HL 80: Challenges in semiconductor theory

Time: Thursday 15:00–17:15   Location: EW 015

HL 80.1   Thu 15:00   EW 015

Full exact exchange and non-local correlation - a next step on the path to a physical Kohn-Sham density of states? — •Tobias Schmidt and Stephan Kümmel — University of Bayreuth, Germany

Density Functional Theory (DFT) provides an efficient and, in principle, exact framework to calculate the electronic structure of matter. In practice, DFT results strongly depend on the exchange correlation (xc) functional used. Hybrid functionals, i.e., combining a semi-local density functional with a fixed percentage of Fock exchange, have been in widespread use. However, there is no "perfect" hybrid: Hybrids with a low (ca. 20%) percentage of Fock exchange describe binding and structural properties well, but much higher amounts of Fock-exchange (ca. 50%) are needed if one aims at eigenvalues that well approximate the density of states.

In an attempt to overcome this gap, a local hybrid functional was designed. The constant mixing parameter of usual hybrids is replaced by a density functional. Thus, full Fock exchange is combined with non-local correlation. We discuss in how far this generalization leads to results that are similar or different from the usual global hybrids. We pay particular attention to the reduction of self-interaction errors, the long-range asymptotic behavior of the potential, and whether the Kohn-Sham density of states becomes more physical. First results on practically relevant, non-trivial systems with d-electrons, e.g., Palladium particles, will be presented.

HL 80.2   Thu 15:15   EW 015

Towards simple orbital-dependent density functionals for molecular dissociation — •Igor Ying Zhang[1], Patrik Rinke[1,2], and Matthias Scheffler[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, 12149 Berlin-Dahlem — [2]COMP/Department of Applied Physics, Aalto University School of Science, P.O. Box 11100, FI-00076 Aalto, Finland

Density functional theory (DFT) is one of the leading first-principles electronic-structure theories. However, molecular dissociation remains a challenge, because it requires a well-balanced description of the drastically different electronic structure at different bond lengths. One typical and well-documented case is the dissociation of both H_2^+ and H_2, for which all popular DFT functionals fail [1]. We start from the Bethe-Goldstone equation to propose a simple orbital-dependent correlation functional which generalizes the linear adiabatic connection approach. The resulting scheme is based on second-order perturbation theory (PT2), but includes the self-consistent coupling of electron-hole pairs, which ensures the correct H_2 dissociation limit and gives a finite correlation energy for systems with a (near)-degenerate energy gap. This coupling PT2-like (CPT2) approximation delivers a significant improvement over all existing functionals for both H_2 and H_2^+ dissociation. We will demonstrate the reason for this improvement analytically for H_2 in a minimal basis.[1] A. J. Cohen *et al.*, Chem. Rev. **112** 289 (2012). [2] F. Caruso *et al.*, Phys. Rev. Lett. **110** 146403 (2013).

HL 80.3   Thu 15:30   EW 015

Hybrid density-functional calculations of polarons in doped MgO. — •Sebastian Kokott, Sergey Levchenko, and Matthias Scheffler — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Charge-carrier conductivity in oxides can influence their surface chemical properties [1]. However, the nature and properties of the charge carriers are not well understood. In this work, we investigate self-trapped polarons in MgO within an *ab initio* framework. Density-functional theory with the hybrid functional HSE06 is used for the atomic relaxtion and electronic structure calculations. The choice of the functional is validated by the analysis of the (de)localization bias according to the generalized Koopmans' condition [2] as a function of the fraction of Hartree-Fock exchange. We find a localized polaronic distortion of the MgO lattice which was not reported previously: two neighboring oxygen atoms forming an O_2^{2-} moiety with two trapped electronic holes. We do not find a self-trapped state for a single hole with the HSE06 functional. The formation energy of the polaron is about 1 eV, *i.e.*, thermally accessible at typical temperatures when *p*-doped MgO is used as a catalyst (800-1,000 K) [3]. We also explore

polarons trapped at intrinsic and extrinsic point defects, in particular Mg vacancies and hydrogen intersitials. □   [1] Richter et al., Phys. Rev. Lett. **111**, 045502 (2013); [2] Lany and Zunger, Phys. Rev. B **80**, 085202, (2009); [3] Arndt et al., Catalysis Rev. **54** (4), 424-514 (2011).

HL 80.4   Thu 15:45   EW 015

Electronic structure and adsorption properties of Ni substitutional defects at MgO(001). — •Aliaksei Mazheika, Sergey V. Levchenko, and Matthias Scheffler — Fritz-Haber-Institut der MPG, Berlin, DE

Ni-MgO solid solutions were demonstrated to be stable and active catalysts for CO_2 activation and conversion. However, despite numerous experimental investigations, the nature of the active sites, in particular at realistic conditions, remains unknown. As a first step towards understanding the nature of the sites, we calculate the electronic and atomic structure of Ni substitutional defects at the (100) surface of MgO, using the density-functional theory with the Heyd-Scuseria-Ernzerhof family of hybrid functionals HSE(α,ω) [1]. We determine the optimal fraction of the exact exchange α by comparing the positions of the defect levels and the valence-band maximum with respect to the vacuum level obtained with DFT and G_0W_0 where self-energy is evaluated with partial self-consistency of the eigenvalues. The obtained optimal value of $\alpha = 0.6$ also gives an agreement within 0.1 eV between the DFT and MP2 energies of CO adsorption at the Ni defect. The MP2 adsorption energy calculated for a small embedded cluster model was previously shown to be very close to the CCSD(T) adsorption energy [2]. An optimal compromise (within 0.05 eV) can be achieved between the accuracy of the defect level alignment and the CO adsorption energy for $\alpha = 0.5$.—[1] A.V. Krukau, O.A. Vydrov, A.F. Izmaylov, G. Scuseria. J. Chem. Phys., **125**, 224106 (2006); [2] I. Mehdaoui, T. Klüner. J. Phys. Chem. A, **111**, 13233 (2007).

HL 80.5   Thu 16:00   EW 015

Atomistic-continuum modeling of short laser pulse melting of Si targets — •Vladimir P. Lipp[1,2], Baerbel Rethfeld[2], Martin E. Garcia[1], and Dmitry S. Ivanov[1,2] — [1]University of Kassel, Kassel, Germany — [2]Technical University of Kaiserslautern, Kaiserslautern, Germany

We present an atomistic-continuum model to simulate ultrashort laser-induced melting processes in semiconductor solids on the example of silicon. The kinetics of transient non-equilibrium phase transition mechanisms is addressed with a Molecular Dynamics method at atomic level, whereas the laser light absorption, strong generated electron-phonon non-equilibrium, fast diffusion of and heat conduction due to photo-excited free carriers are accounted for in the continuum. We give a description of the model, which is then applied to study the mechanism of short laser pulse melting of free standing Si films. The effect of laser-induced pressure and temperature of the lattice on the melting kinetics is investigated. Two competing melting mechanisms, heterogeneous and homogeneous, were identified. Apart of classical heterogeneous melting mechanism, the nucleation of the liquid phase homogeneously inside the material significantly contributes to the melting process. The threshold fluence value, at which homogeneous nucleation of liquid starts contributing to the classical heterogeneous propagation of the solid-liquid interface, is found from the series of simulations at different laser input fluences. On the example of Si, the laser melting kinetics of semiconductors was found to be noticeably different from that of metals with fcc crystal structure.

HL 80.6   Thu 16:15   EW 015

A real-time DFT scheme for electronic transport — •Philipp Schaffhauser and Stephan Kümmel — Universität Bayreuth, Deutschland

We present an approach for calculating the charge transport through molecular systems that relies on solving the time-dependent Kohn-Sham equations in real-time and real-space. So far, propagation methods were mostly used for closed quantum systems and therefore did not allow for studying transport problems which involve a source and a drain. We resolve this problem by introducing absorbing and anti-absorbing boundary conditions. By comparing to established results we verify our computational scheme. Using the new formalism we ex-

amine how molecular distortions, and, e.g., breaks in the conjugation of organic systems, change the molecular conductivity.

HL 80.7   Thu 16:30   EW 015

Piezoelectricity in planar boron nitride via a geometric phase — ●Matthias Droth[1] and Vitor Pereira[2] — [1]University of Konstanz, Germany — [2]Graphene Research Centre, National University of Singapore

Due to their low surface mass density, two-dimensional materials with a strong piezoelectric response are interesting for nanoelectromechanical systems with high susceptibility. In contrast to graphene, the two sublattices in two-dimensional hexagonal boron nitride (hBN) are occupied by different types of atoms, which allows for piezoelectricity. Recently, the piezoelectric tensor of extended hBN has been calculated via density functional theory (DFT). While an analytical description of piezoelectricity does exist for hBN nanotubes, this is, to our knowledge, not the case for two-dimensional hBN. We set up a Hamiltonian that involves the strain-induced pseudomagnetic field and derive the piezoelectric tensor using the modern theory of polarization. Our findings are in exact agreement with symmetry arguments and give an analytical explanation for the piezoelectric electron-phonon coupling in planar hBN. We also provide an estimation of the coupling strength and find a piezoelectric response similar to reported DFT results.

HL 80.8   Thu 16:45   EW 015

Anisotropic Electrostatic Friction of Organic Molecules on ZnO Surfaces — ●Karol Palczynski and Joachim Dzubiella — Institut für Physik, Humboldt-Universität zu Berlin, Newtonstr. 15, 12489 Berlin, Germany

We study the long-time self-diffusion of a single conjugated organic *para*-sexiphenyl (*p*-6P) molecule physisorbed on the inorganic ZnO $(10\bar{1}0)$ surface by means of all-atom molecular dynamics computer simulations. We find strongly anisotropic diffusion processes in which the diffusive motion along the polar [0001] direction of the surface can be many orders of magnitudes slower at relevant experimental temperatures than in the perpendicular direction. The observation can be rationalized by the underlying charge pattern of the electrostatically heterogeneous surface which imposes direction-dependent energy barriers to the motion of the molecule. Furthermore, the diffusive behavior is found to be normal and Arrhenius-like, governed by thermally activated energy barrier crossings. The detailed analysis of the underlying potential energy landscape shows, however, that in general the activation barriers cannot be estimated from idealized zero-temperature trajectories but must include the conformational and positional excursion of the molecule along its pathway. Furthermore, the corresponding (Helmholtz) free energy barriers are significantly smaller than the pure energetic barriers with implications on absolute rate prediction at experimentally relevant temperatures.

HL 80.9   Thu 17:00   EW 015

high resolution nuclear inelastic scattering of antimony trioxide — atefeh jafari[1,2,3], ronnie e. simon[1,2], abdelfattah Mahmoud[1], dimitrios bessas[3], volkr l. Deringer[4], ralf stoffel[4], ●richard dronskowski[4], ilya sergeev[5], benedikt klobes[1], and raphael p. hermann[1,2] — [1]Jülich Centre for Neutron Science JCNS and Peter Grünberg Institute PGI, Forschungszentrum Jülich, Jülich, Germany — [2]Faculty of Sciences, University of Liège, Liège, Belgium — [3]European Synchrotron Radiation Facility, Grenoble, France — [4]Institute of Inorganic Chemistry, RWTH Aachen University Landoltweg 1, Aachen — [5]Deutsches Elektronen-Synchrotron, Hamburg, Germany

The lattices dynamics of the α and β phases of antimony trioxides (Sb2O3) have been carried out using Mössbauer spectroscopy, nuclear inelastic scattering (NIS) and density functional theory (DFT). The experimental results are in very good agreement with our DFT calculations.The acoustic cut-off frequency of α- Sb2O3 is approximately 78 cm-1 as determined from the density of phonon states, DPS. Complementary to other Raman or IR methods, NIS is a powerful technique which yields the element specific density of phonon states of Mössbauer isotopes. Moreover, due to the high resolution achieved with the backscattering sapphire monochromator, we could identify weak and partially superposed bands.