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 3*d* 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 $(\sqrt{3}x\sqrt{3})R30°$ 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 $PdCoO_2$ 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 $PdCoO_2$ after optical excitations. $PdCoO_2$ can be conceived as a natural multilayer system consisting of highly conductive Pd layers separated by insulating 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 $Fe_1/(MgO)_3(001)$ 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.