 0.8 nm thin layer upon annealing to 1000 K. LEED exhibits a complex twelve-fold diffraction pattern.

O 67: Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 3

Time: Thursday 10:30–12:45 Location: S054

O 67.1 Thu 10:30 S054

Quantile Random Forest Model for Extrapolation to the Complete Basis Set Limit in Density Functional Theory Calculations — •Daniel Speckhard[1], Christian Carbogno[2], Sven Lubeck[2], Luca Ghiringhelli[2], Matthias Scheffler[2,1], and Claudia Draxl[1,2] — [1]Humboldt-Universität zu Berlin, Physics Department and IRIS Adlershof, Berlin, Germany — [2]The NOMAD Laboratory at the FHI-MPG and HU, Berlin, Germany

The precision of density-functional theory (DFT) calculations depends on a variety of computational parameters, the most critical being the basis-set size. With an infinitely large basis set, i.e., in the limit of a complete basis set (CBS), the result of the calculation is as precise as possible for the chosen exchange-correlation functional. Our aim in this work is to find a model that can extrapolate the result of an imprecise DFT calculation to the CBS limit. As a starting point, we use a dataset of 63 binary solids investigated with various basis-set sizes [1] with two all-electron DFT codes, `exciting` and *FHI-aims*, which use very different types of basis sets. A quantile random forest model is used to estimate the deviation of the total energy with respect to fully converged calculations as a function of the basis set size. The non-linear random forest model outperforms a previous approach that used a linear model. The quantile random forest model presented also provides prediction intervals which give the user an idea of the model's uncertainty.
[1] C. Carbogno *et al.*, *npj Comput. Mater.* **8**, *69 (2022).*

O 67.2 Thu 10:45 S054

Symmetry and completeness in machine-learning models for atomistic simulations — •Sergey Pozdnyakov and Michele Ceriotti — EPFL, Switzerland

During the last decade, machine learning methods have drastically changed atomistic simulations. On the one hand, they scale linearly with the size of the system and thus, are significantly faster than the quantum mechanical calculations. On the other, they provide a functional form that is much more flexible than so-called classical force fields such as the Lennard Jones potential or embedded atom models. From one point of view, incorporating rotational symmetry is important for ML since it can make models more data-efficient and robust, but can also lead to incompleteness, limiting the ultimate accuracy of the model. I will discuss some examples of this and compare different types of models to show how one can find an optimal balance of the two effects.

O 67.3 Thu 11:00 S054

Fast, robust, interpretable machine-learning potentials — Stephen R. Xie[1,2], Richard G. Hennig[1], and •Matthias Rupp[3] — [1]University of Florida, Gainsville, USA — [2]KBR, NASA Ames Research Center, Mountain View, USA — [3]University of Konstanz, Germany

Machine-learning potentials (MLPs) are increasingly successful in all-atom dynamics simulations where they act as surrogate models for ab-initio electronic structure methods. MLPs often result in two to three orders of magnitude improvements in the number of simulated atoms or duration of simulated time, enabling new insights and applications. Current limitations include data inefficiency, instabilities ("holes" in high-dimensional MLPs [2]), and lack of interpretability.

To address this challenge, we combine effective two- and three-body potentials in a cubic B-spline basis with second order-regularized linear regression. The resulting "ultra-fast potentials" are data-efficient, physically interpretable, sufficiently accurate for applications, can be parametrized automatically, and are as fast as the fastest traditional empirical potentials. [1] We demonstrate these qualities in retrospective benchmarks and present the prediction of thermal conductivities via the Green-Kubo formalism as a first application.

[1] Stephen R. Xie, Matthias Rupp, Richard G. Hennig, Ultra-fast interpretable machine-learning potentials. arXiv:2110.00624, 2021 [2] Jeffrey Li, Chen Qu, Joel M. Bowman: Diffusion Monte Carlo with fictitious masses finds holes in potential energy surfaces, Mol. Phys. 119(17–18): e1976426, 2021.

O 67.4 Thu 11:15 S054

Improving the transferability of high-dimensional neural network potentials by low-order terms — •Alea Miako Tokita and Jörg Behler — Georg-August-Universität Göttingen, Institut für Physikalische Chemie, Theoretische Chemie, Tammannstraße 6, 37077 Göttingen, Germany

High-dimensional neural network potentials (HDNNPs) are able to provide accurate potential energy surfaces suitable for atomistic simulations of large systems. The key to this accuracy is the high flexibility of the atomic neural networks allowing to reproduce energies and forces from reference electronic structure calculations with very small errors. At the same time, this flexibility is limiting the transferability of HDNNPs to atomic configurations that are very different from the reference geometries. Here, we investigate possible improvements in transferability of HDNNPs by the explicit inclusion of low-order terms in the functional form of the potential. The performance is demonstrated for a series of molecular model systems.

O 67.5 Thu 11:30 S054

Predicting condensed-phase electron densities using machine learning — •Alan Lewis[1], Andrea Grisafi[2], Michele Ceriotti[2], and Mariana Rossi[1] — [1]MPI for Structure and Dynamics of Materials, Hamburg, Germany — [2]École Polytéchnique Fédèrale de Lausanne, Lausanne, Switzerland

The electron density is a fundamental quantity for understanding physical phenomena in materials, and is central to electronic structure theories such as density-functional theory. We present the SALTED machine learning method and demonstrate its ability to learn and predict the electronic densities of a range of materials from simple liquids and metals to hybrid organic-inorganic perovskites. This extends the framework presented in ACS Cent. Sci. 5, 57, 2019 to work with periodic boundary conditions and uses a resolution of the identity on a numeric atom-centered orbital basis to expand the all-electron periodic density. A Gaussian process regression model that makes use of local symmetry-adapted representations of the atomic structure is employed, making our method both data-efficient and highly transferable.[1] We also compare various methods of dealing with the non-orthogonality of the basis, accounting for correlations between pairs of off-centered density components, finding that the best compromise between accuracy and computational efficiency comes from approximating the density expansion coefficients by directly minimizing the loss function. The total energies derived from the densities obtained in this way present errors with respect to DFT of just 0.1 meV/atom.

[1] Lewis, Grisafi, Ceriotti, Rossi, JCTC 17, 11, 7203 (2021)

O 67.6 Thu 11:45 S054

Equivariant N-center representations for machine learning molecular Hamiltonians — •Jigyasa Nigam, Michael Willatt, and Michele Ceriotti — Laboratory of Computational Science and Modeling, Institute of Materials, Ecole Polytechnique Federale de Lausanne, 1015 Lausanne, Switzerland

Most of the widely used machine learning schemes that have been successful in predicting chemical and material properties rely on concise, symmetry-adapted descriptions of the underlying atomic structure. A class of these structural descriptions is built on hierarchical correlations of atom-centered densities(ACDC)[1]. These are subsequently used to model corresponding atomic properties or atomic contributions to a global observable. However, many quantum mechanical quantities, such as the effective single-particle Hamiltonian written on an atomic-orbital basis, are associated with multiple atom-centers. This effectively renders ACDCs inadequate to describe the additional degrees of freedom of such multicenter properties. We recently proposed an N-centered representation[2] that extends the ACDC framework to the case of targets that are simultaneously indexed by N atoms. I will demonstrate how devising a family of N-center representations opens avenues for new classes of machine learning models that are fully equivariant and describe their role in assisting electronic structure calculations.

[1] J. Nigam, S. Pozdnyakov, M. Ceriotti, JCP 153,121101, 2020
[2] J. Nigam, M. Willatt, M. Ceriotti, JCP 156, 014115, 2022

O 67.7 Thu 12:00 S054

Similarity-of-materials analysis for reusability and interoperability of data in materials databases — •Šimon Gabaj, Martin Kuban, Santiago Rigamonti, and Claudia Draxl — Humboldt-Universität zu Berlin, Zum Großen Windkanal 2, 12489 Berlin, Germany

Large computational materials databases, such as NOMAD [1-2], make it possible to reuse already generated materials data. All these data typically come from different sources and have been created for a different purpose, be it geometry optimization, electronic structure, or alike. To support interoperability and thus reusability of such data, we devise a data-analysis workflow making use of similarity fingerprints. First, we encode the electronic density of states (DOS) in a vectorial representation [3] to obtain a descriptor. Then, we employ the Tanimoto coefficient to compute the similarity between all pairs of calculations. We demonstrate our workflow with selected materials, chosen from the NOMAD database. In the ideal case, all calculations of the same material in the same geometry should be identical. This is, however, not observed. Using our approach, we can uncover correlations between the DOS similarity and methodology as well as computational parameters. This way, we can also identify parameters that are relevant for the convergence of results.

[1] Draxl, C., Scheffler, M., *MRS Bulletin*, **43**, 676, (2018)
[2] Draxl, C., Scheffler, M., *J. Phys. Mater.*, **2**, 036001, (2018)
[3] Kuban, M., *et al.*, to be published

O 67.8 Thu 12:15 S054

Supervised and unsupervised deep Learning of topological phase transitions from entanglement aspect for one- and two-dimensional chiral p-wave superconductors — •Ming-Chiang Chung — Max-Planck-Institut für Physik komplexer Systeme, Dresden, Germany — National Chung-Hsing University, Taichung, Taiwan

The one-dimensional or two-dimensional chiral p-wave superconductor proposed by Kitaev has long become a classic example for understanding topological phase transitions through various methods, such as examining the Berry phase, edge states of open chains, and, in particular, aspects from quantum entanglement of ground states. In order to understand the amount of information carried in the entanglement-related quantities, here we study topological phase transitions of the model with emphasis of using the deep learning approach. Using both supervised or unsupervised ways, we feed different quantities, including Majorana correlation matrices (MCMs), entanglement spectra (ES) or entanglement eigenvectors (EE) originating from Block correlation matrices, into the deep neural networks for training, and investigate which one could be the most useful input format in this approach. We find that ES is information that is too compressed compared to MCM or EE. MCM and EE can provide us abundant information to recognize not only the topological phase transitions in the model but also phases of matter with different U(1) gauges, which is not reachable by using ES only. We also build a procedure for using unsupervised learning to find the phase transition points. We have used this method for other models.

O 67.9 Thu 12:30 S054

Machine Learning the Square-Lattice Ising Model — •Burak Çivitcioğlu[1], Andreas Honecker[1], and Rudolf A. Römer[2] — [1]Laboratoire de Physique Theorique et Modelisation, CNRS UMR 8089, CY Cergy Paris Universit *e, Cergy-Pontoise, France — [2]Department of Physics, University of Warwick, Coventry, CV4 7AL, United Kingdom

Recently, machine-learning methods have been shown to be successful in identifying and classifying different phases of the square-lattice Ising model. We study the performance and limits of classification and regression models. In particular, we investigate how accurately the correlation length, energy and magnetisation can be recovered from a given configuration. We find that a supervised learning study of a regression model yields good predictions for magnetisation and energy, and acceptable predictions for the correlation length.

O 68: Focus Session: Time-Resolved Momentum Microscopy

Time-resolved momentum microscopy is a new experimental technique to study electron dynamics in momentum space. In momentum microscopy, the reciprocal image plane of photoemitted electrons is mapped onto a position-sensitive detector. In addition, the kinetic energy is extracted by energy dispersive elements, such as Time-of-Flight drift tubes or hemispherical analysers. Hereby, the complete information about the electronic structure of two-dimensional materials can be obtained (kx, ky, E). At the same time the signal can be restricted to micrometer-size spots, which is important for many types of quantum materials. By combining such electron spectrometers with pump-probe laser techniques, optical excitations, photo-induced phase transitions and charge transfer processes within the entire Brillouin zone can be imaged on ultrashort time scales. Several groups, with many of them in Germany, have established this technique during the last years in lab-based setups. In addition, first setups at large scale facilities, like ELI-ALPS or FLASH, are being commissioned or in operation. With higher photon energies available at free electron lasers, also structural dynamics will be observable through time-resolved electron diffraction.

Organizer: Robert Wallauer (Universität Marburg)

Time: Thursday 15:00–18:30 Location: H3

Topical Talk O 68.1 Thu 15:00 H3

Exploring Excitonic Excitations in Momentum Space — •Keshav Dani — Okinawa Inst. of Science and Technology, Graduate University, Onna-son, Japan

Optical techniques have provided us with rich information about the exciton * a two-particle photoexcited state in semiconductors and insulators. Yet, they have left a fundamental degree of freedom of the exciton inaccessible * it*s momentum! In this talk, I will discuss the application of time-resolved photoemission techniques to access the momentum coordinate of excitons in 2D semiconductors, thereby providing us with the formation pathways of momentum-forbidden dark excitons [1], an image of the electron around the hole in the exciton, the observation of the long-predicted anomalous dispersion of the exciton-bound electron [2], the momentum distribution of the exciton-bound hole, and the confinement of the interlayer exciton in a moiré cell [3].

References (*equal authors) [1] J. Madeo*, M. K. L. Man*, et al. Science 370, 1199 (2020). [2] M. K. L. Man*, J. Madeo*, et al. Science Advances 7, eabg0192 (2021). [3] O Karni*, E. Barr*, V. Pareek*, J. D. Georgaras*, M. K. L. Man*, C. Sahoo*, et al. Nature 603, 247 (2022).

Topical Talk O 68.2 Thu 15:30 H3

Moiré interlayer and charge-transfer excitons in space and time: new experiments enabled by time-resolved momentum microscopy — •Stefan Mathias — I. Physikalisches Institut, Georg-August-Universität Göttingen, Germany

In my talk, I introduce the time-resolved momentum microscopy setup that we developed in Göttingen (Germany) [1], which includes a MHz repetition rate extreme-ultraviolet beamline, and has recently been upgraded with a spin-imaging detector. In the following, I will discuss exemplary research projects that we cover with this new instrument. In particular, I will focus on the spatio-temporal identification and dynamics of moiré interlayer excitons in twisted WSe2/MoS2 heterostructures [2], and on an orbital-resolved study [3] of charge-transfer exciton dynamics in C60 thin films [4].

[1] Keunecke et al., Rev. Sci. Instr. 91, 063905 (2020)
[2] Schmitt et al., arXiv:2112.05011(2021)
[3] Jansen et al., New Journal of Physics 22, 063012 (2020)
[4] Stadtmüller et al., Nature Communications 10, 1470 (2019)

O 68.3 Thu 16:00 H3

The complete interface molecular movie: One-stop imaging of orbital, electronic, and structural dynamics — •Markus Scholz[1], Kiana Baumgärtner[2], Marvin Reuner[3], Christian Metzger[2], Michael Heber[1], Dmytro Kutnyakhov[1], Friedrich Reinert[2], Daria Popova-Gorelova[3], Martin Beye[1], and Kai Rossnagel[4] — [1]Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, 22607 Hamburg, Germany — [2]Experimentelle Physik 7, Julius-Maximilians-Universität, Am Hubland, 97074 Würzburg, Germany — [3]I. Institute for Theoretical Physics and Centre for Free-Electron Laser Science, Universität Hamburg, Luruper Chaussee 149, 22607 Hamburg, Germany — [4]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

Reactions of molecules on surfaces involve a complex interplay between electronic reorganization and the motion of atoms. The recent developments of higher-harmonics generation (HHG) and free-electron laser (FEL) based sources open up new opportunities for ultrafast spectroscopy toward simultaneously imaging subtle changes in the electronic structure and atomic positions on a femtosecond time scale and with sub-Ångström spatial resolution. Here, using a dual-electron messenger mode of momentum microscopy, we trace a photoinduced charge transfer process in energy-momentum space using valence electrons and, synchronously, at atomic sites using core electrons, in a single experimental setup for a molecule-2D material interface.

O 68.4 Thu 16:15 H3

Electron dynamics after a spin- and valley-polarized electronic excitation in WS_2 — •Lasse Münster[1], Sarah Zajusch[1], Raul Perea-Causin[1], Samuel Brem[1], Katsumi Tanimura[1], Jens Güdde[1], Yaroslav Gerasimenko[2], Rupert Huber[2], Ermin Malic[1], Ulrich Höfer[1], and Robert Wallauer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Fachbereich Physik, Universität Regensburg, Germany

Atomically thin layers of TMDCs offer an ideal playground to study ultrafast electron dynamics. After optical excitation electrons scatter throughout the Brillouin zone to form a variety of excitonic states. We have shown that this formation process can be imaged by time-resolved momentum microscopy with tunable pump and high harmonic probe [1].

We recently incorporated a new pumping scheme, that allows us to excite the sample under an incident angle close to 0° with circularly polarized light. This results in an excitation, which is located purely within the K valley for one helicity and in the K' valley for the other helicity and provides access to all possible scattering processes. In the case of an excitation at K, electron scattering to K' and Σ is mediated by strong electron-phonon coupling with time constants of a few tens of femtoseconds. In addtion, we observe the formation of spin-forbidden excitons in the K valley and electron scattering towards Σ'. Both of these processes involve a spin-flip and are significantly slower (50 - 100 fs).

[1] R. Wallauer et al., Nano Lett. 21, 5867 (2021)

Topical Talk O 68.5 Thu 16:30 H3

Momentum and energy dissipation of hot electrons in metals and metal-molecular heterostructures — •Benjamin Stadtmueller — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, 67663 Kaiserslautern, Germany — Institute of Physics, JGU Mainz, 55128 Mainz, Germany

The dynamics of optically excited carriers in condensed matter play a crucial role in many fundamental and device-relevant processes in materials. Here, I discuss the potential of time-resolved two-photon momentum microscopy (tr-2PMM) [1] to access the k-space signatures and the corresponding quasi-particle lifetimes of excited carriers in metals [2,3] and metal-molecular heterostructures. For metallic systems, I will demonstrate that tr-2PMM allows us to uncover anisotropies in the orientations of optical transition dipoles for highly free electron like noble metal surfaces [2] and to follow the ultrafast intra- and interband scattering processes of optically excited carriers in metallic quantum well systems [3] in real-time. In this context, I will also introduce two approaches to alter the electron dynamics of metal surfaces by the adsorption of molecular complexes or by periodically modulating the surface potential using porous molecular networks on surfaces. Finally, I will show our efforts to exploit the spatial resolution in tr-2PMM to disentangle the k-space signatures of photo- and plasmon-induced hot carriers at surfaces [4]. [1] F. Haag et al., Rev. Sci. Instrum. 90, 103104 (2019); [2] T. Eul et al. Nat. Commun. (2022); [3] F. Haag et al. Phys. Rev. B 104, 104308 (2021); [4] M. Hartelt et al. ACS Nano 15, 19559 (2021)

O 68.6 Thu 17:00 H3

Coherent response of the electronic system driven by non-interfering laser pulses — •Tobias Eul[1], Eva Prinz[1], Michael Hartelt[1], Benjamin Frisch[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Germany

The strength of light-matter interaction in condensed matter is fundamentally linked to the orientation and oscillation strength of the materials' optical transition dipoles. Structurally anisotropic materials, e.g. elongated molecules, exhibit optical transition dipoles with fixed orientations that govern the angular-dependent light-matter interaction. Contrary, free electron like metals should exhibit isotropic light-matter interaction with the light fields dictating the orientation of the optical transition dipoles. Here, we demonstrate that an anisotropic direction of the optical transition dipoles even exists in highly free electron like noble metal surfaces. Our time- and phase-resolved photoemission experiment reveals coherent interference effects on the (110)-oriented silver surface after optical excitation with two non-interfering cross-polarized pulses. We explain this coherent material response within the density matrix formalism by an intrinsic coupling of the non-interfering light fields mediated by optical transition dipoles with fixed orientations in silver.

O 68.7 Thu 17:15 H3

Time-resolved photoemission orbital tomography of CuPc on Cu(001)-2O — •Alexa Adamkiewicz[1], Miriam Raths[2], Monja Stettner[2], Francois C. Bocquet[2], Christian Kumpf[2], Robert Wallauer[1], F. Stefan Tautz[2], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps Universität Marburg, Germany — [2]Peter Grünberg Institute (PGI-3), Jülich Research Centre, Germany

Charge transfer across molecular interfaces is reflected in the population of electronic orbitals. For ordered organic layers, time-resolved photoemission orbital tomography (tr-POT) is capable of spectroscopically identifying the involved orbitals and deducing their population from the measured angle-resolved photoemission intensity with high temporal resolution. In a first example, we found that for PTCDA/Cu(001)-2O, two distinct excitation pathways could be observed with visible light [1]. The parallel component of the electric field induces a direct HOMO-LUMO transition, the perpendicular component transfers a substrate electron into the molecular LUMO. While in this case, a distinct excitonic signature was not observed, changes in the momentum pattern can in general serve as a measure of detecting excitonic processes. Here, we show such time-dependent change of the pattern for CuPc/Cu(001)-2O. We demonstrate how the temporal evolution of the LUMO momentum distribution can be systematically disentangled from contributions of the projected HOMO. Moreover, we observe LUMO excitation of selected molecular orientation at normal incidence by aligning the pump polarization along the molecular axis. [1] R. Wallauer et al., Science 371, 1056 (2021).

Topical Talk O 68.8 Thu 17:30 H3

Is there a perfect electron analyzer for time-resolved ARPES? — •Laurenz Rettig — Fritz-Haber-Institut der Max-Planck-Gesellschaft

Recent years have seen a huge popularity of time-of-flight based momentum microscopes (MMs), which based on advanced electrostatic optics have revolutionized angle-resolved photoemission spectroscopy (ARPES). Compared to conventional hemispherical analyzers (HAs) with angle-dispersing electron lenses, which access only a small fraction of the reciprocal space at a given configuration, MMs allow for the simultaneous detection of multiple Brillouin zones without the need to rearrange the sample geometry. However, one drawback of such instruments, in particular in time-resolved studies, arises from the large energy and momentum range covered simultaneously, which in combination with detection limitations of delay-line detectors and space charge restrictions can severely reduce the effective detection rate for selected energy-momentum regions compared to conventional HAs.

In my talk I will discuss the advantages and limitations of both types of instruments for several application scenarios in time-resolved ARPES, and present some recent highlights in charge-density wave materials, which take advantage of the combination of both types of instruments.

O 68.9 Thu 18:00 H3

Developing a fs-XUV source for time-resolved momentum microscopy on 2D materials — •Karl Schiller[1], Alan Omar[2], Lasse Sternemann[1], Matija Stupar[1], Clara Saraceno[2], and Mirko Cinchetti[1] — [1]Department of Physics, TU Dortmund University, Germany — [2]Ruhr-Universität Bochum, Germany

We introduce a newly developed setup for time-resolved momentum microscopy with femtosecond extreme ultraviolet (fs-XUV) radiation. The fs-XUV pulses with photon energy up to 30 eV are generated by high-harmonic generation in an Argon gas jet and are coupled to an energy-filtered momentum microscope (KREIOS MM, Specs GmbH). A commercial Ytterbium-based high-power laser system drives the generation with a variable high repetition rate between 100 kHz and 1 MHz (Carbide, Light Conversion). While this configuration allows for high energy resolution of 50 meV, a high temporal resolution can also be reached by coupling the laser to a tunable in-house build Herriot-type multipass cell compressor with peak powers up to 2 GW and pulse durations of ∼ 45 fs [1]. In the talk, we will present the first characterization data of this setup and motivate its capability to examine few-layer 2D materials.

[1] A. Omar, et al. *Advanced Solid State Lasers*, OSA Technical Digest (Optical Society of America, 2021), paper JM3A.55

O 68.10 Thu 18:15 H3

FEL-based time-of-flight momentum microscopy: 3 time-resolved photoemission modalities in 1 experiment — •D. Kutnyakhov[1], R.P. Xian[2], M. Dendzik[2], M. Heber[1], F. Pressacco[3], S.Y. Agustsson[4], L. Wenthaus[1], H. Meyer[3], S. Gieschen[3], K. Bühlman[5], S. Däster[5], R. Gort[5], D. Curcio[6], K. Volckaert[6], M. Bianchi[6], Ch. Sanders[6], J.A. Miwa[6], S. Ulstrup[6], A. Oelsner[7], C. Tusche[8,9], Y.-J. Chen[8,9], D. Vasilyev[4], K. Medjanik[4], G. Brenner[1], S. Dziarzhytski[1], S. Dong[2], J. Hauer[2], L. Rettig[2], J. Demsar[4], H.-J. Elmers[4], Ph. Hofmann[6], R. Ernstorfer[2], G. Schönhense[4], Y. Acremann[5], and K. Rossnagel[1,10] — [1]DESY, Hamburg — [2]FHI Berlin — [3]CFEL, Univ. Hamburg — [4]Univ. Mainz — [5]ETH Zürich — [6]Univ. Aarhus — [7]Surface Concept GmbH, Mainz — [8]FZ Jülich GmbH — [9]Univ. Duisburg-Essen — [10]CAU Kiel

Time-resolved photoemission spectroscopy with ultrashort pump and probe photon pulses is an emerging technique with wide application potential. The ultimate combination of valence-band and core-level spectroscopy with photoelectron diffraction in a single experiment for electronic, chemical, and structural dynamics analysis specifically requires tunable monochromatic soft X-ray pulses at a high repetition rate as well as highly efficient single-shot electron detectors with increased multi-hit capabilities. We have realized such a 3-in-1 ultrafast photoemission experiment at FLASH/PG2, DESY merging free-electron laser capabilities with a multi-dimensional recording scheme.

O 69: Surface Reactions and Heterogeneous Catalysis 2

Time: Thursday 15:00–17:45 Location: H4

Topical Talk O 69.1 Thu 15:00 H4

Theoretical Investigations of Size and Support Effects in Heterogeneous Catalysis — •Felix Studt — Institute of Catalysis Research and Technology, Karlsruhe Institute of Technology

Supported transition metal nanoparticles play an important role as catalysts in heterogeneous catalysis. The extend to which size, shape and metal support interaction influence a catalysts reactivity is still at the forefront of scientific research. Here we use density functional theory calculations on nanoparticles ranging from 0.5 to about 3.5 nm in size in order to gain insight into the particle size effect on a catalysts reactivity. We furthermore investigate how the interaction of nanoparticles with the support alters their reactivity with respect to the binding strength of adsorbates. Using computational models of particle-support interfaces we show how insight into changes in reactivity can be described.

O 69.2 Thu 15:30 H4

Why interlayer exchange is crucial for temperature programmed desorption — •Tobias Dickbreder, Ralf Bechstein, and Angelika Kühnle — Physical Chemistry I, Bielefeld University, Universitätstraße 25, 33615 Bielefeld

Understanding the desorption of molecules from surfaces is fundamental for both natural and application-oriented processes such as dewetting, weathering and catalysis. A powerful method to investigate desorption processes is temperature programmed desorption (TPD) as it offers the possibility to gain mechanistic insights into the desorption kinetics. In the past, several analysis methods have been developed for TPD data. These methods have in common that they rely on the Polanyi-Wigner equation, which requires proposing a desorption mechanism with a single (dominating) desorption path. For real systems, however, several coupled desorption paths can be easily envisioned. Here, we analyze the influence of exchange between the first and the second adsorbate layer on the desorption process. We show that considering this additional desorption pathway alters the desorption spectrum considerably. Thus, our study demonstrates that interlayer exchange can be crucial for the analysis of TPD data.

O 69.3 Thu 15:45 H4

CO oxidation on small size-selected Pt clusters supported on $Fe_3O_4(001)$ — •Johanna Plansky[1], Sebastian Kaiser[1], Farahnaz Maleki[2], Ke Zhang[3], Wolfgang Harbich[4], Ueli Heiz[1], Sergio Tosoni[2], Barbara A.J. Lechner[1], Gianfranco Pacchioni[2], and Friedrich Esch[1] — [1]Technical University of Munich, Garching, Germany — [2]University of Milano-Bicocca, Milano, Italy — [3]Technical University of Denmark, Kgs. Lyngby, Denmark — [4]École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland

Oxide-supported metal nanoparticles and clusters are common catalysts for heterogeneous reactions such as exhaust gas treatment. By studying supported size-selected clusters, we disentangle the influence of cluster size and support on reaction mechanisms. In the $Pt_n/Fe_3O_4(001)$ model system, we observe a complicated interplay between lattice oxygen reverse spillover, cluster encapsulation and gas phase pressure dependence. Sophisticated pulsed-valve reactor experiments (Sniffer-MS) combined with variable temperature scanning tunneling microscopy (STM) reveal the reaction of CO with lattice oxygen occurs on the cluster and not at the interface, but it is rapidly quenched by the strong metal-support interaction (SMSI). We will further demonstrate how UHV-based experiments can be quantified to provide turnover frequencies (TOFs) for comparison with real catalysts.

O 69.4 Thu 16:00 H4

A Model-Free Sparse Approximation Approach to Robust Formal Reaction Kinetics — •Frederic Felsen, Karsten Reuter, and Christoph Scheurer — Fritz-Haber-Institut der MPG, Berlin, Germany

Accurate and transferable models of reaction kinetics are of key importance for chemical reactors on both laboratory and industrial scale. Usually, setting up such models requires a detailed mechanistic understanding of the reaction process and its interplay with the reactor setup. We present a data-driven approach which analyzes the influence of process parameters on the reaction rate to identify effective rate laws without prior knowledge and assumptions. The algorithm we propose determines relevant model terms from a polynomial ansatz employing well established statistical methods. For the optimization of the model parameters special emphasis is put on the robustness of the results by taking not only the quality of the fit but also the distribution of errors [1] into account in a multi-objective optimization [2]. We demonstrate the flexibility of this approach based on synthetic kinetic data sets from microkinetic models. This way, we show that the kinetics of both the classical HBr reaction and a prototypical catalytic cycle are automatically reproduced. Further, combining our approach with experimental screening designs we illustrate how to efficiently explore kinetic regimes by using the example of the catalytic oxidation of CO.

[1] J. J. Filliben, *Technometrics*, 17, 111, **1975**.

[2] K. Deb et al., *IEEE Trans. Evol. Comput.*, 6, 182, **2002**.

O 69.5 Thu 16:15 H4

Combining Planar Laser-Induced Fluorescence with Stagnation Point Flows for Small Single-Crystal Model Catalysts: CO Oxidation on a Pd(100) — Jianfeng Zhou[1], •Sebastian Matera[2,4], Sebastian Pfaff[1], Sara Blomberg[1,3], Edvin Lundgren[1], and Johan Zetterberg[1] — [1]Lund University, SE-22100 Lund, Sweden — [2]Freie Universität Berlin, D-14195 Berlin, Germany — [3]Lawrence Berkeley National Laboratory, Berkeley, CA 94720-8229, USA — [4]Fritz-Haber-Institut der MPG, D-14195 Berlin, Germany

Mass transfer limitations can have a tremendous impact on catalysts characterization and must be accounted for by an appropriate modelling and, if possible, reactor design. We present a stagnation flow reactor for reaction product imaging by planar laser-induced fluorescence (PLIF), which is amenable to efficient low order modeling. Using CO oxidation over a Pd(100) single crystal as a showcase, we discuss the peculiarities for the case of small single-crystal model catalysts. While the ideal stagnation flow equations are not valid in this limit, a slightly modified theory can be derived, which exploits the information encoded in the PLIF signal. This combination of PLIF and half-theory/half-data driven modelling allows to efficiently analyze the experimental data and to estimate the turnover frequency and the CO_2, CO and O_2 concentrations at the surface from solely the CO_2 profile at some distance of the surface.

O 69.6 Thu 16:30 H4

On-surface collision reactions — •Matthew J. Timm, Kelvin Anggara, Lydie Leung, Zhixin Hu, and John C. Polanyi — Lash Miller Chemical Laboratories, Department of Chemistry, University of Toronto, 80 St. George Street, Toronto, Ontario M5S 3H6, Canada

Collisions between atoms and molecules are required for forming chemical bonds, thus they are central to any chemical reaction. The outcome of these collisions depends on the collision energy, geometry, and miss-distance between centers of the colliding reagents (called the impact parameter). As the incoming species - in general - randomly misses the target's center of mass, measurement of the impact parameter is a long-standing problem. Recently, a 'surface-molecular-beam' approach has been demonstrated that can allow for selection of impact parameter in a surface-reaction [1-3]. Energetic, oppositely-recoiling CF2 or F-atom "projectiles" are formed on a Cu(110) surface by dissociation of chemisorbed CF3 molecules with the tip of a Scanning Tunneling Microscope (STM). The inherent corrugation of Cu(110) leads to collimated trajectories of these projectiles, allowing them be aimed to collide with nearby molecular "targets" at chosen impact parameters. The pattern of reactive and non-reactive scattering was then determined by STM with the dynamics of the collision elucidated by density functional theory calculations.

[1] Anggara, K.; Leung, L.; Timm, M. J.; Hu, Z.; Polanyi, J. C.; Sci Adv., 2018, 4, eaau2821. [2] Anggara, K.; Leung, L.; Timm, M. J.; Hu, Z.; Polanyi, J. C.; Faraday Discuss., 2019, 214, 89-103. [3] Leung, L.; Timm, M. J.; Polanyi, J. C.; Chem. Commun., 2021, 57, 12647-12650.

O 69.7 Thu 16:45 H4

Faster oxygen adatom diffusion in a more densely packed CO adlayer on Ru(0001): A high-speed STM and DFT study — •Hannah Illner[1], Sung Sakong[2], Axel Gross[2], and Joost Wintterlin[1] — [1]Ludwig-Maximilians-Universität München, Germany — [2]Universität Ulm, Germany

Oxygen atoms on a Ru(0001) surface covered with 0.33 monolayers (ML) of coadsorbed CO travel through the CO layer by the so-called door opening mechanism. It is facilitated by structural fluctuations in the CO layer and leads to an enhanced diffusion constant [Henß et al., Science 363, 715 (2019)]. Expecting a lower diffusion constant on a more densely CO-covered surface we have investigated how this mechanism changes when the CO coverage is enhanced. The experiments were performed by means of a variable-temperature, high-speed STM. In the analyzed temperature range between 239 and 280 K the CO layer is disordered at a coverage of 0.47 ML. The obtained trajectories of the O atoms show that jumps occur in six equivalent directions with the same probability, in contrast to 0.33 ML, where three directions are preferred. Surprisingly, the Arrhenius plot of the hopping rates suggests a lower activation energy for the diffusion at the higher CO coverage than at 0.33 ML of CO. Density functional theory calculations suggest that at $\theta(\mathrm{CO}) > 0.33$ ML clusters form at which the CO density is locally enhanced, and that the configurations of the O atoms with respect to CO molecules are modified. However, the door opening mechanism is still efficient. A weaker binding of the O atoms in the dense CO layer effectively leads to a lower diffusion barrier.

O 69.8 Thu 17:00 H4

Machine-learning Gaussian Approximation Potentials to solve a longstanding puzzle about RuO_2 surfaces — •Yonghyuk Lee, Jakob Timmermann, Christoph Scheurer, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin, Germany

Machine-learning Gaussian Approximation Potentials (GAPs) have recently evolved as a powerful class of surrogate models to computationally demanding first-principles calculations. Along with structure exploration techniques, they enable us to examine the potential energy surface of interest with a hitherto unforeseen combination of physical accuracy and computational efficiency and to achieve global surface structure determination (SSD) for increasingly complex systems. This can be leveraged e.g. to discover novel surface motifs which are critical in understanding the "living" state of heterogeneous catalysts and their degradation under dynamic operating conditions. In our preceding study, this versatility could be leveraged by a general and data-efficient iterative training protocol that allows for the on-the-fly generation of GAPs via the actual surface exploration process. The iterative refinement of GAPs identifies plenty of unknown low energy terminations of RuO_2 even within the restricted sub-space of (1×1) surface unit-cells. Moreover, by extending the protocol to larger surface unit-cells, we discovered new surface structures, which provide solutions to longstanding questions in heterogeneous catalysis.

[1] J. Timmermann *et al.*, Phys. Rev. Lett. **125**, 206101 (2020)

[2] J. Timmermann *et al.*, J. Chem. Phys., **155**, 244107 (2021)

O 69.9 Thu 17:15 H4

Hydrogen cleaning induced surface changes of GaAs(110) — •Dorothee S. Rosenzweig[1], Moritz N.L. Hansemann[1], Philipp Ebert[2], Michael Schnedler[2], Holger Eisele[1], and Mario Dähne[1] — [1]Institut für Festkörperphysik, Technische Universität Berlin, 10623 Berlin, Germany — [2]Forschungszentrum Jülich GmbH, Peter Grünberg Institut, 52425 Jülich, Germany

For the nanoscopic analysis of III/V nanowire (110) surfaces, hydrogen cleaning is a commonly used procedure. While hydrogen cleaning is reported to be destruction free [1] and to achieve clean, atomically flat surfaces—as they are present directly after growth—the actual processes and dynamics during cleaning are rarely examined. However, a detailed understanding of these issues is crucial for the interpretation of electronic surface properties, of the growth of Nanaowires, as well as of built-in and distribution of dopands.

Here, we investigate the modifications of GaAs(110) as model system upon atomic hydrogen exposure at room temperature and under commonly used cleaning conditions at the atomic level. For depiction and measurement at the atomic scale, we used scanning tunneling microscopy and spectroscopy under UHV conditions. Using these methods we study the geometric arrangement of the adsorbed atoms as well as adsorbation induced additional electronic states, band bending, defect states, and Fermi level pinning.

[1] Webb et al., Nano Lett. 15, 8, 4865-4875 (2015)

O 69.10 Thu 17:30 H4

In-situ characterization of cyclic reduction and reoxidation of CeOx(111) and CeOx(100) islands on Cu(111) — •Linus Pleines[1], Lars Buss[2], Tevfik Onur Menteş[3], Francesca Genuzio[3], Andrea Locatelli[3], Jens Falta[1,4], and Jan Ingo Flege[2] — [1]Institute of Solid State Physics, University of Bremen, Germany — [2]Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus-Senftenberg, Germany — [3]Elettra-Sincrotrone Trieste S.C.p.A., Basovizza Trieste, Italy — [4]MAPEX Center for Materials and Processes, Bremen, Germany

Cerium oxide (CeO_x) is of special interest due to its catalytic activity and various other electronic and optical applications. The inverse model catalyst CeO_x on Cu(111) has a high activity for methanol synthesis from H_2 and CO_2. For the activation of CO_2, Ce^{3+} sites have to be present at the surface, which means that the CeO_x has to be reduced to some extent. This may be achieved by exposure to H_2 at elevated temperatures. We studied the interaction of H_2 and CO_2 with CeO_x islands on Cu(111) with low-energy electron microscopy (LEEM) and X-ray absorption spectroscopy (XAS). From earlier studies, the orientation of the CeO_x is known to be decisive for its catalytic activity. In our experiments (100) and (111) CeO_x islands are grown side by side on the metal substrate, so that identical reaction conditions prevail during the experiment. At a high temperature of 550 °C, exposure to H_2 leads to partial reduction, and exposure to CO_2 leads to reoxidation of the CeO_x. The differences observed for the two island orientations regarding structure and composition will be discussed.

O 70: Supported nanoclusters: Structure, Reactions, Catalysis

Time: Thursday 15:00–17:45 Location: H6

Topical Talk O 70.1 Thu 15:00 H6

Stability and dynamics of cluster catalysts and their supports — Sebastian Kaiser, Johanna Plansky, Fabian Knoller, Alexander Bourgund, Ke Zhang, Ueli Heiz, Friedrich Esch, and •Barbara A. J. Lechner — Department of Chemistry & Catalysis Research Center, Technical University of Munich, Germany

The intrinsic metastability of supported clusters can induce a vast range of dynamics that strongly influence their physical and chemical properties, while being experimentally highly challenging to investigate. Here, I will present a range of surface dynamics that occur in supported cluster dynamics, ranging from confined cluster diffusion, cluster encapsulation, support mobility and reactant spillover to lateral diffusion linked to reactivity. Our experimental approach is to combine static, statistically sound, and dynamic, time-resolved scanning tunneling microscopy (STM) to investigate the diffusion, sintering, and restructuring of size-selected clusters on weakly and strongly interacting supports.

O 70.2 Thu 15:30 H6

The role of water in oxidation of the Pt/Co3O4 interface — •Yaroslava Lykhach[1], Lukáš Fusek[1,2], Maximilian Kastenmeier[1], Tomáš Skála[2], Nataliya Tsud[2], Viktor Johánek[2], Sascha Mehl[3], Josef Mysliveček[2], Olaf Brummel[1], and Jörg Libuda[1] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany — [2]Charles University, Prague, Czech Republic — [3]Elettra-Sincrotrone Trieste SCpA, Basovizza-Trieste, Italy

Electronic metal-oxide interactions (EMSI) play a major role in the design of advanced functional materials for applications in catalysis. We investigated the influence of the EMSI on the oxidation state of ultra-small Pt particles supported on a well-ordered Co3O4(111) substrate in the presence of co-adsorbates, i.e. hydroxyl groups and molecularly adsorbed water, by means of synchrotron radiation photoelectron spectroscopy and scanning tunnelling microscopy. The EMSI gives rise to charge transfer across the metal-oxide interface and results in partial oxidation of Pt deposits coupled with partial reduction of Co3O4(111). We detected ultra-small Ptδ+ aggregates in combination with atomically dispersed Pt2+/4+ species. While the oxidation degree of Pt deposits is not influenced by the presence of co-adsorbates, the magnitude of the charge transfer is enhanced in the presence of molecularly adsorbed water. Subsequent annealing in UHV leads to re-oxidation of Co3O4(111) accompanied by an increase in the amount of Pt4+ species. This observation suggests the re-dispersion of Ptδ+ aggregates to Pt4+ species triggered by the dissociation of water.

O 70.3 Thu 15:45 H6

Ripening mechanism changes with cluster size: *In situ* observation of Pt cluster diffusion on $Fe_3O_4(001)$ — •Sebastian Kaiser, Johanna Plansky, Ueli Heiz, Barbara A. J. Lechner, and Friedrich Esch — Technical University of Munich, Garching, Germany

Ripening of small oxide-supported metal clusters is a common deactivation mechanism in heterogeneous catalysis. We use scanning tunneling microscopy (STM) to follow the diffusion and ripening of size-selected Pt clusters on an $Fe_3O_4(001)$ support. Thanks to a strong cluster-support bonding, ripening and coalescence only set in at elevated temperatures. Particle size analysis of our STM images reveals that Pt_{19} exhibits Ostwald ripening at temperatures above 800 K, i.e. with cluster growth by atom diffusion. In contrast, the smaller Pt_5 and Pt_{10} clusters show Smoluchowski ripening, i.e. the diffusion of entire clusters, already in the temperature range of 600 to 800 K and Ostwald ripening >800 K. We not only observe the ripened clusters in STM, but could successfully follow the diffusion process *in situ* by STM movies. Surprisingly, temperature programmed desorption (TPD) measurements of CO molecules show that in this temperature range the clusters get concomitantly encapsulated by iron oxide via strong metal support interaction (SMSI). Cluster diffusion thus occurs despite a strong interaction with the magnetite support and the diffusing species is most likely a cluster with Fe and O atoms on top.

O 70.4 Thu 16:00 H6

Tuning SMSI Kinetics on Pt-loaded $TiO_2(110)$ by Choosing the Pressure — •Philip Petzoldt[1], Moritz Eder[1], Sonia Mackewicz[1], Monika Blum[2,3], Tim Kratky[4], Sebastian Günther[4], Martin Tschurl[1], Ueli Heiz[1], and Barbara Lechner[5] — [1]Physical Chemistry, Department of Chemistry and Catalysis Research Center, Technical University of Munich — [2]Advanced Light Source, Lawrence Berkeley National Laboratory — [3]Chemical Sciences Division, Lawrence Berkeley National Laboratory — [4]Physical Chemistry with Focus on Catalysis, Department of Chemistry and Catalysis Research Center, Technical University of Munich — [5]Functional Nanomaterials, Department of Chemistry and Catalysis Research Center, Technical University of Munich

The encapsulation of noble metal particles on reducible supports due to a strong metal-support interaction (SMSI) has already been extensively studied. However, there is still an ongoing debate on important aspects such as the influence of oxygen or hydrogen treatments on the encapsulating overlayer. We have utilized synchrotron-based NAP-XPS in order to investigate the SMSI for Pt-loaded $TiO_2(110)$ single crystals under the influence of H_2 and O_2 at different pressures. In an O_2 atmosphere two different, pressure-dependent phenomena, namely, an oxidation of Pt particles and a loss of Pt signal intensity, are observed at 800 K. While the oxidation is partially reversed in vacuum, the intensity of the platinum signal cannot be recovered. H_2 annealing has no significant additional effect compared to vacuum annealing. In the presentation, we discuss possible origins of these observations.

O 70.5 Thu 16:15 H6

Deposition and annealing of FeNi nanoparticles on surfaces — •Mahboobeh Ravankhah[1], Mathias Getzlaff[1], Gerhard Dehm[2], and Philipp Watermeyer[2] — [1]Institut für Angewandte Physik, Universität Düsseldorf — [2]Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf

3d bimetallic nanoparticles have received lots of attention due to their technological applications in ultrahigh density information storage, catalysis and biomedicine. The thermal stability and the magnetic properties of the in-vacuo prepared bimetallic nanoparticles are shown to depend on the composition and their structure. Here we report on new findings of structure and composition of Fe-Ni nanoparticles, synthesized via a magnetron sputtering source and deposited on a Tungsten crystal surface. The elemental distribution of nanoparticles is determined by high resolution transmission electron microscopy (HRTEM) and electron energy loss spectroscopy (EELS). It is found that the nanoparticles have a shell formed by Fe atoms and a core composed of Fe and Ni with the gradient of composition from core to the surface. The melting behavior of nanoparticles was studied under UHV conditions by scanning tunneling microscopy (STM) as a function of heating temperature. The unrolling carpet, surface diffusion and anisotropy spreading are driving processes to form monolayer high islands above the melting point. The relevant result could be helpful for the design and preparation of stable and controllable bimetallic nanoparticles for technological applications.

O 70.6 Thu 16:30 H6

Reaction Pathways in Alcohol Photoreforming on Cluster Co-Catalyst Loaded $TiO_2(110)$ — •Sonia Mackewicz, Moritz Eder, Philip Petzoldt, Martin Tschurl, and Ueli Heiz — Physical Chemistry, Department of Chemistry and Catalysis Research Center, Technical University of Munich

Heterogeneous photocatalysis offers the prospect of utilizing solar energy for the environmentally benign production of chemical fuels such as hydrogen. State-of-the-art materials often comprise co-catalyst loaded semiconductors, but these systems are still limited in efficiency.

In order to systematically optimize photocatalysts, a profound knowledge of reaction mechanisms is crucial, whose details are so far little understood. Alcohols are an ideal model system to investigate reaction mechanisms in photocatalysis. Furthermore, they may serve as renewable sources for hydrogen in the future. In this talk, we give insights into the mechanistics of the alcohol photoreforming reaction on co-catalyst loaded $TiO_2(110)$. It is shown that semiconductor photocatalysis selectively enables new reaction pathways, which are not accessible by thermal or conventional chemical methods.

O 70.7 Thu 16:45 H6

Electron transfer reaction by time resolved (TRS) and core level spectroscopy (XPS) and STM on Au/TiO2(110) single crystal systems. — •Hicham Idriss — Institute of functional Interfaces, KIT, Karlsruhe

Charge transfer from or to a metal deposited on an oxide semiconductor are central to photocatalysis. In order to probe into this phenomenon, the effect of gold coverage on the chemical state of Ti cations, upon photoexcitation of rutile TiO2(110) single crystal, was investigated by X-ray photoelectron spectroscopy (XPS). Photocatalytic reaction of gas phase ethanol (a hole scavenger) on TiO2(110) and Aux/TiO2(110) resulted in the formation of Ti3+ cations. Increasing the Au coverage resulted in the gradual decrease of these Ti3+ cations. The "quasi" total consumption of these reduced states was found at a ratio Au atoms to reacted Ti3+ close to one; this corresponded to about 0.50 at. % of Au. The relationship suggests that electron transfer occurs from the excited semiconductor to Au atoms during the catalytic reaction. In order to complement the work H2 production rates of an electron donor, such as ethanol, over Au clusters with different sizes and coverage deposited on single crystal rutile TiO2(110) were studied by scanning tunneling microscopy, online mass spectrometry and complemented by femto second pump probe spectroscopy. It was also found that there is a non-linear increase of the H2 production rate with increasing gold coverage. The key determining factor appears to be the Au inter-particle distance. Increasing this distance resulted in an increase in the normalized reaction rate.

O 70.8 Thu 17:00 H6

Surface Ligand Infrared Spectroscopy: In-Situ Characterization of Noble Metal Clusters and Metal Oxides at Work — •Eric Sauter[1], Daria Gashnikova[2], Florian Maurer[2], Alexei Nefedov[1], Stefan Heissler[1], Yuemin Wang[1], Jan-Dirk Grunwald[2], and Christof Wöll[1] — [1]Intitute of Functional Interfaces, KIT, Eggenstein-Leopoldshafen, Germany — [2]Institute of Chemical Technology and Polymer Chemistry, KIT, Karlsruhe, Germany

To achieve a full understanding of chemical processes at exposed surfaces in-situ and operando investigations are required. For studies of catalytic processes under real conditions, IR spectroscopy offers a number of advantages. In the present study, surface ligand infrared spectroscopy was used to perform an in-situ investigation of the surface characteristics of cerium oxide single crystals as well as catalytic active NM-clusters on cerium oxide nanoparticles. Low temperature adsorption of carbon monoxide was used to identify the surface structure and morphology, visible through distinctive adsorption bands blue shifted in respect to the gas phase, which can be used as reference for the interpretation of more complicated spectra like powders or nanoparticles. Additionally, noble-metal clusters were investigated in pristine condition as well as in the reduced state. Upon heating, desorption of the probe molecule occurred and at higher temperatures deformation and sintering of the clusters was observed. The investigation shows the power of infrared spectroscopy as a tool for in-situ investigations and characterization of NM-clusters and metal oxides at work.

O 70.9 Thu 17:15 H6

AI with Experimental and Theoretical Data toward the Understanding CO_2 Hydrogenation Catalysis: The Role of the Support Materials — •Ray Miyazaki[1], Kendra Belthle[2], Harun Tüysüz[2], Lucas Foppa[1], and Matthias Scheffler[1] — [1]The NOMAD Laboratory at the Fritz Haber Institute of the Max Planck Society, Germany — [2]Max-Planck-Institut für Kohlenforschung, Germany

The genesis of organic molecules from CO_2 at a hydrothermal vent, which is a fissure on the seafloor, is one of the theories for the origin of life [1]. We focus on CO_2 hydrogenation catalyzed by cobalt nanoparticles supported on SiO_2, which mimic the environment of a hydrothermal vent. In particular, we investigate the role of support materials by using several amorphous SiO_2 supports incorporating different elements (e.g., Ti, Zr). In this study, the experimental selectivity toward organic molecules (e.g., methanol, formic acid) is modeled by the sure-independence screening and sparsifying operator (SISSO) AI approach [2]. Both experimental and theoretical data are adopted as the input features for SISSO, such as atomic-scale features calculated by density functional theory and experimental characterization data. Our approach identifies the key descriptive parameters correlated to the selectivity, which lead to a better understanding of the origin of life and to design of novel CO_2 hydrogenation catalysts.
[1] M. Preiner *et al.*, *Nat. Ecol. Evol.*, **4**, 534-542 (2020).
[2] R. Ouyang *et al.*, *Phys. Rev. Mater.*, **2**, 083802 (2018).

O 70.10 Thu 17:30 H6

Near-ambient pressure studies of size selected clusters on ultrathin silica films — •Matthias Krinninger, Florian Kraushofer, Friedrich Esch, and Barbara A.J. Lechner — Department of Chemistry, Technical University of Munich, 85748 Garching, Germany

Silicon oxide is a widely used catalyst support material for clusters and nanoparticles. Understanding the relationship between these clusters and the support is challenging, however, because SiO_2 is insulating, and in most applications not crystalline, which limits the use of diffraction-based experimental techniques. Some progress has been made by growing ultrathin, quasi-2D silica bilayer films on a variety of metal supports [1], which can then be measured by scanning tunneling microscopy (STM). Here, we show first results for ultrathin silica films grown on Pt(111) and their interaction with deposited metal clusters, examined by near-ambient pressure (NAP) XPS and NAP-STM. We investigate the stability of the films, their ability to stabilize small clusters without sintering, and the dependence of this stability on the crystallinity of the film.

[1] C. Büchner, M. Heyde, Two-dimensional silica opens new perspectives, Prog. Surf. Sci., 92 (2017) 341-374.

O 71: Focus Session: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy 2

Time: Thursday 15:00–18:00 Location: S051

Topical Talk O 71.1 Thu 15:00 S051

Theory for Electron Spin Resonance based on electron transport — •Nicolas Lorente[1], José Reina[2], and Christoph Wolf[2] — [1]Centro de Física de Materiales & DIPC, Donostia, Spain — [2]Center for Quantum Nano Science, Seoul, Korea

Recent progress in electron spin resonance with the scanning tunneling microscope (ESR-STM) [1] is greatly advancing the experimental possibilities of manipulating atomic spins by all-electrical means. Two-qubit operations have been made possible using a pulse-mode in the ESR-STM [2], and addressing remote qubits has been rendered possible by creating a new muti-frequency operational mode [3]. We aim at developing a computational tool that permits us to interpret and predict the outcome of experiments in ESR. The first results of such a simulation tool have addressed one and two spins under an STM current [4,5]. We use a non-equilibrium Green's function approach with Hubbard operators that allows us to write quantum adiabatic Markovian master equations in the presence of an electron current and under the driving of an external electric field. The results are enticing and the modelling is flexible enough to treat many different physical situations. References: [1] S. Baumann et al, Science 350, 417 (2015). [2] K. Yang et al, Science 366, 509 (2019). [3] S.-H. Phark et al, ArXiv:2108.09880. [4] J. Reina et al, Phys. Rev. B 100, 035411 (2019). [5] J. Reina et al, Phys, Rev. B 104, 245435 (2021).

O 71.2 Thu 15:30 S051

Modeling the Electron Spin Resonance Spectrum in Scanning Tunneling Microscopy — •Christian R. Ast[1], Piot Kot[1], Maneesha Ismail[1], and Juan Carlos Cuevas[2] — [1]MPI for Solid State Research, 70569 Stuttgart — [2]Universidad Autónoma de Madrid, 28049 Madrid, Spain

The theory of electron spin resonance (ESR) spectroscopy in scanning tunneling microscopy (STM) has been debated for some time now with a number of different proposals having different origin, but essentially leading to very similar results. While the focus so far has been on the ESR signal itself, the measured DC tunneling spectrum offers more details that allow for a more precise verification of the underlying theory. Here, we discuss the ESR signal from a theory point of view by allowing the tunneling electrons to interact with both the driven spin system and the incident microwave during the tunneling process. We find a more complete description of the whole tunneling current also going beyond the typical approximation of a constant density of states.

O 71.3 Thu 15:45 S051

A new view on the origin of zero-bias anomalies of Co atoms atop noble metal surfaces — Juba Bouaziz[1], Filipe S. M. Guimaraes[1], and •Samir Lounis[1,2] — [1]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich & JARA, Jülich 52425, Germany — [2]Faculty of Physics & CENIDE, University of Duisburg-Essen, 47057, Duisburg, Germany

Many-body phenomena are paramount in physics. In condensed matter, their hallmark is considerable on a wide range of material characteristics spanning electronic, magnetic, thermodynamic and transport properties. In this talk, we address systematically zero-bias anomalies detected by scanning tunneling spectroscopy on Co atoms deposited on Cu, Ag and Au(111) substrates, which remarkably are almost identical to those obtained from first-principles [1]. These features originate from gaped spin-excitations induced by a finite magnetic anisotropy energy, in contrast to the usual widespread interpretation relating them to Kondo resonances. Resting on relativistic time-dependent density functional and many-body perturbation theories, we furthermore unveil a new many-body feature, the spinaron, resulting from the interaction of electrons and spin-excitations localizing electronic states. Besides Co, we will show examples of anomalous spin-excitations characterising adatoms on Nb(110) surface [2,3].

[1] Bouaziz, Guimaraes, Lounis, Nat. Commun. 11, 6112 (2020); [2] Brinker, Küster, Parkin, Sessi, Lounis, Science Adv. 8, eabi7291 (2022); [3] Küster, Montero, Guimaraes, Brinker, Lounis, Parkin, Sessi, Nat. Commun. 12, 1108 (2021).

O 71.4 Thu 16:00 S051

Real-space observation of the Kondo effect in MoS_2 mirror twin boundaries — Camiel van Efferen[1], Jeison Fischer[1], Achim Rosch[2], Thomas Michely[1], and •Wouter Jolie[1] — [1]II. Physikalisches Institut, Universität zu Köln — [2]Institut für Theoretische Physik, Universität zu Köln

Finite mirror twin boundaries in monolayer MoS_2 on graphene confine strongly correlated one-dimensional electronic states [1]. Using scanning tunneling microscopy and spectroscopy, we observe a resonance at the Fermi energy when the highest occupied confined state is filled with one electron. Magnetic field and temperature-dependence of the resonance unambiguously point to the Kondo effect, i.e., screening of the spin-$\frac{1}{2}$ confined state within the mirror twin boundary. Theoretical models for both Kondo resonance and spin-$\frac{1}{2}$ state are used to extract the Kondo coupling strength. Real-space mapping gives access to the correlated beating of both confined state and Kondo resonance along the boundary. Hence, our experiments reveal the behavior of the Kondo effect for a delocalized electronic state on the atomic scale.

[1] Jolie et al., Phys. Rev. X 9, 011055 (2019)

O 71.5 Thu 16:15 S051

Moiré tuning of spin excitations: individual Fe atoms on MoS_2/Au(111) — •Christian Lotze[1], Sergey Trishin[1], Nils Bogdanoff[1], Felix von Oppen[2], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany — [2]Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany

Magnetic adatoms have been investigated on various surfaces in regard to stabilizing, controlling and manipulating single quantum spins. Here, we study individual iron atoms adsorbed on a single layer of molybdenum disulfide (MoS_2) on a Au(111) crystal. MoS_2 has been recently reported as a well-suited system for decoupling molecules. We show that the Fe atoms are largely decoupled from the Au(111) substrate with the remaining coupling strength varying along the moiré structure. As a consequence, the spectroscopic fingerprints range from pure inelastic excitations to Kondo resonances. Moreover, we see spatial variations of those excitations over one atom, which result from the formation of Fe-S hybrid states and interference effects. In conclusion, our work establishes MoS_2 on Au(111) as a tuning layer for quantum spin properties. This tuning can be realized continuously.

O 71.6 Thu 16:30 S051

Spin excitations on hexagonal zinc oxide — •Lukas Arnhold, Henrik Lichtl, Leon Rullkötter, Nicolaj Betz, Susanne Baumann, and Sebastian Loth — University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany

Few-layer materials are widely used to tailor different electronic properties, down to the atomic level. We use a double layer of ZnO, a hexagonal wide bandgap semiconductor [1], to mitigate electron scattering between the Ag (111) substrate and Co atoms deposited on the ZnO surface. With low-temperature scanning tunneling microscopy we observe spin excitations and the ability to manipulate transition metal atoms on the surface into hexagonal arrangements. These findings make ZnO a viable candidate for resonant spin spectroscopy methods [2,3] and construction of geometrically frustrated magnetic structures.

[1] A. Shiotari et al., J. Phys. Chem. C 118, 27428 (2014). [2] S. Baumann et al., SCIENCE 350, 417 (2015). [3] M. Hänze et al., SCIENCE ADVANCES 7, eabg2616 (2021).

Topical Talk O 71.7 Thu 16:45 S051

Stochastic resonance as a new tool to investigate spin dynamics — •Susanne Baumann[1], Nicolaj Betz[1], Max Hänze[1], Gregory McMurtrie[1], Susan Coppersmith[2], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]School of Physics, University of New South Wales, Sydney, Australia

Stochastic resonance is an unusual phenomenon in which noise can be used as a resource to synchronize stochastic dynamics to a control signal [1,2]. In this talk, I will show how stochastic resonance can be induced in the spin switching of magnetic nanostructures on surfaces [3], and, more importantly, how we can use this as a tool to investigate the magnetization dynamics of these spin systems.

With this tool, one can get insight into the interaction of these structures with their environment. This also enables the observation of ultrafast dynamics of excited spin states that are not easily accessible to other scanning probe techniques. The new frequency resolved spectroscopy method allows for the broadband observation of spin dynamics with previously inaccessible bandwidth ranging from milliseconds to picoseconds.

References: [1] R. Benzi, J. Phys. A: Math. Gen 14, L453 (1981). [2] R. Löfstedt, S. N. Coppersmith, Phys Rev. Lett. 72, 1947 (1994). [3] M. Hänze*, G. McMurtrie* et al. Science Adv. 7 eabg2616 (2021).

O 71.8 Thu 17:15 S051

Path-resolved measurement of ultrafast spin dynamics — •Nicolaj Betz[1], Max Hänze[1,2], Gregory McMurtrie[1], Susanne Baumann[1], and Sebastian Loth[1,2] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Max Planck Institute for Solid State Research, Stuttgart, Germany

Transitions between quantum mechanical states are fundamentally random processes. While it is possible to directly observe individual quantum jumps [1] in a time resolved measurement [2], the dynamics of many systems exceed the resolution of real time measurements. This requires the use of time-averaged measurements such as pump probe experiments. These methods typically measure state occupation times and contain little information about the relaxation process itself. Here, we introduce a dynamic response measurement that is sensitive to the switching path between spin states and can be applied in scanning tunneling microscopy. By using stochastic resonance [3], this method resolves spin- switching dynamics of magnetic atoms and nanostructures ranging from milliseconds to picoseconds. Crucially, in more complex spin structures the measurement can distinguish multiple switching paths between higher excited states. This provides deeper insight into ultrafast spin dynamics than possible with relaxometry.

[1] Th. Sauter, et al. Phys. Rev. Lett. 57, 1696 (1986).

[2] M. Hänze, et al. Sci. Adv. 7, 33 (2021).

[3] R. Löfstedt, et al. Phys. Rev. Lett. 72, 1947 (1994).

O 71.9 Thu 17:30 S051

Growth and magnetic characterization of thermally robust cobalt islands on Cu3Au(111) — •Aleš Cahlík, Danyang Liu, Berk Zengin, and Fabian Natterer — Institute of Physics, UZH, Zurich, Switzerland

Due to a larger tunability of the effective lattice parameter, bimetallic alloys can be an appealing choice as an alternative substrate for the growth of thin films and nanostructures. In this respect, we investigate $Cu_3Au(111)$ as a platform for the growth of cobalt nano-islands. Using STM, we demonstrate unique thermal stability of Co/$Cu_3Au(111)$ up to $\sim 340°C$, compared to the fast intermixing of Co/Cu(111) at room temperature. We explore the structural and magnetic properties of the Co islands with spin-polarized and nickelocene functionalized tips. Finally, we find an effective method to produce spin-polarized tips by deliberately lifting off an entire island from the substrate and transferring it to the STM tip.

O 71.10 Thu 17:45 S051

Transport in the Rashba-split surface state of $(\sqrt{3}\times\sqrt{3})$Bi/Ag(111)$R30°$ revealed by MONA — •Markus Leisegang, Patrick Härtl, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

Transport measurements that are sensitive to the band structure of a material require techniques that operate on the length scale of the charge carrier's mean free path. A novel method that fulfills this requirement is the molecular nanoprobe (MONA), which uses a single molecule to detect charge carriers [1].

In this study, we investigate the rotation and tautomerization of phthalocyanine molecules on the $(\sqrt{3}\times\sqrt{3})$Bi/Ag(111)$R30°$ surface and utilize these excitations to investigate transport in the Rashba-split surface state characteristic for this surface [2]. We find that both excitation processes are driven by the N-H stretching mode and can be triggered by a single electron [3]. Our transport measurements proof the sensitivity to hot charge carriers which preferably propagate in the Rashba-split surface state of the $BiAg_2$ alloy. The expected impact of the spin–momentum-locking of this Rashba-split surface state on the surface transport is discussed and first experimental results obtained with spin-polarized tips will be presented.

[1] M. Leisegang *et al.*, Nano Lett. **18**, 2165–2171 (2018)
[2] C. R. Ast *et al.*, Phys. Rev. Lett. **98**, 186807 (2007)
[3] J. Kügel *et al.*, Journ. Phys. Chem. C **121**, 28204–28210 (2017)

O 72: 2D Materials 3: hBN and Electronic Structure

Time: Thursday 15:00–17:00 Location: S052

O 72.1 Thu 15:00 S052

Electronic Structure of Two-Dimensional CoO2 — Ann Julie U. Holt[1], Sahar Pakdel[1], Jonathan Rodríguez-Fernández[2], Yu Zhang[3], Davide Curcio[1], Zhaozong Sun[4], Paolo Lacovig[5], Yong-Xin Yao[6,7], Jeppe V. Lauritsen[4], Silvano Lizzit[5], Nicola Lanatà[1,8], Philip Hofmann[1], Marco Bianchi[1], and •Charlotte E. Sanders[3] — [1]Dept. of Physics and Astronomy, Interdisciplinary Nanoscience Center (iNANO), Aarhus Univ., 8000 Aarhus, DK — [2]Dept. of Physics, Univ. of Oviedo, Oviedo 33007 ES — [3]UK Central Laser Facility, RCaH, STFC RAL, Didcot, Oxfordshire OX11 0QX, UK — [4]Interdisciplinary Nanoscience Center (iNANO), Aarhus Univ., 8000 Aarhus, DK — [5]Elettra Sincrotrone Trieste S.C.p.A., AREA Science Park, Strada Statale 14, km 163.5, 34149 Trieste, IT — [6]Ames Laboratory U.S.-DOE, Ames, IA 50011, USA — [7]Dept. of Physics and Astronomy, Iowa State Univ., Ames, IA 50011, USA — [8]Nordita, KTH Royal Institute of Technology and Stockholm Univ., Roslagstullsbacken 23, 10691 Stockholm, SE

The transition metal oxide CoO2 forms bulk layered structures that exhibit complex correlated electronic states. However, little has been known about the electronic properties of the isolated single layer. We have now [1] studied CoO2/Au(111), using angle-resolved photoemission spectroscopy, x-ray photoelectron diffraction, and density functional theory. The results of our study show single-layer CoO2 to be metallic, with electronic correlations. They emphasize the interest of oxides as a new subject within two-dimensional materials research. [1] 2D Mater. 8 (2021) 035050.

O 72.2 Thu 15:15 S052

Unified Treatment of Magnons and Excitons in Monolayer CrI3 from Many-Body Perturbation Theory — •Thomas Olsen — Technical University of Denmark

We present first principles calculations of the two-particle excitation spectrum of CrI3 using many-body perturbation theory including spin-orbit coupling. Specifically, we solve the Bethe-Salpeter equation, which is equivalent to summing up all ladder diagrams with static screening, and it is shown that excitons as well as magnons can be extracted seamlessly from the calculations. The resulting optical absorption spectrum as well as the magnon dispersion agree very well with recent measurements, and we extract the amplitude for optical excitation of magnons resulting from spin-orbit interactions. Importantly, the results do not rely on any assumptions of the microscopic magnetic interactions such as Dzyaloshinskii-Moriya (DM), Kitaev, or biquadratic interactions, and we obtain a model independent estimate of the gap between acoustic and optical magnons of 0.3 meV. In addition, we resolve the magnon wave function in terms of band transitions and show that the magnon carries a spin that is significantly smaller than $\hbar$. This highlights the importance of terms that do not commute with S_z in any Heisenberg model description.

[1] T. Olsen, Phys. Rev. Lett. 127, 166402, (2021)

O 72.3 Thu 15:30 S052

Excitons in two-dimensional magnetic semiconductors — •Marie-Christin Heissenbüttel, Thorsten Deilmann, Peter Krüger, and Michael Rohlfing — Institute of Solid State Theory, University of Münster, Germany

Semiconducting two-dimensional magnets exhibit peculiar interrelation between magnetic properties and light-matter interaction. We will present and discuss ab-initio *GW*/Bethe-Salpeter equation calculations to examine excitonic states in different magnetic systems.

Due to the hexagonal crystal structure the out-of-plane ferromagnetic monolayer CrI_3 came to the fore for the construction of heterobilayers with transition-metal dichalcogenides [1]. Due to coupling effects the magnetic properties are transferred and can be observed in a splitting and a modified Zeeman effect of the excitons. On the other hand, CrSBr shows in-plane magnetization in combination with a large crystal anisotropy. This is reflected in the quasi-1D behavior of different opto-electronic properties [2].

[1] Nano Lett. Lett. 21, 5173-5178 (2021)
[2] https://arxiv.org/abs/2205.13456

O 72.4 Thu 15:45 S052

Nanoscale view of massive Dirac quasiparticles in lithographic superstructures — •Alfred Jones[1], Lene Gammelgaard[2], Deepnaryan Biswas[1], Mikkel Sauer[3], Roland Koch[4], Chris Jozwiak[4], Eli Rotenberg[4], Aaron Bostwick[4], Kenji Watanabe[5], Takashi Taniguchi[5], Cory Dean[6], Thomas Pedersen[3], Antti-Pekka Jauho[2], Peter Bøggild[2], Bjarke Jessen[6], and Søren Ulstrup[1] — [1]Aarhus University, Denmark — [2]Technical University of Denmark, Denmark — [3]Aalborg University, Denmark — [4]Advanced Light Source, USA — [5]National Institute for Materials Science, Japan — [6]Columbia University, USA

Massive Dirac quasiparticles play a central role in a number of emerging physical phenomena such as topological phase transitions and anomalous Hall effects. Single-layer graphene appears to be an ideal platform to explore such properties, however engineering the transition from massless to massive Dirac quasiparticles in a controllable fashion remains a significant challenge. Here, we employ angle-resolved photoemission with a nanoscale light spot (nanoARPES) to directly measure the electronic structure modifications induced by lithographic patterning of an antidot superlattice onto a graphene device. We observe a transition from massless Dirac fermions in the pristine graphene to a massive character in patterned regions, and determine that the mass scales linearly with antidot diameter, consistent with theory. Gate-induced electron-doping of the patterned graphene produces an enhancement of the mass, highlighting the versatility of nanopatterned graphene as a platform for engineering such quasiparticles.

O 72.5 Thu 16:00 S052

Tuning lower dimensional superconductivity with hybridization at a superconducting-semiconducting interface — •Anand Kamlapure[1], Manuel Simonato[1], Emil Sierda[1], Manuel Steinbrecher[1], Umut Kamber[1], Elze J. Knol[1], Peter Krogstrup[2], Mikhail I. Katsnelson[1], Malte Rösner[1], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University, 6525 AJ Nijmegen, the Netherlands — [2]Center for Quantum Devices, Niels Bohr Institute, University of Copenhagen, 2100 Copenhagen, Denmark

Study of influence of interface electronic structure on the superconductivity (SC) in lower dimensions is important to tune SC in view of its applications to gated superconducting electronics, and superconducting layered heterostructures. Here, using ultra-low temperature scanning tunneling microscopy and spectroscopy, we demonstrate the formation of hybrid electronic structure at the interface between a lead film and black phosphorus. We show that interfacial hybridization weakly modifies the confinement potential and leads to a renormalization of the superconducting gap and a strong modification of the observed vortex structure. Using ab initio methods combined with analytical modeling, we link the renormalized gap to a weighting of the superconducting order parameter in reciprocal space. These results illustrate that interfacial hybridization can be used to tune SC in quantum technologies based on lower dimensional superconducting electronics. Reference: arXiv:2109.08498 (2021).

O 72.6 Thu 16:15 S052

Diversity of defect-related excitons in hBN from ab initio calculations — •Alexander Kirchhoff, Thorsten Deilmann, Peter Krüger, and Michael Rohlfing — Westfälische Wilhelms-Universität Münster, Institut für Festkörpertheorie, Wilhelm-Klemm-Straße 10, 48149 Münster

While pristine hexagonal boron nitride (hBN) is an insulator with an optical gap of ∼5 eV, point defects in this material are discussed as single-photon emitters in the visible optical spectrum. In this study, we examine different defects consisting of carbon or oxygen substitutions and vacancies in an hBN monolayer from an ab initio approach, via the GW/BSE approximation. Our results show deep defect states and defect-related excitations with energies in the visible regime. We present a detailed analysis of their structure and energetic composition and furthermore discuss the dependence of the excitonic spectrum on the geometry. Finally, we present a defect of two carbon substitutions adjacent to a divacancy, which shows an antiferromagnetic ground state.

O 72.7 Thu 16:30 S052

Engineering magnetic interactions in magnetic thin films with two-dimensional materials — •Hangyu Zhou[1,2], Manuel dos Santos Dias[1,3,4], Weisheng Zhao[2], and Samir Lounis[1,3] — [1]Peter Grünberg Institut and Institute for Advanced Simulations, Forschungszentrum Jülich & JARA, 52425 Jülich, Germany — [2]School of Integrated Circuit Science and Engineering, MIIT Key Laboratory of Spintronics, Beihang University, Beijing 100191, China — [3]Faculty of Physics, University of Duisburg-Essen and CENIDE, 47053 Duisburg, Germany — [4]Scientific Computing Department, STFC Daresbury Laboratory, Warrington WA4 4AD, United Kingdom

Two-dimensional (2D) materials have received great attention due to their unique physical and chemical properties and ease of integration in heterostructures, which can lead to improved magnetic properties. Here, we explore with density functional theory calculations the impact of monolayers of graphene and hexagonal boron nitride (h-BN) on the magnetism and structural properties of a Co monolayer placed on Pt(111) and Au(111) surfaces. In particular, we investigate how the magnetic interactions, such as the Heisenberg exchange interaction and the Dzyaloshinskii-Moriya interaction (DMI), are influenced by the 2D monolayer and by structural reconstructions, which in turn can be utilized to ignite complex spin-textures. These results may contribute to an enhanced tunability of skyrmion formation in such composite magnetic heterostructures.

O 72.8 Thu 16:45 S052

Threshold Energies for Defect Production in 2D Materials under Low Energy Ion bombardment: Insights from ab-initio Molecular Dynamics — •Silvan Kretschmer, Sadegh Ghaderzadeh, Stefan Facsko, and Arkady V. Krasheninnikov — Helmholtz-Zentrum Dresden-Rossendorf, Germany

Low energy ion implantation (LEII) provides a valuable tool to tune the mechanical, electronic and catalytic properties of 2D materials by the targeted implantation of impurities. In contrast to ion irradiation at higher energies the commonly applied binary collision formula fails to describe the outcome of the irradiation process for ions close to the displacement energy, that is the minimum ion energy needed to displace the target atom. The dominating influence of the chemical interaction of projectile and target atoms and its effect on the displacement energy are adressed in this work. For that, we carried out ab-initio molecular dynamics (MD) simulations for a broad range of projectiles (elements Hydrogen to Argon) impacting on graphene and h-BN, and determined the energies needed to displace C, N and B atoms, respectively. We further present and validate a scheme to incorporate the effect of spin-polarization on the displacement process - as spin-polarized ab-initio MD runs tend to fail at bond-breaking.

O 73: Electronic Structure of Surfaces 1

Time: Thursday 15:00–17:45 Location: S053

O 73.1 Thu 15:00 S053

Rashba-split image-potential state at Re(0001) — •Fabian Schöttke, Sven Schemmelmann, Peter Krüger, and Markus Donath — Westfälische-Wilhelms-Universität Münster, Germany

Since image states are located mainly in the vacuum in front of the surface, the influence of spin-dependent interactions on these states is a topic of ongoing debate in the literature. In particular, calculations predict Rashba-type spin splittings for image states [1], but experimental results of small spin splittings are controversial [2,3]. In addition, these two-photon-photoemission results using circular dichroism provide only indirect information about the spin polarization of states.

We measured the unoccupied surface electronic structure of Re(0001) with spin- and angle-resolved inverse photoemission. This method allows to investigate the spin orientations directly. We identified the $n = 1$ image state at a binding energy of $E_V - E = 0.68 \pm 0.04\,\mathrm{eV}$ and with an effective mass of $m^*/m_e = 1.2 \pm 0.1$. Careful spin-resolved measurements for several angles of electron incidence allowed us to detect Rashba-type spin-dependent energy splittings of this state with a Rashba parameter of $\alpha_\mathrm{R} = 105 \pm 33\,\mathrm{meV\,Å}$ [4].

[1] McLaughlan *et al.*, J. Phys.: Condens. Matter **16**, 6841 (2004).
[2] Tognolini *et al.*, Phys. Rev. Lett. **115**, 046801 (2015).
[3] Nakazawa *et al.*, Phys. Rev. B **94**, 115412 (2016).
[4] Schöttke *et al.*, Phys. Rev. B **105**, 155419 (2022).

O 73.2 Thu 15:15 S053

Rashba-split surface state and spin-dependent photon emission from Re(0001) at $\bar{\Gamma}$ — •Sven Schemmelmann[1], Fabian Schöttke[1], Peter Krüger[2], and Markus Donath[1] — [1]Physikalisches Institut, Westfälische Wilhelms-Universität, Münster — [2]Institut für Ferstkörpertheorie, Westfälische Wilhelms-Universität, Münster

The unoccupied electronic structure of the Re(0001) surface around the center of the surface Brillouin zone was investigated by spin- and angle-resolved inverse photoemission [1]. The detected states were studied with respect to intrinsic and extrinsic spin-polarization effects. A surface state with Rashba-type spin splitting is detected close to the Fermi level, that disperses downward below the Fermi energy. Furthermore, for normal electron incidence, we observed spin-dependent photon emission from unpolarized bulk and surface states. The sign of the observed spin asymmetry varies for different states and depends on experimental parameters such as electron spin-polarization direction and photon-detection angle. Maximum spin asymmetry is observed if the electron spin polarization and the plane of photon emission are perpendicular. The asymmetry is zero if both are parallel [2]. The effect is traced back to the spin-orbit-induced hybridization of the involved states.

[1] S. Schemmelmann *et al.*, Physical Review B **104**, 205425 (2021)

[2] E. Tamura and R. Feder, Solid State Commun. **79**, 989 (1991)

O 73.3 Thu 15:30 S053

Distinct Tamm and Shockley surface states on Re(0001) - mixed by spin-orbit interaction — •Marcel Holtmann[1], Peter Krüger[1], Koji Miyamoto[2], Taichi Okuda[2], Pascal J. Grenz[1], Shiv Kumar[2], and Markus Donath[1] — [1]Physikalisches Institut, Westfälische Wilhelms-Universität Münster, Wilhelm-Klemm-Straße 10, 48149 Münster, Germany — [2]Hiroshima Synchrotron Radiation Center, Hiroshima University, 2-313 Kagamiyama, Higashi-Hiroshima 739-0046, Japan

Tamm and Shockley states - two paradigmatic concepts are used to describe surface states not only in electronic systems but also in photonic and phononic crystals. The Re(0001) surface hosts both types of electronic surface states in neighboring but qualitatively different energy gaps. Interestingly, spin-orbit interaction generates a double "W"-shaped energy vs. $\mathbf{k}_{\|}$ dispersion by mixing both types of states and lifting their spin degeneracy. By combining spin- and angle-resolved photoemission, tight-binding model calculations as well as density functional theory including the photoemission process, we develop verifiable criteria to distinguish between the two types of surface states and arrive at a consistent picture of the role of spin-orbit interaction in such a scenario.

O 73.4 Thu 15:45 S053

Electronic structure of a square Te adlayer on Au(100) surface — •Begmuhammet Geldiyev[1], Tilman Kisslinger[2], Philipp Eck[3], Maximilian Ünzelmann[1], Tim Figgemeier[1], Jakub Schusser[1], Nikolai Tezak[1], Lutz Hammer[2], M. Alexander Schneider[2], Domenico Di Sante[3], Giorgio Sangiovanni[3], Hendrik Bentmann[1], and Friedrich Reinert[1] — [1]Experimentelle Physik 7 and Cluster of Excellence ct.qmat, Universität Würzburg — [2]Lehrstuhl für Festkörperphysik, Universität Erlangen-Nürnberg — [3]Institut für Theoretische Physik und Astrophysik and Cluster of Excellence ct.qmat, Universität Würzburg

In light of several theoretical predictions regarding the so-called square tellurene [1, 2], the growth and the electronic structure of Te deposited on a Au(100) surface is reported. At a coverage of 1/4 monolayer, Te forms a $p(2\times2)$ square lattice of adatoms on Au(100) as determined by a thorough LEED-IV characterization ($R_\mathrm{P} = 0.085$) and STM. Utilizing angle-resolved photoemission spectroscopy and density functional theory an interface-like state with mixed Te $p_{x,y}$ and Au d orbital character is identified in addition to a complex multitude of backfolded bands of the substrate. Vastly differing Rashba parameters along the $\overline{\mathrm{XG}}$ and $\overline{\mathrm{XM}}$ lines cannot be explained within an (anisotropic) Rashba picture. The spin splitting rather depends on the momentum space texture of the atomic orbitals. Our work may stimulate further experimental explorations of symmetry and topology effects in 2D square-lattice systems. [1] Xian et al., 2D Mater. **4**, 041003, (2017)

[2] Zhang et al., Phys. Rev. B **98**, 115411, (2018)

O 73.5 Thu 16:00 S053

Interplay of exchange and spin-orbit interaction in ultrathin Ni films on W(110) — •Pascal Jona Grenz[1], Peter Krüger[2], and Markus Donath[1] — [1]Physikalisches Institut, Westfälische Wilhelms-Universität Münster, 48149 Münster, Germany — [2]Institut für Festkörpertheorie, Westfälische Wilhelms-Universität Münster, 48149 Münster, Germany

Ferromagnetic adsorbates on high-Z substrates are prototypical systems for studying the combined influence of spin-orbit and exchange interaction on electronic states [1,2]. The unoccupied electronic states of ultrathin Ni films on W(110) were investigated with spin- and angle-resolved inverse photoemission. Measurements were performed along $\overline{\Gamma}\,\overline{\mathrm{N}}$ ($\overline{\Gamma}\,\overline{\mathrm{M}}$ with respect to the Ni(111) surface Brillouin zone), where the quantization axis of the Rashba-spin component is collinear to the easy magnetization axis. Remarkably, the observed unoccupied Ni-derived exchange-split states change their energy-momentum dispersion upon magnetization reversal. Density functional theory calculations show that this phenomenon is a consequence of substrate-induced spin-orbit coupling within the Ni adlayers.

[1] P. Moras *et al.*, Phys. Rev. B **91**, 195410 (2015)

[2] P.J. Grenz *et al.*, J. Phys.: Condens. Matter **33**, 285504 (2021)

O 73.6 Thu 16:15 S053

ARPES studies of Hf(0001) monocrystal: experiment and theory — •Saleem Ayaz Khan[1], Laurent Nicolai[1], Jean Zaraket[2,3], Maria Christine Richter[2,3], Olivier Heckmann[2,3], Laxman Nagi Reddy[2,3], Waly Ndiaye[2,3], Mauro Fanciulli[2,3], Karol Hricovini[2,3], and Jan Minar[1] — [1]NTC, Univiversity of West Bohemia, Plzeň, Czech Republic — [2]LPMS, CY Cergy Paris University, France — [3]DRF, IRAMIS, LIDYL, CEA Saclay, France

We present first ARPES studies of the electronic structure of the Hf(0001) surface. High-Z materials have attracted much interest, because the strong spin-orbit coupling in combination with the broken inversion symmetry and an important effective electric field at the surface results in a spin-momentum locking. Spin-polarized electrons at the surface are of interest in physics and novel applications in electronics and data processing. Extra sharp peaks observed in experiment are identified thanks to *ab-initio* calculations performed within the SPR-KKR package. These extra states come from oxygen contamination of the highly reactive surface of Hf(0001). Further comparison is done on the ARPES level, thanks to the one-step model which include all matrix elements effect, resulting in an excellent agreement.

O 73.7 Thu 16:30 S053

Establishing fundamentals of ARPES spin textures with model material PtTe$_2$ — •Mohammed Qahosh, Honey Boban, Xiao Hou, Claus-Michael Schneider, and Lukasz Plucinski — Electronic Properties-Peter Grünberg Institute (PGI-6).

A novel quantum material PtTe$_2$ is used to establish the connection between ARPES spin textures and initial state spin textures. PtTe$_2$ is predicted to host Dirac type-II fermions and a number of application-relevant properties. The crystal structure of the most stable 1T-PtTe$_2$ polytype is trigonal, belongs to the space group 164 (P$\bar{3}$m1), and exhibits 3-fold mirror planes and inversion symmetry. Since bulk 1T-PtTe$_2$ is both inversion-symmetric and non-magnetic, no bulk spin-polarized bands are allowed due to the Kramers degeneracy. At the surface, spin polarization is expected due to the broken inversion symmetry, however, it must obey the mirror and time-reversal symmetries.

We measured the dependence of spin-polarization on the symmetries of the ARPES experiment. This is performed in two geometries, with the reaction plane parallel to K-Γ-K and M-Γ-M reciprocal directions, i.e. either along or orthogonal to the crystal mirror plane. The measured spin texture is symmetric when the reaction plane is parallel to K-Γ-K. However, we could see asymmetries in the spin texture when the reaction plane is parallel to M-Γ-M. Such asymmetries are not allowed in the initial state and illustrate the mechanism of geometry-induced spin filtering in ARPES.

O 73.8 Thu 16:45 S053

Surface doping of the MnBi$_2$Te$_4$ family by rubidium deposition — •Klara Volckaert[1], Paulina Majchrzak[1], Raphaël Dubourg[1], Zhihao Jiang[1], Xing-Chen Pan[2], Yong Chen[1], and Søren Ulstrup[1] — [1]Department of Physics and Astronomy, Aarhus University, Denmark — [2]Advanced Institute for Materials Research, Tohoku University, Japan

Intrinsic magnetic topological insulators, in the form of MnBi$_2$Te$_4$, have recently been realised as a remarkable platform to study quantised magnetoelectric phenomena. Here we look into the surface electronic structure during rubidium deposition in a combined angle-resolved photoemission and core level study. We find that for MnBi$_2$Te$_4$, the initial electron doping effect from the adsorbed rubidium atoms is small. However, deposition on the higher stoicheometry compound MnBi$_4$Te$_7$ leads to a dramatic modification of the electronic structure, which is different for the surface terminated by a Bi$_2$Te$_3$ quintuple layer compared to a MnBi$_2$Te$_4$ septuple layer. Additionally, high rubidium deposition rates lead to a change of the electronic structure including a shift of the valence bands towards the Fermi level for both compounds, presumably due to Rb-Te-Bi alloying. A distinct quantization of the valence states is simultaneously observed. These results are the first to explore the tunability of electronic states the surface terminations of MnBi$_4$Te$_7$ with in situ alkali doping and substitution, which could modify the surface magnetic ordering.

O 73.9 Thu 17:00 S053

Theoretical and experimental HARPES study of Weyl-semimetal TaAs: The application of machine-learning — •Trung-Phuc Vo[1], Arian Arab[2], Sunil Wilfred D'Souza[1], Laurent Nicolaï[1], Tien-Lin Lee[3], Nitesh Kumar[4], Claudia Felser[4], Alexander Gray[2], and Ján Minár[1] — [1]New Technologies - Research Centre, University of West Bohemia, 301 00 Pilsen, Czech Republic — [2]Department of Physics, Temple University, Philadelphia, Pennsylvania 19122, USA — [3]Diamond Light Source Ltd., Didcot, Oxfordshire OX11 0DE, United Kingdom — [4]Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 40, 01187 Dresden, Germany

The electronic structure properties of tantalum arsenide (TaAs), a Weyl semimetal, have been studied by soft and hard X-ray angle-resolved photoemission spectroscopy (ARPES) at energies of 440 eV and 2150 eV, respectively. For the first time, TaAs is experimentally investigated by the bulk sensitive photoemission in the hard X-ray regime. In order to interpret experimental data we performed one-step model of photoemission calculation which includes all matrix elements and final state effects. Due to the strong photon momentum effects and uncertainty in the tilt of experimental geometry we used a so-called machine learning algorithm combined with a free-electron final-state model to find best possible experimental parameters. Our findings re-emphasize the overwhelming accuracy of hard X-ray ARPES compared to the traditional ultraviolet and soft X-ray one in case of bulk electronic structure, motivating further material discoveries.

O 73.10 Thu 17:15 S053

Electron correlation in SrTiO3 studied by double photoemission spectroscopy with a MHz high-order harmonics laser source — •Robin Kamrla[1], Cheng-Tien Chiang[1,2], Frank Oliver Schumann[3], and Wolf Widdra[1] — [1]Institute of Physics, Martin-Luther-Universität Halle-Wittenberg, Halle (Saale), Germany — [2]Academica Sinica, Institute of Atomic and Molecular Sciences, Taiwan — [3]Max Planck Institute of Microstructure Physics, Halle (Saale), Germany

Photoelectron spectroscopy (PES) has provided deep insights into the electronic structure of solids. However, correlation effects can only be addressed indirectly. To observe such phenomena directly, double photoemission (DPE) spectroscopy is able to detect pairs of correlated photoelectrons that are emitted upon absorption of a single photon. Upon surface near doping, SrTiO3 (001) with a bandgap of 3.4 eV forms a two-dimensional electron gas (2DEG). In this contribution we present PES and DPE data for SrTiO3 (001) with and without 2DEG states at the surface, obtained by a high-order harmonic (HHG) light source at 25.2 and 30.0 eV. PES reveals a change in spectral weight of the O2p states and a surface band bending of 250 meV upon flipping SrTiO3 (001) into the 2DEG state and oxygen vacancy derived states emerging within the bandgap. In DPE, a band-bending induced shift of 500 meV, changes in the intensity of the O2p derived two hole states as well as emission from vacancy-valence pairs are identified via the two-electron sum energy spectrum. In addition, the role of Auger decays for the shallow Sr4p and O2s core level will be discussed.

O 73.11 Thu 17:30 S053

Doping of 1D topologically protected edge states on the (001) surface of the topological crystalline insulator (Pb,Sn)Se — •Florian Keller, Artem Odobesko, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Topological crystalline insulators (TCI) are a class of materials with topological protected surface states protected by crystalline symmetry. A particularly popular representative of this material class is (Pb,Sn)Se which exhibits four Dirac cones per Brillouin zone. It has been shown that surface step edges with a height equivalent to an odd number of atomic layers results in a topologically protected one-dimensional edge state which is characterized by a peak at the Dirac energy [1]. Theoretical analysis suggests that this state is caused by the broken translation-invariance at the step edge and originates from flat-dispersing bands which connect pairs of surface Dirac nodes [1]. Due to intrinsic doping, the energy of the edge modes of as-grown crystals is usually well separated from the Fermi level. Here we investigate the behavior of these one-dimensional edge modes during Fe surface doping. Since Fe donates charge to p-doped PbSnSe, it results in a downwards-bending of the surface band structure. We observe a peak splitting as the Dirac energy gets close to the Fermi level. We discuss the potential origins of this observation in terms of electron correlations.

[1] Sessi, Paolo, Science 354, 6317 (2016)

O 74: Organic Molecules at Surfaces 5: Molecular Switches

Time: Thursday 15:00–18:00 Location: S054

O 74.1 Thu 15:00 S054

Reprogrammable molecular memory array based on chemical switching — •Tobias Birk[1], Anja Bauer[1], Fabian Paschke[1], Rainer Winter[2], and Mikhail Fonin[1] — [1]Fachbereich Physik, Universität Konstanz, 78457 Konstanz, Germany — [2]Fachbereich Chemie, Universität Konstanz, 78457 Konstanz, Germany

Technology on a molecular base promises highly interesting innovations for ultra-dense information storage devices and molecular electronics. Recently, a molecular three-state switch based on triazatruxene (TAT) on Ag(111) has been shown to have a great potential as data storage unit [1].

Here, we demonstrate the precise manipulation of the switching characteristics of individual TAT molecules within a 2D array using a low temperature scanning tunneling microscope (STM). By using the tip of the STM and the field applied within the contact, a successive pinning of the molecule via alkyl groups is achieved, leading to a gradual suppression of the current induced molecular switching. The possibility to reversibly switch between differently pinned molecules in combination with the three-level switching of the unpinned molecule offers the possibility to realize 9 states on a single molecule. We also demonstrate that the intermolecular interaction between the switches within the 2D array leads to a strong increase of the number of states, which can be detected on a single TAT unit, yielding up to 24 distinguishable states.

[1] A. Bauer et al., Adv. Mater 32, 1907390 (2020)

O 74.2 Thu 15:15 S054

Uni-directional rotation of molecular motors on Cu(111) — •Monika Schied[1,2], Deborah Prezzi[3], Dongdong Liu[4], Peter Jacobson[1,5], Elisa Molinari[3], James M. Tour[4], and Leonhard Grill[1] — [1]University of Graz, Austria — [2]Elettra Sincrotrone Trieste, Italy — [3]Nanoscience Institute of CNR, Italy — [4]Rice University, USA — [5]The University of Queensland, Australia

Artificial molecular motors that convert external energy into controlled motion have seen great developments in the last decades [1]. While many studies exist in solution, little is known how such functional molecules behave on a surface. However, such a solid support can be advantageous as it offers fixed points of reference as well as confinement in two dimensions, making it easier to study the directionality of their motion.

We have used low-temperature scanning tunnelling microscopy (STM) to study single molecules with a so-called Feringa motor [2,3] on a Cu(111) surface. It was found that rotations of individual molecules can be induced over rather long distances by voltage pulses with the STM tip. Importantly, these rotations show high directionality (clockwise or anticlockwise), which will be discussed in view of their specific chemical structure and adsorption.

[1] W. R. Browne and B. L. Feringa, Nat. Nanotech. 1, 25 (2006)
[2] T. Kudernac et al., Nature 479, 208 (2011)
[3] A. Saywell et al., ACS Nano 10, 10945 (2016)

O 74.3 Thu 15:30 S054

Precise control of single-molecule motion on Ag(111) — •Donato Civita, Grant Simpson, and Leonhard Grill — Department of Physical Chemistry, University of Graz, Austria

The motion of molecules adsorbed on metal single crystal surfaces is of fundamental importance in various fields such as heterogeneous catalysis, and on-surface polymerization. During diffusion, however, the motion of adsorbed molecules is characterised by random direction changes and thus control is limited. Moreover, adsorbates in the surroundings as well as substrate defects can strongly influence molecular motion.

With the use of a scanning tunnelling microscope (STM) at low temperature, we can control the motion of single di-bromo-ter-fluorene (DBTF) molecules on a Ag(111) surface over distances of more than

100 nm with picometric precision [1]. We find that a single molecule can move strictly along one atomic row across the surface. The molecule can be repelled or attracted by the STM tip, driven by an interplay of van der Waals and electrostatic interactions. The large spatial extension of the motion, and its unidimensional confinement allow the direct measurement of the molecular velocity. Ultimately, this system demonstrates the possibility of studying the influence of surrounding adsorbates, crystal defects, and STM tip on the molecular motion and velocity.

[1] D. Civita, M. Kolmer, G. J. Simpson, A.-P. Li, S. Hecht, L. Grill, Control of long-distance motion of single molecules on a surface, Science, Vol. 370, Issue 6519, pp. 957-960 (2020).

O 74.4 Thu 15:45 S054

Dynamics of a chiral molecular rotor under a scanning-tunneling microscope — •Richard Korytár[1] and Ferdinand Evers[2] — [1]Charles University, Prague — [2]Universität Regensburg

Motivated by an experimentally realized chiral molecular switch, we devise a classical theory of the switching process, relevant for molecular electronics. The system of interest is a chiral molecular rotor in a scanning-tunneling setup, i.e. the switching occurs under the electric current. The molecule is modeled by a path in three dimensions. The path is massive and can rotate around a fixed axis. The incident electron traverses the path, inducing a torque on the path. We represent this two-body dynamics in a Lagrangian formalism. Switching mechanism and switching rates are discussed.

O 74.5 Thu 16:00 S054

Bipolar single-molecule electrofluorochromism — •Tzu-Chao Hung[1], Roberto Robles[2], Brian Kiraly[1], Julian H. Strik[1], Bram A. Rutten[1], Alexander A. Khajetoorians[1], Nicolas Lorente[2], and Daniel Wegner[1] — [1]Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands — [2]Centro de Fisica de Materiales, CFM/MPC (CSIC-UPV/EHU), Paseo de Manuel de Lardizabal 5, 20018 Donostia-San Sebastián, Spain

The interplay between the charge state and the fluorescence of a molecule is not only important for the spectroscopic analysis of chemical reactions, but electrofluorochromic molecules can also be utilized in displays, sensors, and switches. To understand the fundamental mechanisms on the single-molecule level, we studied the transient charged state of zinc phthalocyanine (ZnPc) adsorbed on ultrathin NaCl films on Ag(111) by combining scanning tunneling microscopy (STM) and spectroscopy (STS) with STM-induced luminescence (STML). We found evidence for both cationic ($[\mathrm{ZnPc}]^+$) and anionic ($[\mathrm{ZnPc}]^-$) fluorescence, depending on the polarity of the tip-sample bias. By carefully mapping the molecular frontier orbitals over a wide energy range, correlating them with onset energies for light emission and comparing with results from DFT calculations, we propose an alternative charging and electroluminescence mechanism. Our study provides new insights into the tunability of molecular optical response, as well as novel aspects toward utilization of bipolar electrofluorochromism in devices.

O 74.6 Thu 16:15 S054

Electronic Motor Based on Single Tripodal Chiral Molecule — •Julian Skolaut[1], Lukas Gerhard[1], Nico Balzer[2], Michal Valasek[2], Jan Wilhelm[4], Philipp Markus[5], Marcel Mayor[2,3], Ferdinand Evers[4], and Wulf Wulfhekel[1,5] — [1]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, Eggenstein- Leopoldshafen, Germany — [2]Institute of Nanotechnology, KIT, Eggenstein-Leopoldshafen, Germany — [3]Department of Chemistry, University Basel, Basel, Switzerland — [4]Institute of Theoretical Physics, University of Regensburg, Regensburg, Germany — [5]Physikalisches Institut, KIT, Karlsruhe, Germany

We present our results concerning a single molecular motor driven by the current in an STM. Three anchoring groups fix the molecules to a Au(111) surface. In specific ordered structures, the molecules adsorb such, that the protruding head group is free to rotate. This chiral group is supposed to perform a rotation in a preferred direction, proposed to be driven based on the chiral-induced spin selectivity (CISS) effect. At fixed tip positions above the molecules, three distinguishable current levels can be observed. These are interpreted as metastable rotational states. That way, two rotation directions can be defined. Via binomial tests, we verify that the surplus of rotational switches in one direction compared to the other is statistically significant. In voltage and current dependent measurements, two interesting trends are observed. Firstly, the rate of events decreases with increasing current. Secondly, the asymmetry in the switching events shows non-monotonic behavior, depending on the voltage.

O 74.7 Thu 16:30 S054

Proton Transfer in Single Asymmetric Porphycene Molecules — •Simon Jaekel[1,2], Jacek Waluk[3], and Leonhard Grill[1] — [1]Department of Physical Chemistry, University of Graz, Austria — [2]Chair of Physical Chemistry II, University of Erlangen-Nürnberg (FAU), Germany — [3]Polish Academy of Science, Warsaw, Poland

Studying single molecular switches is of interest for a better understanding of fundamental physical and chemical processes, but also in view of their possible use in smart materials and nanoscale applications. It has been shown that switches based on tautomerization, i.e. hydrogen transfer, are especially suited for scanning tunneling microscopy (STM) studies, because the electronic states near the Fermi level are very sensitive to the position of the hydrogen atoms. Experiments on symmetric molecules such as naphthalocyanine [1], tetraphenyl-porphyrin [2], and porphycene [3] showed that the energies of the tautomers are degenerate, barring environmental modulation. Thus, if one is interested in switching processes with preferential switching directions between multiple states, structurally asymmetric molecules are of particular interest.

Here, we report STM results of the tautomerization properties of 22-Oxahemiporphycene, a derivative of porphycene with an asymmetric macrocycle, on a metal surface.

[1] Liljeroth et al. Science, 317 (2007), 1203-1206

[2] Auwärter et al. Nature Nanotechnology, 7 (2011), 41-46

[3] Kumagai et al. Physical Review Letters 111 (2013), 246101

O 74.8 Thu 16:45 S054

Tuning the chirality change of a single molecule by van der Waals interactions — •Yunjun Cao and Karina Morgenstern — Physical Chemistry I, Ruhr-Universität Bochum, Bochum, Germany

The chiral induction and control of molecules by non-covalent intermolecular interactions, like hydrogen bonding and van der Waals interactions, is crucial to understand the origin of homochirality in nature. However, it remains challenging to address these subtle intermolecular interactions at a single-molecular level, especially the weakest of them, the van der Waals interactions. Here, by adsorbing a specifically designed carbene molecule on a copper surface, we examine the influence of the van der Waals interactions on the chirality induction by scanning tunneling microscopy. While the strongly binding carbene center of this molecule suppresses any side-reactions upon excitation, its two low-interacting phenyl rings facilitate a chirality change induced by inelastically tunneling electrons. The potential energy of the chirality change is modified by the van der Waals interactions in the presence of a tip. A marginal change induces an asymmetric distribution of the carbene molecule between its two enantiomers during this chirality change. Our study shows how the weak van der Waals interactions alter the dynamics of chirality changes at the molecular level, enabling an in-depth understanding of the origin of homochirality in nature and providing new insights into the construction of homochiral supramolecular assemblies in solutions and on solid surfaces.

O 74.9 Thu 17:00 S054

Lateral Force Microscopy Reveals the Energy Barrier of a Molecular Switch — Alfred John Weymouth, Elisabeth Riegel, •Bianca Simmet, Oliver Gretz, and Franz Josef Giessibl — Universität Regensburg, Regensburg, Germany

Copper phthalocyanine (CuPc) is a small molecule often used in organic light emitting diodes where it is deposited on a conducting electrode. Previous scanning tunneling microscopy (STM) studies of CuPc on Cu(111) have shown that inelastic tunneling events can cause CuPc to switch between a ground state and two symmetrically equivalent metastable states in which the molecule is rotated. We investigated CuPc on Cu(111) and Ag(111) with STM and lateral force microscopy (LFM). Even without inelastic events, the presence of the tip can induce rotations and upon closer approach, causes the rotated states to be favored. Combining STM measurements at various temperatures and LFM measurements, we show that the long-range attraction of the tip changes the potential energy landscape of this molecular switch. We can also determine the geometry of the rotated and ground states. We compare our observations of CuPc on Cu(111) to CuPc on Ag(111). On Ag(111), CuPc appears flat and does not rotate. Stronger bonding typically involves shorter bond lengths, larger shifts of energy levels, and structural stability. Although the binding of CuPc to Cu(111) is stronger than that on Ag(111), the nonplanar geometry of CuPc on Cu(111) is accompanied by two metastable states which are not present on the Ag(111) surface.

ACS Nano, 15, 3264 (2021)

O 74.10 Thu 17:15 S054

Evidence of trion-libron coupling in chirally adsorbed single molecules — •Jiří Doležal[1,2], Sofia Canola[1], Prokop Hapala[1], Rodrigo Ferreira[1], Pablo Merino[3], and Martin Švec[1] — [1]Institute of Physics, Czech Academy of Sciences; Cukrovarnická 10/112, CZ16200 Praha 6, Czech Republic — [2]Faculty of Mathematics and Physics, Charles University; Ke Karlovu 3, CZ12116 Praha 2, Czech Republic — [3]Instituto de Ciencia de Materiales de Madrid; CSIC, Sor Juana Inés de la Cruz 3, E28049 Madrid, Spain

Interplay between the motion of nuclei and excited electrons in molecules plays a key role both in biological and artificial nanomachines. Here we provide a detailed analysis of coupling between quantized librational modes (librons) and charged excited states (trions) on single phthalocyanine dyes adsorbed on a surface. By means of tunnelling electron-induced electroluminescence, we identify libronic progressions on a μeV energy range in spectra of chirally adsorbed phthalocyanines, which are otherwise absent from spectra of symmetrically adsorbed species. Experimentally measured libronic spectra match very well the theoretically calculated libron eigenenergies and peak intensities (Franck-Condon factors) and reveal an unexpected depopulation channel for the zero libron of the excited state that can be effectively controlled by tuning the size of the nanocavity. Our results showcase the possibility of characterizing the dynamics of molecules by their low-energy molecular modes using μeV-resolved tip-enhanced spectroscopy.

O 74.11 Thu 17:30 S054

Design Principles for Metastable Standing Molecules — Hadi H. Arefi[1], Marvin Knol[1], Daniel Corken[2], James Gardner[2], F. Stefan Tautz[1], Reinhard J. Maurer[2], and •Christian Wagner[1] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]Department of Chemistry, University of Warwick, Coventry, UK

Molecular nanofabrication with a scanning probe microscope is a promising route towards the prototyping of metastable functional molecular structures and devices which do not form spontaneously. The aspect of mechanical stability is crucial for such structures especially if they extend into the third dimension vertical to the surface. A prominent example are freestanding molecules on a metal which can function as field emitters or electric field sensors[1,2]. Improving the stability of such molecular configurations is an optimization task involving many degrees of freedom. Here, we present a combination of scanning probe experiments with ab initio potential energy calculations to investigate the stability of a prototypical standing molecule. We cast our results into a simple set of universal design principles for such metastable structures, the validity of which we demonstrate in two computational case studies. This offers the intuition needed to fabricate new devices without tedious trial and error.

[1] T. Esat, N. Friedrich, F. S. Tautz, R. Temirov, *Nature* **558**, 573 (2018)

[2] C. Wagner, M. F. B. Green, P. Leinen, T. Deilmann, P. Krüger, M. Rohlfing, R. Temirov, F. S. Tautz, *PRL* **115**, 026101 (2015)

O 74.12 Thu 17:45 S054

Improving the Switching Efficiency in Azobenzene Derivative Film on Graphite-Air Interface — •Thiruvancheril G. Gopakumar[1], Khushboo Yadav[1], Hariom Birla[1], Showkat H. Mir[1,2], Thomas Halbritter[2], Alexander Heckel[2], and Jayant K. Singh[3] — [1]Department of Chemistry, Indian Institute of Technology Kanpur, Kanpur 208016, India — [2]Institute for Organic Chemistry and Chemical Biology, Goethe-University Frankfurt, Max-von-Laue-Str. 9, 60438 Frankfurt, Germany — [3]Department of Chemical Engineering, Indian Institute of Technology Kanpur, Kanpur 208016, India

The trans isomer of azobenzene (AB) and its derivatives is the most abundant under equilibrium-thermodynamical conditions and is known to switch between its trans and cis states when triggered by light and electrons/holes on graphite.[1] In this work, we show that AB derivatives are switching between two cis states (cis, cis') when electrons/holes induced switching is performed on a cis dominant non-equilibrium initial condition at HOPG-air interface. The switching efficiency in the cis adlayer is several folds higher than that in the trans adlayer. This is related to the low switching barrier for cis-cis' switching compared to that of trans-cis switching as revealed by density functional theory (DFT) calculations.

1) K. Yadav, S. Mahapatra, T. Halbritter, A. Heckel, T. G. Gopakumar, J. Phys. Chem. Lett., 2018, 9, 6326-6333.

O 75: Members' Assembly

Topics: Report of the Chairman; Presentation of the Gerhard Ertl Young Investigator Award; Miscellaneous

Time: Thursday 19:00–19:30 Location: H1

All members of the Surface Science Division are invited to participate.

O 76: Post-Deadline Session

Time: Thursday 19:30–20:30 Location: H1

Contributed Post-Deadline Talks

O 77: Overview Talk Guillaume Schull

Time: Friday 9:30–10:15 Location: S054

Invited Talk O 77.1 Fri 9:30 S054

Sub-molecular fluorescence microscopy with STM — •Guillaume Schull — IPCMS - CNRS/Unistra - Strasbourg

The electric current traversing the junction of a scanning tunneling microscope (STM) may lead to a local emission of light that can be used to generate sub-molecularly resolved fluorescence maps of individual molecules [1]. Combined with spectral selection and time-correlated measurements, this hyper-resolved fluorescence microscopy approach allowed us to scrutinise the vibronic structure of individual molecules [2] to characterise the emission properties of charged species [3], to track the motion of hydrogen atoms within free-base phthalocyanine molecules [4] and to follow energy transfers between multi-molecular architectures [5].

[1] A. Roslawska et al., PRX 12, 011012 (2022)
[2] B. Doppagne et al., PRL 118, 127401 (2017)
[3] B. Doppagne et al. Science, 361, 251 (2018)
[4] B. Doppagne et al. Nature Nanotechnol. 15, 207 (2020) .
[5] S. Cao et al. Nature Chem. 13, 766 (2021)

O 78: Plasmonics and Nanooptics 3

Time: Friday 10:30–12:30 Location: H3

O 78.1 Fri 10:30 H3

Reconfiguring magnetic resonances with the plasmonic phase-change material In$_3$SbTe$_2$ — •Lukas Conrads, Andreas Hessler, Konstantin Wirth, Matthias Wuttig, and Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

For miniaturized active nanophotonic components, resonance tuning of nanoantennas is a key ingredient. Phase-change materials (PCMs) have been established as prime candidates for non-volatile resonance tuning based on a change in refractive index [1]. Currently, a novel material class of switchable infrared plasmonic PCMs, like In$_3$SbTe$_2$ (IST), is emerging. Since IST can be locally optically switched between dielectric (amorphous phase) and metallic (crystalline phase) states in the whole infrared range, it becomes possible to directly change the geometry and size of nanoantennas to tune their infrared resonances [2]. Here, crystalline IST split-ring resonators (SRRs) are directly optically written and reconfigured in their arm size to continuously tune their magnetic dipole resonances over a range of 2.4 μm without changing their electric dipole resonances. The SRRs are further modified into crescents and J-antennas, which feature more complex resonance modes dependent on the polarization of the incident light [3]. Our concepts are well-suited for rapid prototyping, speeding up workflows for engineering ultrathin, tunable, plasmonic devices for infrared nanophotonics, telecommunications, or (bio)sensing.

[1] Wuttig et al., *Nat. Photon.* **11**, 465 (2017) [2] Heßler et al., *Nat. Commun.* **12**, 924 (2021) [3] Heßler, Conrads et al. *ACS Photonics* **9**, 5 (2022)

O 78.2 Fri 10:45 H3

Impact of atomistic structure and dynamics on inelastic light scattering in a plasmonic picocavity — •Franco Bonafé[1], Shuyi Liu[2], Heiko Appel[1], Martin Wolf[2], Takashi Kumagai[2,3], and Angel Rubio[1] — [1]MPI for Structure and Dynamics of Matter, Hamburg, Germany — [2]Dpmt. of Physical Chemistry, Fritz-Haber Institute, Berlin, Germany — [3]Center for Mesoscopic Sciences, Institute for Molecular Science, Okazaki, Japan

Atomically sharp metallic tips can focus an electromagnetic field down to the sub-nanometer scale, leading to strong light-matter interactions in plasmonic “picocavities”. This atomic-scale field can be used in plasmon-enhanced spectroscopy, such as tip-enhanced Raman spectroscopy (TERS), which has enabled the visualization of optical properties at sub-molecular resolution. However, a full microscopic understanding of the interplay of structural relaxation, bonding, near-fields and vibrations has not been achieved.

In this work, we combine experimental observations of inelastic light scattering from a single silver adatom in a plasmonic picocavity controlled by a low-temperature scanning tunneling microscope (STM), with ab initio real-time electron dynamics simulations. The experiment demonstrates a dramatic enhancement of Raman scattering that occurs upon the formation of a quantum point contact. We model possible geometries for the plasmonic tips and compute the vibrational modes, electronic current and near-fields localized in the vicinity of the single adatom. These simulations reveal a crucial role of the atomistic structural relaxation in the optical response in a plasmonic nanocavity.

O 78.3 Fri 11:00 H3

Vector Polarimetry - Measuring Electrical Fields on Surfaces — •Alexandra Rödl[1], David Janoschka[1], Pascal Dreher[1], Alexander Neuhaus[1], Bettina Frank[2], Timothy Davis[1,2,3], Michael Horn-von Hoegen[1], Harald Giessen[2], and Frank-J. Meyer zu Heringdorf[1] — [1]Faculty of Physics and Center for Nanointegration, Duisburg-Essen, University of Duisburg-Essen, 47048 Duisburg, Germany — [2]4th Physics Institute, Research Center SCoPE, and Integrated Quantum Science and Technology Center, University of Stuttgart, 70569 Stuttgart, Germany — [3]School of Physics, University of Melbourne, Parkville, Victoria 3010 Australia

Non-linear photoemission microscopy has been established as an excellent tool to investigate nano-optical fields at surfaces, in particular the fields of surface plasmon polaritons (SPPs). In a pump-probe experiment with femtosecond laser pulses we excite SPPs at grooves that are ion-milled into a Au platelet. The electric field of the probe-laser pulse interferes coherently with the electric field of the SPP and electrons are liberated through a nonlinear process by the combined field at the surface. The contrast depends on the alignment of the probe polarization and the orientation of the in-plane component of the SPP’s electric field. Using a set of different polarizations for the probe laser pulse while keeping the same excitation conditions, one can reconstruct the in-plane component of the electric field of the SPP. The out-of-plane field component is calculated by Maxwell’s equations to reconstruct the full electric vector field. Here, we measure and image complex electric vector fields of SPPs and analyze their topology.

O 78.4 Fri 11:15 H3

Spatially-resolved THz near-field spectroscopy — •Moritz B. Heindl[1], Nicholas Kirkwood[2], Tobias Lauster[3], Julia A. Lang[1], Markus Retsch[3], Paul Mulvaney[2], and Georg Herink[1] — [1]Experimental Physics VIII, University of Bayreuth, Germany — [2]ARC Centre of Excellence in Exciton Science, School of Chemistry, University of Melbourne, Australia — [3]Physical Chemistry I, University of Bayreuth, Germany

Spectroscopic access to ultrafast electric waveforms is critical to the understanding of plasmonic and field-driven nonlinear phenomena, yet, microscopic measurements still present a grand challenge. Here, we present a fluorescence-based field microscope for imaging ultrafast THz near-field evolutions employing the quantum-confined Stark-effect in semiconductor quantum dots [1,2]. This Quantum-Probe Field Microscopy (QFIM) scheme [3] allows for detection of strongly confined near-fields in three-dimensional structures. Using QFIM, we demonstrate spatially-resolved near-field spectroscopy of single THz resonators and propagating THz excitations inside wave-guiding structures.

[1] Hoffmann, M. C. et al. Appl. Phys. Lett. 97, 231108 (2010)
[2] Pein, B. C. et al. Nano Lett. 17, 5375-5380 (2017)
[3] Heindl, M. B. et al. Light Sci. Appl. 11, 5 (2022)

O 78.5 Fri 11:30 H3

Mode-selective imaging and control of nano-plasmonic near-fields — •Murat Sivis[1,2], Hugo Lourenço-Martins[1,2], Andre Geese[2], Tyler R. Harvey[2], Thomas Danz[1,2], Radwan M. Sarhan[3], Matias Bargheer[3], Armin Feist[1,2], and Claus Ropers[1,2] — [1]Max Plank Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute - Solids and Nanostructures, University of Göttingen, Germany — [3]Institut für Physik und Astronomie, Universität Potsdam, Potsdam, Germany

Electron energy-loss spectroscopy (EELS) in a transmission electron microscope allows for the study of optical excitations in plasmonic nanostructures with sub-nanometer spatial resolution. While EELS is a powerful tool, it can only provide information about the spontaneous losses in a system with limited spectral resolution (10-100 meV in the most advanced microscopes). Recent developments in ultrafast transmission electron microscopy overcome these limitations and enable the probing of laser-excited modes by using photon-induced near-field electron microscopy (PINEM). Here, we demonstrate how PINEM can be used to measure the modal structure of the optical response of individual plasmonic systems (metal nano-triangle-resonators) at the nanoscale. Using our boundary element method (BEM)-based data analysis, we extract the magnitude and relative phase of each plasmonic mode from optical near-field maps. This approach opens a route to study the influence of the laser polarization, wavelength and incidence angle on the population of each mode, as well as the control of complex mode patterns created by multicolor fields.

O 78.6 Fri 11:45 H3

Tailoring of nonlinear metasurfaces using sampling-based optimization — •David Hähnel, Jens Förstner, and Viktor Myroshnychenko — Paderborn University, Theoretical Electrical Engineering, Warburger Str. 100, 33098 Paderborn, Germany

Various efficient methods for the design and optimization of linear metasurfaces have already been developed in the past [1]. Nowadays, attempts are being made to use these methods also for the design and optimization of nonlinear metasurfaces, which is a quite complex task due to the nonlinear processes involved. Here we present the design and optimization of all-dielectric nonlinear metasurfaces using a simple sampling method combined with Monte Carlo simulation, demonstrating that the use of sophisticated optimization methods is not necessarily required. Furthermore, this combination provides an effective approach in the optimization of nonlinear metasurfaces with a dynamic problem, such as a varying number of optimization parameters that are unknown in advance. We apply this method combination in the optimization of a nonlinear beam deflector metasurface for the third harmonic, which consist of an array of elliptical silicon disks and obtained an significant improvement in the radiated intensity compared to literature results [2]. [1] Chen, S. et al., Advanced Optical Materials 2018, 6, 1800104. [2] Lei Wang et al., Nano Lett. 2018, 18, 6, 3978-3984.

O 78.7 Fri 12:00 H3

Polarization selective investigation of plasmonic Bloch modes with dark-field spectroscopy — •Maximilian Johannes Black and Nahid Talebi — Institute for Experimental and Applied Physics, Kiel University, 24118 Kiel, Germany

Plasmonic and photonic crystals are widely used to mold the flow of light. Owing to its symmetry even the seemingly simple structure of a periodic square lattice of holes within a thin gold film shows a broad variety of plasmonic Bloch modes that are distinguished by the momenta and polarization. Therefore, the response of the system to the excitation is formed by a superposition of these modes. In this work we use polarization-selective optical dark-field microscopy to decompose these modes. Dark-field microspectroscopy is used to enhance the resolution and to suppress the background illumination, whereas linear polarizers both in the illumination and the detection path enable the selection of Bloch modes by polarization. We find that the dominating signal is formed by the selective excitation and detection of radiating magnetic moments. These and a variety of other modes are visualized by hyperspectral imaging. In the far-field reciprocal space interference fringes are resolved, indicating the propagation of the observed modes along the surface of the plasmonic crystal. Our results prove the possibility to thoroughly investigate plasmonic Bloch modes using dark-field microscopy, adding its versatility to the range of methods for measuring light-matter interactions.

O 78.8 Fri 12:15 H3

A Ginzburg Landau model for femtosecond charge-density wave dynamics at the atomic scale — •Kurt Lichtenberg[1], Mohamad Abdo[1,2], Shaoxiang Sheng[1], Luigi Malavolti[1,2], and Sebastian Loth[1,2] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Max Planck Institute for Solid State Research, Stuttgart, Germany

Charge-density waves (CDWs) feature collective excitations that can be observed as an oscillatory response of the electron system of a material to a fast optical stimulus [1]. At the same time, scanning tunneling microscopy (STM) measurements reveal highly localized interactions with atomic defects [2]. THz pump-probe spectroscopy in the STM reveals highly heterogeneous dynamics with spatial variations down one unit cell of the CDW. To improve the understanding of such local CDW dynamics, we developed an empirical model that is motivated by former approaches [3,4] and based on time-dependent Ginzburg Landau Theory. The combination of this theory with THz-STM measurements establishes a possibility to study the interplay between collective CDW dynamics with atomic pinning sites.

[1] M.-A. Méasson et. al, PRB 89, 060503(R) (2014)
[2] C. J. Arguello et. al, PRB 89, 235115 (2014)
[3] W. L. McMillan, PRB 12, 1187-1196 (4) (1975).
[4] G. Grüner, Density Waves in Solids. Perseus Publishing - Cambridge, Massachusetts, (2000).

O 79: Surface Reactions and Heterogeneous Catalysis 3

Time: Friday 10:30–12:45 Location: H4

Topical Talk O 79.1 Fri 10:30 H4

Exploitation of Heterocycles for N-doped Graphene Nanomaterials — Rémy Pawlak[1], Ulrich Aschauer[2], Silvio Decurtins[2], Jascha Repp[3], Pavel Jelinek[4], Ernst Meyer[1], Laerte L. Patera[5], and •Shi-Xia Liu[2] — [1]Department of Physics, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland — [2]Department of Chemistry, Biochemistry and Pharmaceutical Sciences, University of Bern, 3012 Bern, Switzerland — [3]Institute of Experimental and Applied Physics, University of Regensburg, 93053 Regensburg, Germany — [4]Institute of Physics of Czech Academy of Sciences, 16200 Prague, Czech Republic — [5]Department of Chemistry and Catalysis Research Center, Technical University of Munich, 85748 Garching, Germany

On-surface chemical reactions have been intensively investigated in order to obtain carbon-based functional nanomaterials which very often cannot be synthesized by wet chemistry. Thus, tailored heterocyclic precursors are becoming increasingly important for the development of highly symmetric 2D-conjugated porous architectures and 1D N-doped graphene nanoribbons (GNRs) with desired functions due to their intrinsic electronic properties. This presentation will focus on our collaborative work on C-C coupling reactions on various surfaces leading to the formation of a range of nanostructures, including molecular wires, fully fused porphyrin-GNR hybrids, as well as 1D- and 2D-conjugated porous networks. All of these atomically precise nanostructures can be directly visualized by STM and AFM. The fine-tuned electronic properties by chemical modification are discussed.

O 79.2 Fri 11:00 H4

On-surface cyclomerization of oxygen heterocycles: Controlling the ring size by tuning the molecule-surface interaction — •Andreas Dörr[1], Nemanja Kocic[1], Lukas Fromm[2], Vladimir Akhmetov[3], Konstantin Y. Amsharov[3], Andreas Görling[2], and Sabine Maier[1] — [1]Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen (Germany) — [2]Department of Chemistry and Pharmacy, Chair of Theoretical Chemistry, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen (Germany) — [3]Institute for Chemistry, Martin-Luther-University Halle-Wittenberg, 06099 Halle (Germany)

Heterocycles with nitrogen, oxygen, or sulfur atoms are the basic units that incorporate chemical functionalization into carbon scaffolds. For oxygen-doped nanographenes, furan and pyran, having five- and six-membered rings, respectively, are the most common ones. However, their on-surface synthesis via cyclomerization reactions has so far been elusive. Here, we present a low-temperature scanning tunneling microscopy study combined with density functional theory calculations to understand the on-surface synthesis of furan and pyran derivatives from ketone-functionalized precursors on metal surfaces. We first discuss the self-assemblies of the precursors, which are strongly influenced by molecule-surface interactions. Upon annealing, the different intermolecular binding motifs resulted in selective cyclomerization reactions toward furan and pyran moieties.

O 79.3 Fri 11:15 H4

Proximity-induced superconductivity in atomically precise nanographene — •Jung-Ching Liu[1], Rémy Pawlak[1], Xing Wang[2], Philipp D'Astolfo[1], Carl Drechsel[1], Ping Zhou[2], Hongyen Chen[3], Silvio Decurtins[2], Ulrich Aschauer[2], Shi-Xia Liu[2], Wulf Wulfhekel[3], and Ernst Meyer[1] — [1]Department of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel — [2]Department of Chemistry, Biochemistry and Pharmaceutical Sciences, University of Bern, Freiestrasse 3, CH-3012 Bern — [3]Physikalisches Institut, Karlsruhe Institute of Technology, Wolfgang-Gaede-Str. 1, D-76131 Karlsruhe

Atomically precise nanographenes (NGs) can be efficiently synthesized through on-surface reactions. On such designed NGs, topological superconductivity could be fostered via proximity to a s-wave superconductor. However, on-surface synthesis of NGs is still missing on superconducting surfaces [1-3]. To fill the gap, we first fabricate a Ag buffer layer on the Nb(110) superconductor [4], and grow atomically precise NGs on the Ag/Nb substrate using DBBA as the precursor. Through the investigation of low temperature STM/AFM, we demonstrate successful synthesis of polymeric chains and NGs on the superconducting Ag/Nb(110) substrate. We believe our method provides a promising platform for studying the role of topology in the interaction between carbon magnetism and superconductivity [5]. [1]Cai et al., Nature 466, 470-473 (2010) [2]K. A. Simonov et al., Sci. Rep. 8, 3506 (2018) [3]M. Kolmer et al., Science 369, 571-575 (2020) [4]T. Tomanic et al., Phys. Rev. B 94, 220503 (2016) [5]J.-C. Liu et al., arXiv:2202.00460

O 79.4 Fri 11:30 H4

Real-space Imaging of Unprecedented Phenyl Group Migration Reaction on Metal Surfaces — •Zilin Ruan[1,2], Baijin Li[2], Shijie Sun[2], Yong Zhang[2], Lei Gao[3], Jianchen Lu[2], Michael Gottfried[1], and Jinming Cai[2] — [1]Department of Chemistry, Philipps University Marburg, Hans-Meerwein-Straße 4, 35037 Marburg (Germany) — [2]Faculty of Materials Science and Engineering, Kunming University of Science and Technology, Kunming, Yunnan 650093 (China) — [3]Department of Chemistry, Philipps University Marburg, Hans-Meerwein-Straße 4, 35037 Marburg (Germany)

We report on-surface identification and visualization of an unprecedented and universal phenyl group migration reaction of 1,4-dimethyl-2,3,5,6-tetraphenyl benzene (DMTPB)a precursor on Au(111), Cu(111) and Ag(110) substrates by a combination of bond-resolved scanning tunneling microscopy (BR-STM) and density functional theory (DFT) calculations. The phenyl group migration reaction of DMTPB precursor results in formations of variously unprecedented polycyclic aromatic hydrocarbons on the substrates. DFT calculations reveal that the multiple-steps phenyl group migration reactions are facilitated by the radical attack and rearomatization of the DMTPB precursor. Our study provides unprecedented insights into complex surface reaction mechanisms at the single molecule level, which may guide the design of chemical species.

O 79.5 Fri 11:45 H4

Deciphering the intramolecular C-C coupling mechanism of a model aryl radical via bond-level AFM imaging — •Qigang Zhong[1], Jannis Jung[2], Daniel Kohrs[3], Daniel Ebeling[1], Doreen Mollenhauer[2], Hermann A. Wegner[3], and André Schirmeisen[1] — [1]Institute of Applied Physics, Justus-Liebig University Giessen (JLU), Germany — [2]Institute of Physical Chemistry, JLU, Germany — [3]Institute of Organic Chemistry, JLU, Germany

Although on-surface dehalogenative and dehydrogenative C-C coupling has proved to be a versatile and prevailing approach to constructing atomically precise carbon-based nanostructures, understanding of the reaction mechanisms remains limited by the elusive intermediates. Here, we studied the intramolecular cyclodehydrobromination of 1-bromo-8-phenylnaphthalene on Cu(111) and Ag(111) using bond-level atomic force microscopy (BL-AFM). The reaction occurs at room temperature on both metal surfaces, while the reaction rate on Cu(111) is much higher than that on Ag(111) presumably due to the higher catalytic activity of copper. Surface-bound radicals, cyclized intermediates and dehydrogenated product were captured by BL-AFM imaging and verified by DFT calculations, suggesting a multi-step reaction process, i.e. debromination, radical cyclization and dehydrogenation. The large proportion (up to 65 percent) of cyclized intermediates on Cu(111) indicates that dehydrogenation is the rate-determining step, which is corroborated by DFT calculations of activation barriers. To achieve a reasonable activation barrier for dehydrogenation, multiple pathways were theoretically evaluated.

O 79.6 Fri 12:00 H4

Interplay between π-conjugation and exchange magnetism in one-dimensional porphyrinoid polymers — •Kalyan Biswas[1], Maxence Urbani[1], Ana Sánchez-Grande[1], Diego Soler[2], Koen Lauwaet[1], Adam Matěj[2], Pingo Mutombo[2], José M. Gallego[1], Rodolfo Miranda[1], David Écija[1], Pavel Jelínek[2], Tomás Torres[1], and José I. Urgel[1] — [1]IMDEA Nanoscience, Madrid, Spain — [2]CATRIN, Olomouc, Czech Republic

The field of carbon magnetism has gained an increased attention in view of the recent progress made in the synthesis and characterization of open-shell polycyclic aromatic hydrocarbons following a bottom-up synthetic approach. In this work, we introduce an exemplary approach toward the bottom-up fabrication of unprecedented magnetic porphyrinoid-based polymers homocoupled via surface-catalyzed [3 + 3] cycloaromatization of isopropyl substituents studied on Au(111) under ultra-high vacuum conditions. The chemical structure of the polymer, formed by thermal-activated intra- and intermolecular oxidative ring closure reactions followed by controlled tip-induced hydrogen dissociation from the porphyrinoid units, have been clearly elucidated by scanning tunneling microscopy and non-contact atomic force microscopy. Scanning tunneling spectroscopy, complemented by computational investigations reveals the antiferromagnetic singlet ground state (S=0), which display singlet-triplet inelastic excitations observed between spins of adjacent porphyrinoid units only along a specific π-conjugation pathway. We envision that our approach can be a highly relevant in nanoscale spintronic devices.

O 79.7 Fri 12:15 H4

Photoactivation of Azide in SURMOFs — •Jimin Song, Xiaojuan Yu, Manuel Tsotsalas, Alexander Knebel, Alexei Nefedov, Stefan Heissler, Yuemin Wang, and Christof Wöll — Institute of Functional Interfaces, Karlsruhe Institute of Technology,76344, Eggenstein-Leopoldshafen, Germany

The ability to turn the surface activation of metal-organic frameworks (MOFs) is essential for developing advanced MOF applications. Here, we have successfully synthesized a surface-mounted MOF (SURMOF) model system with azide side groups in order to investigate the photoactivation of phenyl azide and its reaction pathways. In situ UHV infrared reflection absorption spectroscopy (IRRAS) was applied to precisely monitor the chemical changes taken place on the surface of highly oriented and crystalline SURMOFs under UV irradiation at different temperatures. Combining with in situ XRD, MS and XPS, a two-step mechanism is proposed including the activation and subsequent reaction of azide with adjacent C=C bonds yielding pyrrole species.

O 79.8 Fri 12:30 H4

Differences in the Intermolecular Interaction of Electron-rich Phosphines on a Metal and a Salt Surface — •Vladimir Lykov[1], Florenz Buss[2], Milica Feldt[3], Karina Morgenstern[1], and Fabian Dielmann[4] — [1]Chair of Physical Chemistry I, Ruhr Universität Bochum, Germany — [2]Institute for Inorganic and Analytical Chemistry, Westphalian Wilhelms University of Münster, Germany — [3]Leibniz-Institut für Katalyse e.V. (LIKAT), Rostock, Germany — [4]Department of General, Inorganic and Theoretical Chemistry, University of Innsbruck, Austria

Electron-rich phosphines are attractive as capture molecules, for instance, for carbon dioxide (CO_2) and sulfur dioxide (SO_2) by forming zwitterionic Lewis base adducts with them [1]. The main focus of this project is understanding the interaction behind this capturing in real space. For this purpose, we use a low-temperature (7 K) scanning tunneling microscope (STM). To understand the influence of the metal surface on this capture process, we compare the molecules adsorbed on Ag(100) to those on NaBr(100). As a first step, the different interaction of phosphines with the salt multilayers and with the metal was investigated by step-wise heating from 43 K to 135 K. The difference in intermolecular interaction on these surfaces will be discussed in this presentation.

[1] Buß F., Röthel M.B., Werra J.A., Rotering P., Wilm L.F.B., Daniliuc C.G., Löwe P., Dielmann F., Chem. Eur. J. 10.1002/chem.202104021 (2021)

O 80: Focus Session: Atomic-Scale Studies of Spins on Surfaces with Scanning Tunneling Microscopy 3

Time: Friday 10:30–12:15 Location: S051

O 80.1 Fri 10:30 S051

Synthesis and Characterization of Triangulene: a novel concept of magnetic nanostructure made of carbon — •Francisco R. Lara[1], Silvia Castro[2], Jeremy Hieulle[1], Manuel Vilas-Varela[2], Alessio Vegliante[1], Niklas Friedrich[1], Lorenz Meyer[1], Unai Uriarte-Amiano[1], Sofia Sanz[3], Dulce Rey[2], Natalia E. Koval[1], Martina Corso[4,3], Emilio Artacho[1,5], Thomas Frederiksen[3], Diego Peña[2], and Jose Ignacio Pascual[1] — [1]CIC nanoGUNE BRTA, Donostia, Spain — [2]Centro Singular de Investigación en Química Biolóxica e Materiais Moleculares, Santiago de Compostela, Spain — [3]Donostia International Physics Center, Donostia, Spain — [4]Centro de Física de Materiales, Donostia, Spain — [5]Cavendish Laboratory, Cambridge, United Kingdom

In this talk, we present the synthesis and characterization of a [3]-triangulene ring of 6 units, the [3]-triangulene nanostar (TNS), and an aza-[5]-triangulene (A5T), on a Au(111) surface.

By means of scanning tunneling microscopy (STM) and spectroscopy (STS) the precise spin states of such carbon-based nanostructures have been studied. Such experimental findings, are supported by calculations from density functional theory (DFT), mean-field Hubbard model (MFH), and Heisenberg model.

Both nanostuctures are achieved thanks to on-surface synthesis. TNS hosts antiferromagnetically coupled spin-1 showing a complex many-body inelastic excitation spectrum. A5T ground state has a spin larger than one. Here the presence of the N heteroatom substitution plays an important role in the spin state on surface.

O 80.2 Fri 10:45 S051

Local access to the Ln-Ln bonding orbital in dimetallofullerene molecular magnets — •Fabian Paschke[1], Tobias Birk[1], Fupin Liu[2], Stanislav M. Avdoshenko[2], Alexey A. Popov[2], and Mikhail Fonin[1] — [1]Department of Physics, Universität Konstanz, 78457 Konstanz — [2]Leibniz Institute for Solid State and Materials Research (IFW Dresden), 01069 Dresden

One of the key players in the single-molecule magnet community are members of the lanthanide (Ln) dimetallofullerene family that combine air-stable chemical robustness, easy functionalization, a large magnetic moment and slow magnetic relaxation up to 28 K [1]. Two Ln^{3+} ions are ferromagnetically coupled by a singly occupied Ln – Ln bonding orbital in the void of a C_{80} fullerene cage. In this talk I will first demonstrate the robust on-surface magnetism of $\{\mathrm{Ln}_2\}$ complexes that show outstanding magnetic blocking temperatures [2,3], an important prerequisite to address its molecular magnetism on a local scale. Subsequently, scanning tunneling spectroscopy unambiguously identifies the unoccupied component of the single-electron Ln – Ln bonding orbital in the spectrum of $\{\mathrm{Dy}_2\}$ on a graphene/Ir(111) surface [4]. This finding outlines a new route how to access the molecular spin dynamics of single $\{\mathrm{Ln}_2\}$ complexes and provides a working point for spin manipulation using chemical doping.

[1] F. Liu *et al.*, Nat. Commun. 10, 571 (2019). [2] F. Paschke *et al.*, Adv. Mater. 2102844 (2021). [3] L. Spree *et al.*, Adv. Funct. Mater. 2105516 (2021). [4] F. Paschke *et al.*, Small 2105667 (2022).

O 80.3 Fri 11:00 S051

indirect spin-readout of rare-earth-based single-molecule magnet with STM — •hongyan chen[1], timo frauhammer[1], timofey balashov[2], gabriel derenbach[1,3], svetlana klyatskaya[4], eufemio moreno-pineda[5], mario ruben[3,4,6], and wulf wulfhekel[1,3] — [1]Physikalisches Institut, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany — [2]Physikalisches Institut, RWTH Aachen, 52074 Aachen, Germany — [3]Institute for Quantum Materials and Technologies, KIT, 76021 Karlsruhe, Germany — [4]Institute of Nanotechnology, KIT, 76021 Karlsruhe, Germany — [5]Departamento de Química-Física, Escuela de Química, Facultad de Ciencias Naturales, Exactas y Tecnología, Universidad de Panamá 0824, Panama — [6]Centre Européen de Sciences Quantiques (CESQ) in the Institut de Science et d'Ingénierie Supramoléculaires (ISIS), 8 allée Gaspard Monge BP 70028, 67083 Strasbourg Cedex, France

Rare-earth based SMMs are promising candidates for magnetic information storage as their large magnetic moments are carried by localized 4f electrons. However, this in turn hampers a direct readout of the moment. Here, we present the indirect readout of the Dy moment in DyPc_2 molecules on Au(111) using mK-STM. Because of an unpaired electron on the Pc ligand, the molecules show a Kondo resonance that is, however, split by the ferromagnetic exchange interaction between the unpaired electron and the Dy spin. Using spin-polarized STS, we read out the Dy spin as a function of the applied magnetic field, exploiting the spin polarization of the exchange-split Kondo state.

O 80.4 Fri 11:15 S051

Yu-Shiba-Rusinov states of Fe dimers on $\mathbf{NbSe_2}$ — •Lisa M. Rütten[1], Eva Liebhaber[1], Harald Schmid[1], Gaël Reecht[1], Kai Rossnagel[2,3], Felix von Oppen[1], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany — [2]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24118 Kiel, Germany — [3]Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany

Unpaired adatom spins on superconductors interact with the Cooper pairs of the substrate and induce Yu-Shiba-Rusinov (YSR) states inside the superconducting gap. These can be probed by scanning tunneling spectroscopy at the single-atom scale. $2H$-NbSe_2 is a superconducting, layered van der Waals material, where the YSR wave functions of magnetic impurities extend over several nanometers. This provides a wide range of adatom spacings over which their interaction is sufficiently strong to be potentially observed as a splitting in the tunneling spectra. In addition to superconductivity, $2H$-NbSe_2 hosts an incommensurate charge-density wave (CDW). The imposed variation of the local density of states leads to shifts in the energy of the YSR states and alters the spatial symmetry of YSR wave functions. Here, we arrange Fe atoms on $2H$-NbSe_2 using the tip of a scanning tunneling microscope and realize dimers with different spacings and symmetries. We investigate the influence of spacing and position with respect to the CDW on the interaction between the YSR states.

O 80.5 Fri 11:30 S051

Atomic manipulation of spin structures on the β-$\mathbf{Bi_2Pd}$ Superconductor — Cristina Mier[1], Divya Jyoti[1,2], Jiyoon Hwang[3], Jinkyung Kim[3], Yujeong Bae[3], Andreas Heinrich[3], Nicolas Lorente[1,2], and •Deung-Jang Choi[1,2,4] — [1]Centro de Fisica de Materiales, CFM/MPC (CSIC-UPV/EHU), Paseo Manuel de Lardizabal 5, 20018 Donostia-San Sebastian, Spain — [2]Donostia International Physics Center (DIPC), Paseo Manuel de Lardizabal 4, 20018 Donostia-San Sebastian, Spain — [3]Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS), Seoul 03760, South Korea — [4]Ikerbasque, Basque Foundation for Science, 48013 Bilbao, Spain

Recently, the introduction of impurity states in the superconducting gap has received a lot of attention. Indeed, the search of a new superconducting state called topological superconductivity is strongly based in the combination of doping classical (s-wave) superconductors with magnetic impurities that arrange spins in a chiral fashion. We present the first results of controlled single-atom manipulation to assemble a chain of Cr atoms on a β-$\mathrm{Bi}_2\mathrm{Pd}$ superconductor [1,2]. Such magnetic impurities on different substrates allow us to explore many-body effects and exotic phenomena in different experimental spin systems giving an understanding on the parameters on each system.

[1] C. Mier et al., Atomic Manipulation of In-gap States on the β-$\mathrm{Bi}_2\mathrm{Pd}$ Superconductors, Phys. Rev. B 104 (4), 045406 (2021). [2] Cristina Mier, Deung-Jang Choi and Nicolás Lorente, Phys. Rev. B 104 (24), 245415 (2021).

O 80.6 Fri 11:45 S051

Theory of transport between superconducting states bound to magnetic impurities — •Ciprian Padurariu[1], Haonan Huang[2], Björn Kubala[1,3], Christian R. Ast[2], and Joachim Ankerhold[1] — [1]Institute for Complex Quantum Systems and IQST, Ulm University, Ulm, Germany — [2]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany — [3]Institute of Quantum Technologies, German Aerospace Center (DLR), Ulm, Germany

The realization of the Majorana chain [1], a 1D-chain of superconducting states bound to magnetic impurities, suggests that Majorana edge states can be probed using the superconducting tunneling microscope

(STM). Recently, we have developed an ideal tool to probe and manipulate the edge states of a Majorana chain. It consists of a mK-STM with its own in-gap Yu-Shiba-Rusinov (YSR) state created by a magnetic impurity on the tip. With this device we have studied the sharp resonant transport between the YSR state on the tip and another YSR on the sample, and have developed the theory [2].
Here, we summarize and expand the theory of YSR-YSR tunneling to phenomena that occur when one YSR state is close to zero energy, near its quantum phase transition. If the zero-energy state sits at the edge of a Majorana impurity chain, theory predicts that the topological edge state will transfer from the chain to the tip.
[1] S. Nadj-Perge, *et al.*, "Observation of Majorana fermions in ferromagnetic atomic chains on a superconductor", Science **346**, 602 (2014).
[2] H. Huang, *et al.*, "Tunneling dynamics between superconducting bound states at the atomic limit", Nat. Phys. **16**, 1227 (2020).

O 80.7 Fri 12:00 S051

Many-body Excitations of a Quantum Spin on a Proximitized Superconductor — Stefano Trivini[1], •Jon Ortuzar[1], Katerina Vaxevani[1], Jingchen Li[2], Ane Garro[1], F. Sebastian Bergeret[3], Miguel.A Cazalilla[4,5], and José Ignacio Pascual[1,5] — [1]CICnanoGUNE, San Sebastian,Spain — [2]School of Physics, Sun Yat-sen University, Guangzhou 510275, China — [3]Centro de Fìsica de Materiales (CFM-MPC), 20018 San Sebastiàn, Spain — [4]Donostia International Physics Center (DIPC), 20018 San Sebastian, Spain — [5]Ikerbasque, Basque Foundation for Science, 48013 Bilbao, Spain

In magnetic molecules intrinsic magnetic anisotropy breaks spin degeneracy, allowing inelastic spin excitations that can be protected by a superconducting gap. The coupling of the spin to the superconductor induces Yu-Shiba-Rusinov (YSR) states, detected by scanning tunnelling microscopy (STM) as long-lived quasiparticles excitations inside the superconducting gap.

Here, we observe the signature of both described excitations in an Fe-porphyrin adsorbed on a Au/V(100) proximitized superconducting surface. We found that the STM tip affects in a similar way both in-gap and out-gap states, hint that they are correlated. Solving an effective Hamiltonian of the system, we describe the observed signals as multiple excitations of entangled states formed by the molecular spin and superconductor.

O 81: 2D Materials 4: Heterostructures

Time: Friday 10:30–12:00 Location: S052

O 81.1 Fri 10:30 S052

Sub-angstrom non-invasive imaging of microstructures in 2D hybrid perovskites — •Shayan Edalatmanesh[1,3], Mykola Telychko[2], Kai Leng[2], Ibrahim Abdelwahab[2,4], Na Guo[5], Chun Zhang[5], Jesus Mendieta-Moreno[1], Matyáš Nachtigall[1], Jing Li[4], Kian Ping Loh[2], Pavel Jelínek[1,3], and Jiong Lu[2,4] — [1]Institute of Physics, The Czech Academy of Sciences, Prague, Czech Republic — [2]Department of Chemistry, NUS, Singapore — [3]RCPTM, Palacky University, Olomouc, Czech Republic — [4]CA2DM, NUS, Singapore — [5]Department of Physics, NUS, Singapore

Organic-inorganic hybrid two dimensional (2D) Ruddlesden-Popper perovskites (RPPs), made of soft insulating organic layers sandwiched between conducting inorganic frameworks, have recently gained a great deal of attention as candidates for the next generation of optoelectronic devices. To gain an understanding of the cooperative lattice relaxation governing the optoelectronic properties of 2D RPPs, we present sub-angstrom resolution imaging of both soft organic layers and inorganic framework in a prototypical 2D lead-halide RPP crystal using a tip-functionalized Scanning Tunneling Microscopy (STM), non-contact Atomic Force Microscope (ncAFM) and Kelvin Probe Force Microscopy (KPFM) corroborated by theoretical simulations, namely Density Functional Theory (DFT)[1]. We unveil the overall twin-domain composition of the RPP crystal, with alternating quasi-one-dimensional electron and hole-channels at neighboring twin-boundaries, possibly responsible for the long-distance exciton transport in RPPs. Reference: [1] https://arxiv.org/abs/2109.05878

O 81.2 Fri 10:45 S052

Electronic Structure of Quasi-Freestanding WS_2/MoS_2 Heterostructures — Borna Pielić[1], Dino Novko[1], Iva Šrut Rakić[1], Jiaqi Cai[2], Marin Petrović[1], Alice Bremerich[2], Robin Ohmann[2], Nataša Vujičić[1], Mario Basletić[3], Marko Kralj[1], and •Carsten Busse[2] — [1]Institute of Physics, Zagreb, Croatia — [2]Universität Siegen, Germany — [3]University of Zagreb, Croatia

Quasi-freestanding heterostructures of semiconductiong two-dimensional materials with sharp interfaces, large built-in electric field, and narrow depletion region widths are proper candidates for the future design of electronic and optoelectronic devices.

Here, we epitaxially grow lateral WS_2-MoS_2 and vertical WS_2/MoS_2 heterostructures on graphene under UHV conditions. By means of scanning tunneling spectroscopy (STS), we examine the electronic structure of monolayer MoS_2, WS_2, and WS_2/MoS_2 vertical heterostructure. Moreover, we investigate band bending in the vicinity of the narrow one-dimensional interface of the WS_2-MoS_2 lateral heterostructure. Density functional theory (DFT) is used for the calculation of the band structures, as well as for the density of states maps at the interfaces. For the WS_2-MoS_2 lateral heterostructure, we find type-II band alignment and determine the corresponding depletion regions, charge densities, and the electric field at the interface.

O 81.3 Fri 11:00 S052

Quantum spin Hall edge states and interlayer coupling in twisted bilayer WTe_2 — •Felix Lüpke[1], Dacen Waters[2], Anh Pham[3], Jiaqiang Yan[4], David G. Mandrus[5], Panchapakesan Ganesh[3], and Benjamin M. Hunt[6] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Physics, University of Washington — [3]Center for Nanophase Materials Sciences, Oak Ridge National Lab — [4]Materials Science and Technology Division, Oak Ridge National Lab — [5]Department of Materials Science and Engineering, University of Tennessee — [6]Physics, Carnegie Mellon University

The quantum spin Hall (QSH) effect, characterized by topologically protected spin-polarized edge states, was recently demonstrated in monolayers of the transition metal dichalcogenide (TMD) WTe_2. However, the robustness of this topological protection remains largely unexplored in van der Waals heterostructures containing one or more layers of a QSH insulator. In this work, we use scanning tunneling microscopy and spectroscopy (STM/STS), to study twisted bilayer (tBL) WTe_2 and compare it to topologically trivial natural bilayer. By comparing our experimental observations to first principles calculations, we conclude that the twisted bilayers are weakly coupled, preserving the QSH states and preventing back scattering.

O 81.4 Fri 11:15 S052

Real-time TD-DFTB simulations and modeling of Fano-induced transparency in molecular van der Waals Heterostructures — •Carlos R. Lien-Medrano[1], Franco P. Bonafé[2], Chi Yung Yam[3], Carlos-Andres Palma[4], Cristián G. Sánchez[5], and Thomas Frauenheim[1] — [1]BCCMS, Uni-Bremen, Germany — [2]MPSD, Hamburg, Germany — [3]CSAR, Shenzhen, P. R. China — [4]IOP, Beijing, P.R. China — [5]UNCuyo, Mendoza, Argentina

While gating and doping in two-dimensional (2D) materials is well-known, the physics of photosensitizing and advanced optical properties have not been fully investigated, especially in the context of molecular vdW heterostructures (MVHs), that is, regular monolayer stacks on 2D materials. In a recent work [1], we employed an adapted Gersten-Nitzan (two point dipoles) model and real time time-dependent density functional tight-binding to study the optoelectronics of self-assembled monolayers on graphene nanoribbons. We found Fano resonances that cause electromagnetic induced opacity and transparency and reveal an additional incoherent process leading to interlayer exciton formation with a characteristic charge transfer rate. These results showcase hybrid van der Waals heterostructures as paradigmatic 2D optoelectronic stacks, featuring tunable Fano optics and unconventional charge transfer channels. Our findings open a path for improved design of modular multilayer organic photovoltaic devices.

[1] Lien-Medrano, C. R., et al. Fano Resonance and Incoherent Interlayer Excitons in Molecular van der Waals Heterostructures. Nano Letters (2022), 22(3), 911-917.

O 81.5 Fri 11:30 S052

High-throughput stacking reveals emergent and switchable properties of 2D van der Waals bilayers — •Sahar Pakdel, Asbjørn Rasmussen, Mads Kruse, Alireza Taghizadeh, Thomas Olsen, and Kristian Sommer Thygesen — CAMD, Computational Atomic-Scale Materials Design, Department of Physics, Technical University of Denmark, 2800 Kgs. Lyngby Denmark

Stacking atomically thin two-dimensional monolayers into van der Waals (vdW) heterostuctures offer new opportunities to tune physical properties of 2D materials. Here we provide a systematic ab initio-based study of homo-bilayers created by stacking several hundreds of stable monolayers containing up to 10 atoms per unit cell. We investigate all configurations commensurate with the primitive cell and verify our approach by comparing our stacking orders with available bulk compounds. For the stable bilayers within a 3 meV/Å^2 binding energy distance from the most stable configuration, we calculate a range of electronic and magnetic properties. We explore switchable properties in bilayer pairs related with a slide vector. Our work is a step towards rational design of layered vdW materials and contributes to the systematisation of 2D materials. Our results will be available online and integrated with the Computational 2D Materials Database (C2DB) which allows for comparison between mono- and bilayer properties.

O 81.6 Fri 11:45 S052

1D p-n junction electronic and optoelectronic devices from transition metal dichalcogenide lateral heterostructures grown by one-pot chemical vapor deposition synthesis — •E. Najafidehaghani[1], Z. Gan[1], A. George[1], T. Lehnert[2], G. Q. Ngo[3], C. Neumann[1], T. Bucher[3], I. Staude[3], D. Kaiser[1], T. Vogl[3], U. Hübner[4], U. Kaiser[2], F. Eilenberger[3], and A. Turchanin[1] — [1]Friedrich Schiller University Jena, Institute of Physical Chemistry, Germany — [2]Ulm University, Central Facility of Materials Science Electron Microscopy, Germany — [3]Friedrich Schiller University Jena, Institute of Applied Physics, Germany — [4]Leibniz Institute of Photonic Technology (IPHT), Germany

Lateral heterostructures (LH) of dissimilar monolayer transition metal dichalcogenides provide great opportunities to build 1D in-plane p-n junctions for sub-nanometer thin low-power electronic, optoelectronic, optical, and sensing devices. Electronic and optoelectronic applications of such p-n junction devices fabricated using a scalable chemical vapor deposition process yielding $MoSe_2$-WSe_2 LHs are reported here. Their growth is achieved by in situ controlling the partial pressures of the oxide precursors by a two-step heating protocol. The grown LHs are characterized structurally and optically using optical microscopy, Raman spectroscopy, and photoluminescence spectroscopy. High-resolution transmission electron microscopy further confirms the high-quality 1D boundary between $MoSe_2$ and WSe_2 in the LH. p-n junction devices are fabricated from these LH and their applicability solar cells, photodetectors, and electroluminescent emitters are demonstrated.

O 82: Electronic Structure of Surfaces 2

Time: Friday 10:30–12:15 Location: S053

O 82.1 Fri 10:30 S053

Electron- and Hole-Like Transport in Shockley Type Surface States Detected by MONA — •Andreas Christ, Markus Leisegang, Patrick Härtl, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

In recent years, we have established the STM-based molecular nanoprobe (MONA) technique to detect the transport of hot charge carriers over distances of a few nanometers [1,2]. In short, MONA uses a charge carrier-driven molecular switching events, such as a tautomerization or rotation, to detect currents injected a few nanometers away. Earlier experiments performed on Ag(111) showed that the surface state characteristic for fcc(111) surfaces facilitates effective charge transport between the charge injection point at the tip position and a single phthalocyanine (HPc) detector molecule [3]. Since, however, the energy threshold for tautomerization of HPc exceeds the energy onset of the Ag(111) surface state ($E = -63\,\mathrm{meV}$), hole transport remained inaccessible. In order to investigate the influence of the band structure on the propagation of hot charge carriers we compare results of MONA experiments performed on Ag(111) with the isoelectronic Cu(111) surface. Our results reveal that due to the lower surface state onset in Cu(111) at $E = -440\,\mathrm{meV}$ hole-like charge transport can also be detected.

[1] M. Leisegang *et al.*, Nano Lett. **18**, 2165-2171 (2018)
[2] M. Leisegang *et al.*, Phys. Rev. Lett. **126**, 146601 (2021)
[3] J. Kügel *et al.*, Nano Lett. **17**, 5106 (2017)

O 82.2 Fri 10:45 S053

Exploring polaron stability and defect structures at the $Li_4Ti_5O_{12}$ (LTO) surface: A combined theoretical and experimental approach — •Yu-Te Chan[1], Matthias Kick[2], Cristina Grosu[2,3], Christoph Scheurer[1], and Harald Oberhofer[2] — [1]Fritz-Haber-Institut — [2]TU München — [3]IEK-9, FZ Jülich

Spinel $Li_4Ti_5O_{12}$ (LTO) is a promising anode material for next-generation all-solid-state Li-ion batteries (ASSB) due to its "zero strain" charge/discharge behavior. Pristine, white LTO possesses poor ionic and electronic conductivity. Through tailoring the sintering protocol, one can produce oxygen vacancies, resulting in a performant, blue LTO material. Polarons induced by oxygen vacancies have been proposed as one of the origins of the higher electronic conductivity. However, detailed knowledge about polaron stability, distribution, and dynamics in LTO bulk and surface has been lacking. By performing *Hubbard corrected density functional theory* (DFT+U) calculations we are able to show that in fact polaron formation and a possible polaron hopping mechanism can not only play a significant role in enhancing electronic conductivity but boost Li^+ diffusion nearby through lowering the hopping barrier, in line with the experimentally observed improved conductivities.[1,2] In combination with positron lifetime spectroscopy data and theoretical positron lifetimes, we arrive at a rather complete picture of the bulk vs. surface defect chemistry in LTO particles and the resulting mixed ionic electronic conductivity.

[1] M. Kick et al.,J. Phys. Chem. Lett. 11 (2020), 2535
[2] M. Kick et al., ACS Appl. Energy Mater 4 (2021), 8583

O 82.3 Fri 11:00 S053

Spin-polarized VLEED from Au(111): Surface sensitivity of the scattering process — •Christoph Angrick[1], Jürgen Braun[2], and Markus Donath[1] — [1]Westfälische Wilhelms-Universität Münster — [2]Ludwig-Maximilians Universität München

Low-energy electron diffraction from Au(111) shows the well-known threefold symmetry of the diffracted electron beams despite the sixfold symmetry of the surface layer. This is due to the influence of the second and deeper layers and the probing depth of the electrons. In this work, we investigated Au(111) with spin-polarized very-low-energy electron diffraction (VLEED) [1,2] experimentally and theoretically. We monitor the reflected specular beam at a fixed polar angle of incidence of $\Theta = 45^\circ$ while the azimuthal orientation of the crystal is varied. This puts the surface sensitivity of the VLEED scattering process to a test.

Our results show that the electron reflection and the spin-orbit-induced reflection asymmetry along $\overline{\Gamma}\,\overline{M}$ and $\overline{\Gamma}\,\overline{M}'$ are equivalent. The observed sixfold symmetry suggests a sensitivity to one atomic layer only. At azimuth angles deviating from the high-symmetry directions $\overline{\Gamma}\,\overline{M}$ and $\overline{\Gamma}\,\overline{M}'$, however, the VLEED signal from Au(111) shows a threefold symmetry. To reveal the origin of this effect, we varied the parameters in the calculation. The results indicate a non-negligible influence of the second atomic layer in the VLEED scattering process.

[1] Burgbacher *et al.*, Phys. Rev. B **87**, 195411 (2013)
[2] Angrick *et al.*, J. Phys.: Condens. Matter **33**, 115001 (2020)

O 82.4 Fri 11:15 S053

The quantum corral: Pertubation by adatoms and bonding description by LCAO — •Andreas Bereczuk[1], Martin Steinau[1], Fabian Stilp[2], Franz Josef Giessbil[2], and Klaus Richter[1] — [1]Institut of Theoretical Physics, University of Regensburg, Germany — [2]Institute of Experimental and Applied Physics, University of Regensburg, Germany

The quantum corral, first investigated in 1993 [1] consists of a circle of

48 iron atoms placed on a copper surface and gives rise to a standing wave pattern of the local charge density (LDOS). Afterwards a plurality of different confinement shapes have been investigated [2,3]. We revisited this structure in [4] using atomic force microscope (AFM) and scanning tunneling microscopy (STM). A tight-binding model [5] provides a LDOS in consistency with STM measurements for the original and the perturbed quantum corral. We further consider weak bonds between the AFM tip and the artifical atom, indicated by a widely spread LDOS, by using linear combinations of atomic orbitals (LCAO).
[1] M. F. Crommie et al., Science 262, 218 (1993)
[2] M. Crommie et al., Physica D: Nonlinear Phenomena 83, 98 (1995)
[3] E. Heller et al., Nature 369, 464 (1994)
[4] F. Stilp et al., Science 372, 1196 (2021)
[5] C. W. Groth et al., New J. Phys. 16, 063065 (2014)

O 82.5 Fri 11:30 S053

Single-Molecule Ultrafast Fluorescence-Detected Pump–Probe Microscopy — •Daniel Fersch[1], Pavel Malý[1,2], Jessica Rühe[3], Victor Lisinetskii[1], Matthias Hensen[1], Frank Würthner[3,4], and Tobias Brixner[1,4] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Faculty of Mathematics and Physics, Charles University, Ke Karlovu 5, 121 16 Prague, Czech Republic — [3]Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [4]Center for Nanosystems Chemistry (CNC), Universität Würzburg, Theodor-Boveri-Weg, 97074 Würzburg, Germany

The spectroscopic signatures of bulk samples can differ vastly from those of the respective single molecules. In particular, access to the femtosecond dynamics of single molecules remains a large experimental challenge. Here, we present a novel setup consisting of a spectrally tunable femtosecond laser source and a scanning confocal fluorescence microscope with fully reflective excitation geometry and single-molecule sensitivity. Using a phase-stable interferometer we create a pulse pair with variable time delay to measure the molecular fluorescence excitation spectrum by means of Fourier-transform spectroscopy. By exciting the molecule with an additional prior pump pulse, we gain access to the dynamics of the excited state as a function of the pump–probe delay, resulting in a fluorescence-detected pump–probe spectrum. We have obtained first results on single terrylene bisimide molecules and compare them to a spincoated thin film.

O 82.6 Fri 11:45 S053

Anomalies at the Dirac point in doped graphene (B, N, BN) — •Sangeeta Thakur[1,2], Arindam Pramanik[3], Bahadur Singh[3], Philip Willke[4], Martin Wenderoth[4], Hans Hofsäss[5], Giovanni Di Santo[1], Luca Petaccia[1], and Kalobaran Maiti[3] — [1]Elettra Sincrotrone Trieste, Strada Statale 14 km 163.5, 34149 Trieste, Italy — [2]Freie Universität Berlin, Institut für Experimentalphysik Arnimallee 14, 14195 Berlin, Germany — [3]Department of Condensed Matter Physics and Materials Science, Tata Institute of Fundamental Research,Homi Bhabha Road, Colaba, Mumbai 400005, India — [4]IV. Physikalisches Institut, Georg-August-Universität Göttingen, 37077 Göttingen, Germany — [5]II. Physikalisches Institut, Georg-August-Universität Göttingen, 37077 Göttingen, Germany

The changes in the electronic properties of graphene on SiC, induced by different atomic species, B, N and BN, substituted, via low energy (25eV) ion bombardment were investigated via angle-resolved photoemission spectroscopy. The anomalies at the Dirac point for B, N, and BN doped graphene are attributed to the spectral width arising from the lifetime and momentum broadening in the experiments. An energy gap at the Dirac point of graphene is not observed even after 5 % of B and N substitution [1]. These results will provide new insight to tune the carrier properties of graphene while keeping the Dirac fermionic properties protected, which is important for exploring its technological applications.
[1] A. Pramanik, Sangeeta Thakur et.al. PRL 128, 166401 (2022).

O 82.7 Fri 12:00 S053

Interplay of intrinsic and extrinsic states in pinning and passivation of m-plane facets of GaN n-p-n junctions — •Lars Freter[1,2], Yuhan Wang[1,2], Michael Schnedler[1], Jean-François Carlin[3], Raphaël Butté[3], Nicolas Grandjean[3], Holger Eisele[4], Rafal Edward Dunin-Borkowski[1,5], and Philipp Ebert[1] — [1]Peter Grünberg Institut, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany — [2]Lehrstuhl für Experimentalphysik IV E, RWTH Aachen University, 52056 Aachen, Germany — [3]Institute of Physics, Ecole Polytechnique Fédérale de Lausanne (EPFL), 1015 Lausanne, Switzerland — [4]Technische Universität Berlin, Institut für Festkörperphysik, Hardenbergstr. 36, 10623 Berlin, Germany — [5]Ernst Ruska-Centrum, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany

Intrinsic and extrinsic pinning and passivation of *m*-plane cleavage facets of GaN *n*-*p*-*n* junctions were investigated by cross-sectional scanning tunneling microscopy and spectroscopy. On freshly cleaved and clean *p*-type GaN($10\bar{1}0$) surfaces, the Fermi level is found to be extrinsically pinned by defect states, whereas *n*-type surfaces are intrinsically pinned by the empty surface state. For both types of doping, air exposure reduces the density of pinning states and shifts the pinning levels toward the band edges. These effects are assigned to water adsorption and dissociation, passivating intrinsic and extrinsic gap states. The revealed delicate interplay of intrinsic and extrinsic surface states at GaN($10\bar{1}0$) surfaces is a critical factor for realizing flatband conditions at sidewall facets of nanowires exhibiting complex doping structures.

O 83: Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 4

Time: Friday 10:30–13:00 Location: S054

O 83.1 Fri 10:30 S054

Alchemical machine learning for high entropy alloys — •Nataliya Lopanitsyna, Guillaume Fraux, and Michele Ceriotti — École Polytechnique Fédérale de Lausanne, Switzerland

High entropy alloys (HEAs) are a class of metallic materials composed of five or more principal elements. Interest in HEAs has grown over the last decades due to their exceptional structural and mechanical properties. HEAs are particularly challenging for atomistic modeling. Machine-learning (ML) models have emerged as a promising alternative to inaccurate empirical forcefields and very demanding first-principles simulations, with the ability to deliver the accuracy of first principle methods with lower computational resources. However, the complexity of ML models grows exponentially with the number of different elements due to the unfavourable scaling of their associated feature space sizes, limiting the chemical diversity of the systems tackled thus far. To address the problems arising from the high feature space dimensionality, first, we propose a chemical embedding compression scheme to reduce the dimensionality of the feature space required for multi-component systems, based on the framework of Willatt et al [Phys. Chem. Chem. Phys., 2018], and implemented in PyTorch. Second, we generate a dataset of several thousands configurations, assembled from 25 d-block elements, which aims to represent cross-elemental interactions, evaluating their energies and forces at the DFT level. We demonstrate the effectiveness of the alchemical ML model in learning the energetics of this extremely diverse dataset, and provide showcase calculations of the properties of some realistic HEA compositions.

O 83.2 Fri 10:45 S054

Stacking the odds: Distribution-biased generative deep learning for molecular design — •Joe Gilkes[1,2], Julia Westermayr[1], Rhyan Barrett[3], and Reinhard J. Maurer[1] — [1]Department of Chemistry, University of Warwick, UK — [2]HetSys CDT, University of Warwick, UK — [3]Warwick Mathematics Institute, UK

Organic electronics applications pose a number of often competing requirements on molecular design that are hard to satisfy by conventional synthesis. Devices such as organic light-emitting diodes (OLEDs) must exhibit closely aligned optoelectronic properties, yet their component molecules must be easily synthesisable and stable. The odds of finding suitable molecules when drawing random samples from chemical space are still too low for targeted design of candidate systems for

OLED devices. We develop an automated molecular design approach based on iterative biasing of a generative deep learning model. In successive iterations, the output of this model is filtered with a deep learning surrogate model of electronic structure and then used to retrain the generative model with a bias. This enables us to create models that are progressively biased towards, e.g., higher ionisation potentials, or smaller fundamental gaps. We also demonstrate how we can bias towards multiple properties simultaneously by filtering our results with the SCScore model for synthetic complexity. This creates more synthetically viable molecules while still meeting optoelectronic requirements. Our approach efficiently creates novel molecules with tuned optoelectronic properties. Clustering analysis reveals trends in bonding patterns which can be utilised in molecular design.

O 83.3 Fri 11:00 S054

Machine learning TCP phases with domain knowledge of the interatomic bond — •Mariano Forti, Alesya Burakovskaya, Ralf Drautz, and Thomas Hammerschmidt — ICAMS, Ruhr-Universität Bochum, Universitätsstr. 150, 44801 Bochum, Germany.

The understanding of the precipitation of topological close packed (TCP) phases in single-crystal superalloys is of central importance for the design of these materials for high-temperature applications. However, the structural complexity of these intermetallic compounds and the chemical complexity of the superalloys with typically N=5-10 elements hampers the exhaustive sampling of chemical space by density-functional theory (DFT) calculations. For example, the computation of the convex hull of the R phase with 11 inequivalent lattice sites would require N^{11} DFT calculations in an N-component system. We overcome this computational limitation by combining machine learning (ML) techniques with descriptors of the local atomic environment of the TCP phases. We present descriptors that are derived from bond order potential (BOP) theory which retain domain knowledge of the interatomic interaction from tight-binding Hamiltonians. We demonstrate that these descriptors enable us to predict the structural stability of TCP phases with simple regression algorithms. We apply this methodology to several systems with experimental evidence of R phase formation.

O 83.4 Fri 11:15 S054

Ab initio random structure search of organic molecules at substrates — •Dmitrii Maksimov[1,2] and Mariana Rossi[1,2] — [1]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [2]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

Finding stable structures of molecular adsorbates (in isolation or forming layers) from calculations is challenging, exacerbated when the adsorbates are flexible. To make matters more complicated, in these situations, it is often difficult to find good and cheap potentials of such complex interfaces that allow a thorough and reliable global search of the structural space. To make this problem tractable with *ab initio* potentials, we present a random global geometry optimization package that can explicitly take into account the internal degrees of freedom of molecules, their position and orientation with respect to fixed surroundings, as well as periodic boundary conditions [1]. Electronic structure calculations and local geometry optimizations are performed through a connection to the ASE software [2], making it possible to interface this algorithm with various codes. To increase the efficiency of geometry optimizations, we introduce a framework to construct initial approximate Hessians for BFGS algorithms that are specially tailored to accelerate the relaxation of van der Waals bonded structures and handle large structural changes. We showcase the algorithm for the adsorption of di-L-alanine at Cu(110). [1] https://github.com/sabia-group/gensec [2] Larsen et. al., J. Phys.: Condens. Matter **29**, 273002 (2017).

O 83.5 Fri 11:30 S054

Active learning and element-embedding approach in neural networks for infinite-layer versus perovskite oxides — Armin Sahinovic and •Benjamin Geisler — Fakultät für Physik, Universität Duisburg-Essen

The observation of superconductivity in $NdNiO_2$ films on $SrTiO_3(001)$ by Li *et al.* [1] has sparked considerable interest in the materials class of infinite-layer oxides. Here we combine first-principles simulations and active learning of neural networks to explore formation energies of oxygen vacancy layers, lattice parameters, and their statistical correlations in infinite-layer versus perovskite oxides across the periodic table, and place the superconducting nickelate and cuprate families in a comprehensive context. Neural networks accurately predict these observables, which act as a fingerprint of the complex reduction reaction, using only a fraction of the data for training. Unbiased by external knowledge, element embedding autonomously identifies chemical similarities between the individual elements in line with human knowledge. Active learning renders the training highly efficient, based on the physical concepts of entropy and information, and provides systematic accuracy control [2]. We recently applied this concept also to nitrides and fluorides [3]. This exemplifies how AI may assist on the quantum scale in discovering novel materials with optimized properties.

[1] D. Li *et al.*, Nature **572**, 624 (2019)

[2] A. Sahinovic and B. Geisler, PR Research **3**, L042022 (2021)

[3] A. Sahinovic and B. Geisler, J. Phys.: Condens. Matter **34**, 214003 (2022)

O 83.6 Fri 11:45 S054

Indirect learning interatomic potential models for accelerated materials simulations — •Joe D. Morrow and Volker L. Deringer — Department of Chemistry, Inorganic Chemistry Laboratory, University of Oxford, Oxford OX1 3QR, United Kingdom

Machine learning (ML) based interatomic potentials are emerging tools for materials simulations but require a trade-off between accuracy and speed. We show how one ML potential can be used to train another: we use an existing, accurate, but more computationally expensive model to generate reference data (labels and locations) for a series of much faster “indirectly-learned” potentials. Extensive reference datasets can be easily generated without the need for quantum-mechanical reference computations at the indirect learning stage, and we find that the additional data significantly improve the predictions of fast potentials with less flexible functional forms.

We apply the technique to disordered silicon, including a simulation of vitrification and polycrystalline grain formation under pressure with a system size of a million atoms. When comparing indirectly learned potentials to models learned directly from a DFT-labelled database, the latter make unphysical predictions for large systems (10^5 atoms) that are not apparent in smaller simulations ($\leq 10^4$ atoms). This emphasises the importance of carefully validating ML potentials *chemically*, not only via numerical error measures. Our work provides conceptual insight into the machine learning of interatomic potential models, and it suggests a route toward accelerated simulations of nanostructured materials.

O 83.7 Fri 12:00 S054

Predicting hot electrons free energies from ground-state data — •Chiheb Ben Mahmoud, Federico Grasselli, and Michele Ceriotti — EPFL, Lausanne, Switzerland

Machine-learning potentials, while extremely successful in describing the stability of condensed phases, are usually trained on ground-state electronic-structure calculations depending exclusively on the atomic positions and ignoring the electronic temperature. Hence, they are limited in their ability to describe hot electrons. We introduce a rigorous framework to calculate the finite-temperature electron free energy based exclusively on ground-state total energy and electronic density of states, while allowing to sample on-the-fly the electronic free energy at any temperature [1]. Our physically-motivated approach facilitates modeling material properties in extreme conditions with a fraction of the usual cost. We demonstrate it by computing the equation of state and heat capacity of hydrogen in planetary conditions. This approach demonstrates the impact of a universal model describing structural and electronic properties inexpensively and its ability to enable more accurate and predictive materials modeling and design.

[1]: C Ben Mahmoud, F Grasselli, M Ceriotti*- arXiv preprint arXiv:2205.05591, 2022

O 83.8 Fri 12:15 S054

Machine Learning the RPA density-density response function — •Mario Zauchner, Johannes Lischner, and Andrew Horsfield — Imperial College London, London, United Kingdom

Clusters and nanoparticles are used in a variety of scientific and industrial applications, including optoelectronics, photocatalysis, single electron transistors and medical imaging, among others. Electronic excitations often play a key role in these applications, but theoretical techniques for calculating excited-state properties of materials, such as the first-principles GW/Bethe-Salpeter method, are typically limited to very small systems. A key bottleneck of such excited-state calculations of clusters and nanoparticles is the determination of the static density-density response function, which is often calculated using a sum-over-states technique. In this talk, we present a technique to decompose the density-density response function into atomic contributions. This can be achieved by exploiting the locality of the density-density response function in non-metallic systems. These atomic contributions can then be used to train a machine-learning model using a set of structural features with the same rotational symmetry as the atomic response functions, thus allowing direct prediction of the density-density response function using only structural information.

O 83.9 Fri 12:30 S054

MD-based Raman Spectra using Machine Learning — •Manuel Grumet[1], Karin S. Thalmann[1], Tomáš Bučko[2,3], and David A. Egger[1] — [1]Department of Physics, Technical University of Munich, Garching, Germany — [2]Comenius University in Bratislava, Slovakia — [3]Slovak Academy of Sciences, Slovakia

Theoretical calculations of Raman spectra based on molecular dynamics (MD) trajectories allow to directly incorporate both anharmonic and temperature-dependent effects and thus yield more realistic spectra compared to a phonon-based approach [1]. The spectra can be calculated from the Fourier-transformed velocity correlation function of the polarizability tensor α. However, this requires evaluating α for a large number of MD configurations along each trajectory, which has high computational cost if done by ab-initio methods.

We therefore use kernel-based machine learning (ML) methods with density-based descriptors [2, 3] to predict α based on atomic positions. Ab-initio calculations are then only needed for obtaining a training data set, reducing the computational cost significantly. We use a number of test systems, including both solids and small molecules, to test and optimize several different variants of this approach and compare the achieved prediction performances. We also test transferability of the trained models to trajectories at different temperatures.

[1] M. Thomas et al., Phys. Chem. Chem. Phys. **15**, 6608 (2013)
[2] A. P. Bartók et al., Phys. Rev. B **87**, 184115 (2013)
[3] A. Grisafi et al., Phys. Rev. Lett. **120**, 036002 (2018)

O 83.10 Fri 12:45 S054

Thermal Transport via Green-Kubo Method and Message-Passing Neural-Network Potentials — Marcel F. Langer[1,2], Florian Knoop[2,3], Christian Carbogno[2], Matthias Scheffler[2], and •Matthias Rupp[2,4] — [1]TU Berlin, Germany — [2]The NOMAD Laboratory, FHI-MPG & HU Berlin, Germany — [3]Theoretical Physics Division, Linköping U, Sweden — [4]Konstanz U, Germany

Accurate, precise, and efficient computational access to thermal conductivities of materials is relevant for scientific understanding and industrial applications. The Green-Kubo method with first-principles calculations enables the determination of thermal conductivities, even for strongly anharmonic materials [1]. However, the high computational cost of long dynamics simulations of large supercells required for convergence limits applicability for large-scale, high-throughput materials discovery. Machine-learning potentials can reduce this cost [2].

Message passing neural networks (MPNNs) are a promising, but for this task yet untested, class of models due to their relational inductive bias, implicit long-range nature, and ability to incorporate directional information. We adapt the heat flux definition for MPNNs, investigate the impact of equivariance, present a systematic account of their convergence behavior and performance, and compare them to a simpler baseline model.

[1]: C. Carbogno, R. Ramprasad, and M. Scheffler, Phys. Rev. Lett. 118 175901 (2017) [2]: P. Korotaev et al., Phys. Rev. B 100 144308 (2019); C. Mangold et al., J. Appl. Phys. 127, 244901 (2020); C. Verdi et al., NPJ Computer. Mat. 7 156 (2021)

O 84: Overview Talk Claus M. Schneider (joint session O/CPP)

Time: Friday 13:15–14:00 Location: S054

Invited Talk O 84.1 Fri 13:15 S054

Exploring the Mysteries of Topology in Quantum Materials — •Claus M. Schneider — Peter Grünberg Institut PGI-6, Forschungszentrum Jülich, 52425 Jülich

A characteristic feature of emergent or quantum materials is the competition of various spin-dependent interactions, such as spin-orbit coupling and exchange interaction. In addition, depending on the material system, there may be a breaking of time-reversal and/or inversion symmetries at play. As a consequence, topological materials may range from metals to insulators. In the vicinity of the Fermi level, this situation leads to peculiar electronic dispersions associated with Dirac and Weyl points, eventually also resulting in complex spin textures in momen- tum space. The interplay of competing mechanisms often results in unusual charge and spin transport phenomena in such materials. In order to understand the physical properties of quantum materials on a fundamental level, we need to explore these electronic states in detail and disentangle the role of the various interactions. For this purpose, we employ electron spectroscopic approaches, which explicitly take the electron spin as an experimental quantity into account. In this contribution we discuss an avenue starting from simple single-crystalline systems (e.g, W(011), Fe(100) and Co(100)) to more complex 2D and 3D quantum materials and detail the role of the individual interactions and symmetry-breaking mechanisms by experimental examples.