TT 81: Electronic Structure Theory: Many-Body Effects (organized by O)

Time: Wednesday 18:15–21:00 Location: Poster A

TT 81.1 Wed 18:15 Poster A

Spin Entanglement in Atoms and Molecules — •Stefano Pittalis[1], Filippo Troiani[1], Carlo Andrea Rozzi[1], and Giovanni Vignale[2] — [1]Istituto Nanoscienze, Consiglio Nazionale delle Ricerche, Modena, Italy — [2]Department of Physics, University of Missouri, Columbia, Missouri, USA

We investigate spin entanglement in many-electron systems within the framework of density functional theory. We show that the entanglement length of a Kohn-Sham system, which is extracted from the spatial dependence of the local concurrence, is a sensitive indicator of atomic shells, and reveals the character, covalent or metallic, of chemical bonds. These findings shed light on the remarkable success of modern density functionals, which tacitly employ the entanglement length as a variable. This opens the way to further research on entanglement-based functionals.

TT 81.2 Wed 18:15 Poster A

The role of the quantum well states in the oscillating behavior of the magnetic anisotropy energy and the orbital moment anisotropy — •Leonid Sandratskii — Max Planck Institute of Microstructure Physics, Halle, Germany

We report detailed first-principles theoretical study of the correlated behavior of the magnetic anisotropy energy and the orbital moment anisotropy as a function of the thickness of the ferromagnetic films. The role of the quantum well states in the formation of the oscillations is discussed. The analysis of the contributions of different points in the 2D Brillouin zone is performed. The special role of the surface layer is considered. The symmetry of the quantum mechanical problem is studied. It is shown that the electronic states possess transversal components of the orbital moment that compensate each other after summation over the Brillouin zone.

TT 81.3 Wed 18:15 Poster A

Ab initio calculation of the first order Raman spectrum of graphene and graphite — •Albin Hertrich, Caterina Cocchi, Pasquale Pavone, and Claudia Draxl — Department of Physics, Humboldt-Universität zu Berlin, Germany

Raman scattering produced by fluctuations of the dielectric function caused by phonons is an important non-destructive method for characterizing carbon-based materials. By adopting a fully ab initio approach, we compute the first order Raman spectrum of graphene and graphite using the full-potential all-electron density-functional-theory (DFT) package exciting [1]. This code is based on the augmented plane-waves approach and allows for the calculation of both phonon-dispersion curves, within the frozen-phonon approximation, and frequency-dependent dielectric tensors, from time-dependent DFT and the Bethe-Salpeter equation. Starting from these ingredients, the first-order Raman scattering intensity (G peak) within the adiabatic approximation is obtained from the derivative of the dielectric function with respect to the normal coordinates of the optical phonons at the Γ point of the Brillouin zone and the vibrational matrix element. Our results are interpreted and discussed in comparison with the existing literature.

[1] A. Gulans *et al.* J. Phys.: Condens. Matter **26** (2014) 363202

TT 81.4 Wed 18:15 Poster A

Thermoelectric clathrates: stability of the $Ba_8Al_xSi_{46-x}$ and $Sr_8Al_xSi_{46-x}$ compounds — •Maria Troppenz, Santiago Rigamonti, and Claudia Draxl — Humboldt-Universität zu Berlin

On the search for high-efficiency thermoelectric materials, promising candidates are clathrate compounds. Their cage-like structure, capable of containing guest atoms, allows for exploiting the idea of the phonon-glass electron-crystal, proposed as a way to reach a large figure of merit.

In this work, we focus on the clathrates $Sr_8Al_xSi_{46-x}$ and $Ba_8Al_xSi_{46-x}$. In contrast to the well-studied Ga-Ge based clathrates, Al-Si clathrates are of technological interest in terms of price, weight, and low environmental impact. The use of Sr and Ba as guests is motivated by the glass-like conductivity of $Sr_8Ga_{16}Ge_{30}$ and the high thermoelectric efficiency of $Ba_8Ga_{16}Ge_{30}$. The electronic transport properties are optimized at the Zintl composition, corresponding to $x = 16$. However, it has not been possible to synthesize samples with $x > 10$ for Sr or $x > 15$ for Ba [1].

We explore the structural stability of the compositional range $0 \leq x \leq 16$ as well as the stability against competing phases that are observed in experiments (e.g. $SrAl_2Si_2$). Due to the large number of substitutional configurations we resort to a cluster expansion based on accurate calculations of a subset of substitutional configurations.

[1] J.H. Roudebush, N. Tsujii, A. Hurtando, H. Hope, Y. Grin, and S.M. Kauzlarich, *Inorg. Chem.* **51**, 4161 (2012).

TT 81.5 Wed 18:15 Poster A

Atomistic Modeling of Optical Coefficients in Layered Materials — •Christian Vorwerk, Caterina Cocchi, and Claudia Draxl — Humboldt Universität zu Berlin and IRIS Adlershof, 10099 Berlin, Germany

The optical properties of a material are fully determined by the complex dielectric tensor. Comparison with measured data however, requires accounting for the experimental setup. This is in particular important for molecular materials, owing to their anisotropic nature.

We apply a 4x4-matrix formalism [1] to calculate optical coefficients of layered anisotropic materials, combining *ab initio* calculations of the dielectric tensor with the solution of Maxwell's equations. This approach [2] allows us to investigate the impact of polarization and incidence angle of the incoming light beam on the spectra of layered materials, having a specific orientation with respect to the substrate. We apply the formalism to optical as well as to X-ray absorption spectra, computing the full dielectric tensor with the all-electron full-potential code `exciting` [3] by means of many-body perturbation theory. Our prototypical target systems are different polymorphs of sexithiophene, crystals of functionalized pentacenes, self-assembled monolayers of azobenzenes, and more.

[1] P. Yeh, Surf. Sci. 96, 41 (1980). [2] P. Puschnig and C. Ambrosch-Draxl, Adv. Eng. Mater. 8, 1151 (2006). [3] A. Gulans et al., J. Phys.: Condens. Matter 26, 363202 (2014).

TT 81.6 Wed 18:15 Poster A

The Novel Materials Discovery (NoMaD) Repository — •Evgeny Blokhin[1], Fawzi Mohamed[2], Karsten Hannewald[1], Luca Ghiringhelli[2], Pasquale Pavone[1], Christian Carbogno[2], Johann-Christoph Freytag[3], Matthias Scheffler[2], and Claudia Draxl[1] — [1]Humboldt-Universität zu Berlin, Institut für Physik und IRIS Adlershof, Zum Großen Windkanal 6, 12489 Berlin — [2]Fritz Haber Institute of the Max Planck Society, Theory Department, Faradayweg 4-6, 14195 Berlin — [3]Humboldt-Universität zu Berlin, Institut für Informatik, Rudower Chaussee 25, 12489 Berlin

Since many years our community is producing an enormous amount of data by CPU-intensive calculations. Most of these data are not used, although the information content is significant. We propose to change our scientific culture following the idea of open access. The NoMaD Repository (https://nomad-repository.eu) was established to host, organize, and share materials data. NoMaD also copes with the increasing demand of storing scientific data and making them available for longer periods. NoMaD facilitates research groups to share and exchange their results. Upload of data is possible without any barrier. Results are accepted in their raw format as produced by the underlying electronic-structure package. At present, the repository is being filled with results for inorganic and organic semiconductors, catalysis and corrosion, optoelectronics, thermoelectrics, and biophysics. These areas will be continuously complemented by others.

TT 81.7 Wed 18:15 Poster A

Acoustic magnons in the long-wavelength limit: resolving the Goldstone violation in many-body perturbation theory — •Mathias C.T.D. Müller, Christoph Friedrich, and Stefan Blügel — Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

Ferromagnetic materials exhibit a spontaneously broken global rotation symmetry in spin space leading to the appearance of massless quasiparticles (zero gap) in the long-wavelength limit. These magnons are formed by the correlated motion of electron-hole pairs with opposite spins, which we describe from first principles employing the T-matrix formalism in the ladder approximation within the FLAPW method [1]. Due to approximations used in the numerical scheme,

the acoustic magnon dispersion exhibits a small but finite gap at Γ. We analyze this violation of the Goldstone mode and discuss possible correction schemes. One of the correction schemes, which involves an adjustment of the Kohn-Sham (KS) exchange splitting, is motivated by the spin-wave solution of the one-band Hubbard model. The new exchange splittings turn out to be closer to experiment. We present corrected magnon spectra for Co, Ni, and Fe. In addition, we discuss an approach that implements the magnetic susceptibility using a renormalized Green function instead of the KS one. The latter, much more expensive approach is expected to fulfill the Goldstone condition without correction.

[1] E. Şaşıoğlu *et al.*, Phys. Rev. B **81**, 054434 (2010); C. Friedrich *et al.* Top. Curr. Chem. **347**, 259 (2014).

TT 81.8 Wed 18:15 Poster A

Electronic Structure of Hybrid Materials by Means of Self-Consistent *GW* — •Nora Salas-Illanes and Claudia Draxl — Humboldt-Universität zu Berlin, Institut für Physik, Theoretische Festkörperphysik, Zum Großen Windkanal 6, 12489 Berlin

Nano-structured hybrid materials, typically consisting of two or more components that exhibit different nature, are very promising for optoelectronic applications. Unexpected new electronic properties can arise in these systems, which are absent in either of the building blocks. Unfortunately, state-of-the-art electronic-structure methods are not well suited or can even badly fail for such interfaces.

For a large number of materials, DFT provides accurate results for most ground-state properties. However, in order to obtain realistic results of electronic excitations, we have to go beyond DFT, using many-body perturbation theory (MBPT). State-of-the-art MBPT calculations are performed using the quasiparticle approach in the *GW* approximation.

In principle, the *GW* approach requires the self-consistent solution of the Hedin equations. However, most of the up-to-date calculations are performed using the results of the first iteration (one-shot *GW*). Unfortunately, this procedure cannot be used for obtaining accurate results for hybrid materials where, instead, some kind of self-consistent *GW* needs to be employed.

We implemented the quasi-particle self-consistent *GW* scheme in the all-electron full-potential code **exciting**. We present here the first results of our implementation for selected prototype materials.

TT 81.9 Wed 18:15 Poster A

Size-dependent optical gaps of Cu chalcogenide nanocrystals from *GW* and the Bethe-Salpeter equation — •Sabine Körbel[1,2], Miguel Alexandre Lopes Marques[1,2,3], and Silvana Botti[4,2,3] — [1]Martin-Luther-Universität Halle-Wittenberg, Germany — [2]Université Claude Bernard Lyon 1, France — [3]European Theoretical Spectroscopy Facility — [4]Friedrich-Schiller-Universität Jena, Germany

In semiconductor nanocrystals, quantum confinement, which opens the optical gap with decreasing nanocrystal size, allows to tune the optical absorption edge. Hence, nanoparticles of different sizes may be used in multilayer thin-film solar cells, with each layer consisting of nanocrystals of a different size, absorbing light at a different frequency, therefore enhancing the overall efficiency of the solar cell, all the while using one and the same photovoltaic absorber material. Here we present size-dependent optical gaps of nanocrystals of $Cu_2ZnSn(S,Se)_4$ [CZTS(e)], an earth-abundant semiconductor with a direct optical gap suitable for solar-cell absorbers, calculated with *GW* and the Bethe-Salpeter equation, and compared to experimental data, where available.

TT 81.10 Wed 18:15 Poster A

2D Metal Dichalocogenides and Oxides for Hydrogen Evolution: A Computational Approach — •Mohnish Pandey[1], Aleksandra Vojvodic[2], Kristian S. Thygesen[1], and Karsten W. Jacobsen[1] — [1]Center for Atomic-scale Materials Design, Department of Physics, Technical University of Denmark, DK - 2800 Kongens Lyngby, Denmark — [2]SUNCAT Center for Interface Science and Catalysis, Department of Chemical Engineering, Stanford University, Stanford, California 94305, United States

We explore the possibilities of hydrogen evolution by basal planes of 2D metal dichalcogenides and oxides in the 2H and 1T phases using the hydrogen binding energy as a computational descriptor. The binding energies are calculated using density functional theory with the BEEF-vdW functional which includes uncertainty estimates on the calculated binding energies. For some groups of systems like the Ti, Zr, and Hf dichalcogenides the hydrogen bonding the bonding to the 2H structure is stronger than to the 1T structure while for the Cr, Mo, and W dichalcogenides the behavior is opposite. This is rationalized investigating shifts in the chalcogenide *p*-levels comparing the two structures.