O 42: Focus Session: Frontiers of Electronic-Structure Theory – Advances in Time-Dependent and Nonequilibrium Ab Initio Methods III

Time: Wednesday 10:30–12:30 Location: HE 101

O 42.1 Wed 10:30 HE 101

Pauli blocking and core-hole screening in X-ray transient absorption spectroscopy at the Zn K-edge in ZnO — •Lu Qiao[1], Thomas C. Rossi[2], Keith Gilmore[1], Renske M. van der Veen[2], and Claudia Draxl[1] — [1]Physics Department and IRIS Adlershof, Humboldt-Universität zu Berlin, 12489 Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 14109 Berlin, Germany

X-ray transient absorption (XTA) spectroscopy can provide valuable insight into the atomic structure, the electronic properties, and the dynamics of materials. Zinc oxide (ZnO) is a prototypical wide band gap band insulator with many applications in optoelectronics and photovoltaics. Combining state-of-the-art first principle calculations and picosecond XTA spectroscopy, we reveal XTA spectra at the Zn K-edge of ZnO thin films are composed of overlapping contributions from lattice heating and core-hole screening. In particular, we analyze the computed results obtained from the Bethe-Salpeter equation and time-dependent density functional theory to disentangle the contributions from Pauli blocking and core-hole screening. Our combined experimental and theoretical approach reveals that core-hole screening enhances with increasing excitation densities; its contribution to the XTA spectra dominates over Pauli blocking.

O 42.2 Wed 10:45 HE 101

Photoemission tomography of excitons in periodic systems through Wannier interpolation — •Christian Simon Kern and Peter Puschnig — Institute of Physics, NAWI Graz, University of Graz, Austria

Excitons, bound optical excitations below the band gap of a material, are theoretically described as correlated electron-hole states. Their wave functions can be obtained by many-body perturbation theory in the framework of the Bethe-Salpeter equation or within time-dependent density functional theory. Experimentally, time- and angular-resolved photoemission spectroscopy (t-ARPES) is arguably the most direct method to measure the excitons' momentum-space signatures, which is also boosted by the recent developments in ultra-fast laser physics.

For molecules in the gas-phase, a formal connection of the theoretical exciton wave function with measured t-ARPES momentum maps in the spirit of photoemission orbital tomography was recently established [1] and has lead to interesting consequences for t-ARPES: the photoemission signature of an exciton is measured at all kinetic energies that are in accordance with the energy conservation from its hole contributions, while the respective momentum maps result from a coherent sum over all electronic contributions. Here, we extend this formalism to periodic systems and momentum-dark excitons, which allows for the simulation of photoemission from excitons in crystals, van der Waals hetero-structures or metal-organic interfaces.

[1] C. S. Kern et. al., Phys. Rev. B 108, 085132 (2023).

O 42.3 Wed 11:00 HE 101

Unveiling electronic correlation and many-body effects in the CrMnFeCoNi high-entropy alloy — •David Redka[1,2], Marco Caputo[5], Trpimir Ivsic[3], Heinz Huber[2], Hubert Ebert[4], J. Hugo Dil[3,5], László Forró[3,6], and Ján Minár[1] — [1]University of West Bohemia, Czech Republic — [2]Hochschule München, Germany — [3]Institute of Physics, EPFL, Switzerland — [4]Universität München, Germany — [5]Paul Scherrer Institut, Switzerland — [6]University of Notre Dame, USA

The impact of electronic correlation effects in the CrMnFeCoNi high-entropy alloy on electronic transport, spectroscopies, and optics is investigated. Here we combine experiments like resonant photoemission, and optical response with DFT calculations utilizing the KKR Green's function method including many-body effects via dynamical mean-field theory (DMFT). Our results show excellent agreement between theory and experiment, particularly for onsite Coulomb interactions, mirroring those in the containing pure elements. Through linear response calculations, we demonstrate the influence of the imaginary part of the self-energy of localized d-band electrons. In terms of electrical resistivity, we confirm the known dominance of chemical and magnetic disorder at low temperatures, whereas at higher temperatures, the DMFT contribution becomes significant. In general, we find that many-body correlation effects especially affect states at higher binding energies, next to the strong band smearing due to chemical disorder, revealing the impact on different material properties and response.

O 42.4 Wed 11:15 HE 101

Mott Metal-Insulator Transition from Steady-State Density Functional Theory — •Stefan Kurth[1,2,3], David Jacob[1,2], and Gianluca Stefanucci[4,5] — [1]Univ. of the Basque Country UPV/EHU, San Sebastian, Spain — [2]IKERBASQUE, Basque Foundation for Science, Bilbao, Spain — [3]Donostia International Physics Center DIPC, San Sebastian, Spain — [4]Univ. of Rome Tor Vergata, Rome, Italy — [5]INFN, Section Rome Tor Vergata, Rome, Italy

We present a computationally efficient method to obtain the spectral function of bulk systems in the framework of steady-state density functional theory (i-DFT) using an idealized scanning tunneling microscope (STM) setup. We calculate the current through the STM tip and then extract the spectral function from the finite-bias differential conductance. 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 the Mott metal-insulator transition is captured by i-DFT.

O 42.5 Wed 11:30 HE 101

Quasiparticle Self-Consistent *GW* Study of Simple Metals — •Christoph Friedrich, Stefan Blügel, and Dmitrii Nabok — Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

The *GW* method is a standard method to calculate the electronic band structure from first principles. It has been applied to a large variety of semiconductors and insulators but less often to metallic systems, in particular, with respect to a self-consistent employment of the method. In this work, we take a look at all-electron quasiparticle self-consistent *GW* (QS*GW*) calculations for simple metals (alkali and alkaline earth metals) based on the full-potential linearized augmented-plane-wave approach and compare the results to single-shot (i.e., non-selfconsistent) G_0W_0 calculations, density-functional theory (DFT) calculations in the local-density approximation, and experimental measurements. We show that, while DFT overestimates the bandwidth of most of the materials, the *GW* quasiparticle renormalization corrects the bandwidths in the right direction, but a full self-consistent calculation is needed to consistently achieve good agreement with photoemission data. The results mainly confirm the common belief that simple metals can be regarded as nearly free electron gases with weak electronic correlation. The finding is particularly important in the light of a recent debate, in which this seemingly established view has been contested. D.N. and S.B. gratefully acknowledge financial support by the MaX Center of Excellence funded by the EU through the H2020-INFRAEDI-2018 (project: GA 824143).

O 42.6 Wed 11:45 HE 101

FAIR Spectroscopy Data in NOMAD: from Theory towards Experiments — •José M. Pizarro[1], Nathan Daelman[1], Joseph F. Rudzinski[1], Luca M. Ghiringhelli[2], and Silvana Botti[3] — [1]Institut für Physik und IRIS-Adlershof, Humboldt-Universität zu Berlin, Berlin — [2]Department of Materials Science and Engineering, Friedrich-Alexander-Universität, Erlangen-Nürnberg — [3]RC-FEMS and Faculty of Physics, Ruhr University Bochum, Bochum

The emergence of big data in science underscores the need for FAIR (Findable, Accessible, Interoperable, Reusable) [1] data management. NOMAD [nomad-lab.eu][2, 3] is an open-source data infrastructure that meets this demand in materials science, enabling cross-disciplinary data sharing and annotation for both computational and experimental users.

In this talk, I will present our recent work in extending NOMAD to support a range of many-body and excited state calculations, including *GW*, BSE, and DMFT, among others. I will demonstrate how NOMAD captures these workflows in an automated but flexible fash-

ion, enabling findability and clear, visual overviews. Finally, I will present an outlook on NOMAD′s potential for large-scale interoperability and harmonization between computational and experimental data in the field of spectroscopy.
[1] Wilkinson, M. D. *et al.*, Sci. Data **3**, 160018 (2016).
[2] Scheffler, M. *et al.*, Nature **604**, 635-642 (2022).
[3] Scheidgen, M. *et al.*, JOSS **8**, 5388 (2023).

O 42.7 Wed 12:00 HE 101

Electronic structure of oxides and perovskites: a benchmark database

— •Kshitij Sinha[1,2] and Claudia Draxl[2] — [1]Indian Institute of Science Education and Research, Tirupati, India — [2]Humboldt-Universität zu Berlin, Institut für Physik and IRIS Adlershof, 12489 Berlin, Germany

Both perovskites and oxide materials have gained significant attention in recent years due to their unique properties and, thus, a wide range of possible applications. In this work, we investigate from first principles metal oxide and halide perovskites, like $\mathrm{BaSnO_3}$, $\mathrm{CsPbI_3}$, etc., and large band-gap oxide materials, like $\mathrm{Ga_2O_3}$, $\mathrm{ZrO_2}$, etc. We focus on their electronic properties, computing their electronic band structure and band gap, the density of states, effective masses, and more by applying density-functional theory. We employ a range of density functionals, i.e. the local spin density approximation (LDA), the generalized gradient approximation (PBEsol), as well as the hybrid functional PBE0 and HSE06. All calculations are carried out with the full potential all-electron package **`exciting`** [1], that implements the linearized augmented planewave plus local orbital (LAPW+lo) basis to achieve highly precise results. **`exciting`** exhibits thus proven benchmark quality, reaching μHa precision [2]. Our calculations present benchmark calculations that can serve as valuable reference data for the community.

[1] A. Gulans *et al.* J. Phys.: Condens. Matter **26** (2014) 363202
[2] A. Gulans *et al.* Phys. Rev. B, **97**, 161105

O 42.8 Wed 12:15 HE 101

Beyond the GW approximation using effective interactions

— •Abdallah El Sahili — École Polytechnique

Electron addition and removal spectra as well as the total ground state energy can be expressed in terms of the one-body Green*s function. Often, the Dyson equation with an approximate self-energy is solved to access this quantity. Today, the probably most widely used approximation is Hedin's GW self-energy [1], which has become the state-of-the-art for the band structure calculations of solids. However, GW still suffers from the existence of a variety of different flavors that lead to a sometimes significant spread of results, it has problems to describe satellite structures in the spectral function, and it is limited to a range of materials where correlations are weak to moderate. Especially total energy calculations present a number of open questions. In the present work we concentrate on the screened interaction W. On one side, we discuss the different perturbation expansions for the self-energy that can be explored according to the choice of W. On the other side, we generalize the concept of the screened interaction to a generalized effective interaction, for which we derive in principle exact equations as well as promising approximations. We show that this allows one to go beyond the GW approximation in an efficient way, avoiding the computational complexity of full vertex corrections. Illustrations for the general findings are given using the half-filled symmetric Hubbard dimer.

[1] Lars Hedin, Phys. Rev. 139, A796 (1965)