O 124: Development of Novel Methods II

Time: Friday 10:30–13:30 Location: WIL C107

O 124.1 Fri 10:30 WIL C107

Michelson Interferometry with Spiral Phase Plates — •Michael Reichenspurner[1], Nicola Kerschbaumer[1], Michael Fedoruk[2], Theobald Lohmüller[1], and Jochen Feldmann[1] — [1]Chair for Photonics and Optoelectronics, Nano-Institute Munich and Department of Physics, Ludwig-Maximilians-Universität, Königinstr. 10, 80539 Munich, Germany — [2]Vortex Photonics, Lindwurmstr. 115, 80337 Munich, Germany

Interferometry for highly accurate distance or surface profile measurements is widely applied in various fields of research and engineering. The basic principle of most interferometric approaches is that two propagating transverse beams are superimposed to generate a Newton*s rings interference pattern. Here, we demonstrate a different idea, using an optical vortex instead of a transverse beam for distance measurements in a Michelson configuration. By using a spiral phase plate, we generate a donut-shaped laser beam with helical propagation. The interference of two such helical beams, when they have opposite rotations, results in a characteristic flower shaped intensity distribution. Any phase difference introduced between these beams, either by a length difference or a change in refractive index in one of the beam paths, leads to a specific rotation of the flower pattern, which is even observable by the naked eye.

O 124.2 Fri 10:45 WIL C107

Cryogenic Single Nanoparticle Action Spectroscopy (cryo-SNAS) - A New Tool for Surface Science of Single Nanoparticles — •Tim Esser[1], Benjamin Hoffmann[2], and Knut Asmis[2] — [1]University of Oxford, Oxford, UK — [2]Universität Leipzig, Leipzig

Nanoparticles (NPs) with diameters from 10 to 100 nm have unique size and shape dependent properties due to their large surface-to-volume ratio and quantum effects. They are relevant in atmospheric and astro chemistry and applied in catalysis, energy storage, opto-electronics and bio-medicine. Their fundamental properties can be obscured in experiments that rely on averaging and interaction with surfaces or solutions. Action spectroscopy is a sensitive alternative to direct absorption spectroscopy for gas phase clusters and macroscopic surfaces, but was not previously applied to NPs due to limited sensitivity and resolution of most mass spectrometers above 1 MDa. Here we present a nanoparticle mass spectrometer which enables cryogenic single nanoparticle action spectroscopy (cryo-SNAS) for the first time. NPs from an electrospray ion source are trapped in a temperature controllable (8 to 350 K) split-ring electrode ion-trap. The mass-to-charge ratio and absolute mass of a single NP is determined non-destructively by optical means. Mass variations can then be monitored as a function of the trap temperature, pressure, laser power and wavelength.

First cryo-SNAS spectra of dye labeled SiO_2 NPs are shown followed by a discussion of future applications.

O 124.3 Fri 11:00 WIL C107

Observation of bulk electronic states with HAXPES on the heavy fermion system YbRh2Si2 — •Steinn Ymir Agustsson[1], Sergey Chernov[1], Sergey Babenkov[1], Olena Fedchenko[1], Dmitry Vasilyev[1], Katerina Medjanik[1], Christoph Schlueter[2], Andrei Gloskovskii[2], Yury Matveyev[2], Kristin Kliemt[3], Cornelius Krellner[3], Gerd Schoenhense[1], Jure Demsar[1], and Hans-Joachim Elmers[1] — [1]JGU Mainz — [2]DESY Hamburg — [3]Goethe Universität Frankfurt

The study of elastic strain induced effects on strongly correlated systems requires in situ mechanical deformations, which is best achieved with bulk single crystal samples. Observation of the electronic band structures using low energy photoemission techniques, however, requires additional surface cleaning methods, such as cleaving, which result challenging in combination with applying mechanical deformation. We demonstrate the use of hard x-ray photoemission (HAXPES) as an effective probe for observing bulk electronic band structures, overcoming the surface quality bottleneck, on the prototypical heavy fermion (HF) system YbRh2Si2. We successfully observed bulk states at the Fermi surface, probed with 5keV photons in a time-of-flight momentum microscope spectrometer at different temperatures. Our results agree with previous low energy photoemission experiments as well as with ab-initio calculations, and highlight the changes in the hybridized valence band and Yb 4f band dispersions between 25K and 300K.

O 124.4 Fri 11:15 WIL C107

In-plane and out-of-plane nanomechanical characterization of HOPG at the atomic scale — •Anna Lisa Eichhorn and Christian Dietz — TU Darmstadt

Multifrequency atomic force microscopy enables high resolution imaging of flat surfaces such as HOPG down to the atomic scale. The technique is based on the simultaneous excitation and detection of two or more cantilever eigenmodes. Depending on the type of the oscillation modes (flexural, torsional or lateral), out-of-plane elastic and dissipative sample properties or the in-plane shear behavior can be analyzed. Here, a bimodal approach was developed where the second flexural eigenmode amplitude was used for the topographical feedback. Additionally, either the first torsional or the first lateral eigenmode was excited at a constant amplitude while the frequency shift was recorded. Using the described setup atomic resolution was achieved in both imaging channels at ambient conditions, yet in the flexural topography images only every second carbon atom could be resolved, resulting in a triangular appearance. This effect is a result of the Bernal stacking of graphite monolayers, leading to two distinguishable carbon atom sites. Mapping the torsional/lateral frequency shift, however, provided a more comprehensive image, resolving the complete hexagonal arrangement of the carbon atoms. We aim to study the change in nanomechanical properties originating from single defects artificially generated within the structure by oxygen plasma treatment.

O 124.5 Fri 11:30 WIL C107

Electrospray Ion Beam Deposition as Universal Preparation Method for Electron Microscopy Imaging of Molecules on Ultrathin Supports — •Stephan Rauschenbach — Department of Chemistry, University of Oxford Mansfield Road, Oxford, UK

Recent developments in electron microscopy (EM) instrumentation, such as aberration corrected lenses and high speed, single electron detectors have enabled the observation of individual atoms and molecules at sub-angstrom resolution. This finds applications in structural biology through cryo-EM, however the imaging of many species is hindered by absence of a general sample preparation method.

Here we present soft-landing electrospray ion beam deposition (ES-IBD)as universal preparation method for single molecule imaging on ultra-thin substrates (graphene/carbon membranes) EM. We show applications ranging from native folded proteins to small, few-atom cluster species. The experiment enables structure determination based on direct imaging or averaging in the case of radiation sensitive molecules. In addition, the control of the ion beam offered in ES-IBD allows for chemical isolation of a target species and for the controlled activation in surface collision.

O 124.6 Fri 11:45 WIL C107

Developing descriptors for the prediction of adsorption energies on metal oxides — •Wenbin Xu, Mie Andersen, and Karsten Reuter — Chair for Theoretical Chemistry and Catalysis Research Center, Technical University of Munich, Garching, Germany

Linear scaling relations of adsorption energy arise at transition metal (TM) catalysts as a consequence of the adsorbate valency, together with the properties of the d electrons of the surface. Already, this linearity is only approximate, with outlier adsorption energies easily deviating from the trend by up to 1 eV. At other materials classes, deviations can be even more significant [1], calling for improved approaches that yield reliable adsorption energies at still comparably low computational cost.

To this end, we have recently demonstrated the usefulness of a compressed sensing approach for TM and TM alloy catalysts [2]. The corresponding SISSO (sure independence screening and sparsifying operator) approach [3] allows the prediction of adsorption energies from descriptors that are expressed as nonlinear functions of intrinsic properties of the clean catalyst surface (so-called primary features), e.g. coordination numbers, d-band moments, and work function. Here, we extend this approach to TM oxide catalysts and oxygen evolution catalysis. Next to establishing a systematic first-principles database for the SISSO training, a key aspect of our work is to identify primary features for this class of materials. [1] X. Hong, et al., ACS Catal., 6,

4428 (2016). [2] M. Andersen, et al., ACS Catal., 9, 2752 (2019). [3] R. Ouyang, et al., Phys. Rev. Mater., 2, 083802 (2018).

O 124.7 Fri 12:00 WIL C107

Towards an efficient sarin detector: a combined theory-experiment approach — •Hazem Aldahhak[1], Paulina Powroźnik[2], Piotr Pander[3], Fernando B. Dias[3], Wolf Gero Schmidt[1], Uwe Gerstmann[1], and Maciej Krzywiecki[2] — [1]Lehrstuhl für Theoretische Materialphysik, Universität Paderborn, 33095 Paderborn, Germany — [2]Institute of Physics, Center for Science and Education, Silesian University of Technology, Gliwice, Poland — [3]Department of Physics, Durham University, South Road, Durham, DH1 3LE, United Kingdom

Detecting hazardous agents is vital for efficiently preventing their effects. A prominent example in this respect is sarin gas, one of the most toxic nerve agents. Therefore, sensing devices are required to are able to detect sarin rapidly and in extremely concentration.

Here, density-functional theory (DFT) is used to analyze the interaction of sarin with single metal-centered phthalocyanines (MPc) as well as MPc layer structures, and to identify a benign model system. The calculations show almost coincident behaviors of sarin and dimethyl methylophosphonate (DMMP) on different MPcs. Among NiPc, CuPc, CoPc and ZnPc we find the interaction of both sarin and DMMP to be strongest with ZnPc, both in terms of interaction energy and adsorption induced work function change. Zinc phthalocyanine is thus proposed as a promising sensor for sarin detection. Using X-ray photoelectron spectroscopy (XPS), the theoretically predicted charge transfer to ZnPc is confirmed for the model system DMMP and identified as a key component in the sensing mechanism.

O 124.8 Fri 12:15 WIL C107

Self-interaction correction applied to molecules in solution — •Jakob Kraus[1], Sebastian Schwalbe[1], Kai Trepte[2], and Jens Kortus[1] — [1]TU Bergakademie Freiberg, Germany — [2]Central Michigan University, USA

The Fermi-Löwdin orbital self-interaction correction (FLO-SIC [1]) combats the self-interaction error [2] found in common exchange-correlation functionals of density functional theory (DFT), which is otherwise known as numerically efficient and reasonably accurate. While DFT calculations assume $T = 0$ K and the gas phase, many chemical reactions happen near room temperature and in an aqueous solution, frequently leading to shifts in energies. Thus, SIC and other approaches might profit from including thermochemical corrections and solvation effects. Here, electrostatic solvation was studied by combining the conductor-like screening model (COSMO [3]) with DFT, SIC-DFT, and wavefunction methods (HF, CCSD(T)). Using the PySCF [4], PyFLOSIC [5], and ERKALE [6] codes, ionization potentials and standard enthalpies of formation were evaluated in the gas phase and in an aqueous solution.

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[2] Perdew and Zunger, PRB 23, 5048 (1981)
[3] Klamt and Schüürmann, JCS-PT 2, 799 (1993)
[4] Sun et al., CMS 8, e1340 (2017)
[5] Schwalbe et al., arXiv:1905.02631 (2019)
[6] Lehtola et al., JCC 33, 1572 (2012)

O 124.9 Fri 12:30 WIL C107

Reliable electrostatic energies in MPE implicit solvation — •Jakob Filser, Konstantin Jakob, Markus Sinstein, Karsten Reuter, and Harald Oberhofer — Technical University of Munich

Implicit solvation models like the multipole expansion (MPE) model [1] are widely used in first-principles calculations to incorporate solvent effects without the necessity of sampling solvent degrees of freedom. MPE divides the free energy of solvation into the electrostatic interaction between the solute and a dielectric medium, and a remaining, 'nonelectrostatic' term, fitted to experimental reference data. The medium is defined to fill all space outside a 'cavity' around the solute.

In the present work, we solve two shortcomings of the state-of-the-art treatment of electrostatic interactions in MPE: First, for larger and more complex solutes the multipole basis for the potential becomes insufficient to solve the electrostatic problem. Currently, this is partially compensated for in the nonelectrostatic energy contribution. However, an accurate solution which does not rely on such error cancellation is obviously more desirable. We achieve this by dividing space into approximately spherical domains inside each of which a multipole basis is sufficient to express arbitrary harmonic potentials.

Second, the shape of the cavity crucially influences the electrostatic interaction, but there is no unique and straightforward definition of the cavity. This can lead to a systematic error in the electrostatic interaction. As a remedy, we choose a cavity definition which – at least on average – neither over- nor underestimates this term.

[1] M. Sinstein et al., J. Chem. Theo. Comput. 13, 5582, 2018.

O 124.10 Fri 12:45 WIL C107

Boosting size convergence for slab supercell calculations of materials exhibiting spontaneous polarization — •Su-Hyun Yoo[1], Mira Todorova[1], Chris Van de Walle[2], and Jörg Neugebauer[1] — [1]Department of Computational Materials Design, Max-Planck-Insitut für Eisenforschung GmbH, Düsseldorf, Germany — [2]Computational Materials Group, Materials Dept., UCSB, USA

The most common approach to describe surfaces in density-functional theory is the repeated slab geometry. A common strategy to avoid artificial charge transfer from one side of the slab to the other is to passivate the backside of the slab. Using the example of ZnO(0001) surfaces we find that conventionally used passivation schemes (e.g. pseudo H or surface reconstructions) break down for materials exhibiting internal polarization. We have therefore developed a generalized passivation method that accounts for the effect of spontaneous polarization and correctly describes the electric field limit for pyroelectric materials. It is robust and ensures quick convergence of total energies and electronic structure with respect to slab thickness as will be demonstrated using the example of wurtzite ZnO.

O 124.11 Fri 13:00 WIL C107

Theoretical study of Ln atoms in complexes and surfaces: valence electrons effect — •Stanislav Avdosheno and Alexey Popov — Leibniz Institute for Solid State and Materials Research (IFW), D-01069 Dresden, Germany

Detailed electronic structure of single atomic magnets is the most crucial bit in the further understanding and design of a new generation of monatomic functional elements on surfaces and in another structural setting. Recently, the inspirational single atomic experiment with Ho on MgO surfaces brought into a new light a possibility to control and manipulate the quantum states of an atom.[1] However, these convincing experiments are puzzling by the insufficient theoretical description. In this contribution, we looked at the exact ab initio model for the Ho atom at the MgO surface under the various levels of complexity in an attempt to resolve an experimental dilemma in the reported data. The research is based on the idea of the imminent need to implement the local d- and p-shell electrons of Ho atom into the active space for proper system consideration. By doing so, we have obtained the solution which complements experimental observations without any additional assumptions.[2]

References

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O 124.12 Fri 13:15 WIL C107

Spin-dependent electron reflection from Au(111) and single-layer MoS_2/Au(111): A comparative study — •Christoph Angrick[1], Andre Reimann[1], Nicole Mutzke[1], Moritz Ewert[2,3], Lars Buss[2,3], Jens Falta[3], Jan Ingo Flege[2,3], and Markus Donath[1] — [1]University of Münster, Germany — [2]Brandenburg University of Technology Cottbus-Senftenberg, Germany — [3]University of Bremen, Germany

In the field of spin-integrated photoemission experiments, major parts of the occupied electronic band structure are obtained in a short time by parallel detection of electrons with different energies and/or angles. Spin-polarizing electron mirrors (spin mirrors) are added to the experimental setups to obtain spin resolution [1,2]. The concept of these spin mirrors is based on the reflection of very-low-energy electrons from targets influenced by spin-orbit or exchange interaction.

In this work, Au(111) and single-layer MoS_2/Au(111) are put to a test as scattering targets. Both are influenced by spin-orbit interaction and therefore, are possible candidates for the use as a spin mirror. Maps of the reflectivity, Sherman function and figure of merit are derived from spin-dependent very-low-energy electron reflection measurements for a wide range of incident electron polar and azimuthal angles and energies [3]. The targets are compared with respect to preparation procedure, target stability and efficiency.

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[2] Tusche *et al.*, Appl. Phys. Lett. **99**, 032505 (2011).
[3] Thiede *et al.*, Phys. Rev. Applied **1**, 054003 (2014).