HL 56: Electronic-Structure Theory: New Concepts and Developments in Density Functional Theory and Beyond - IV

Time: Wednesday 10:30–13:00   Location: GER 38

Invited Talk   HL 56.1   Wed 10:30   GER 38
Electronic excitations in 2D materials and heterostructures —
●Kristian Sommer Thygesen — Technical University of Denmark,
Lyngby, Denmark

Atomically thin two-dimensional (2D) materials have recently emerged
as a new class of materials with unique and highly tunable opto-
electronic properties. Different 2D crystals can be stacked to form
van der Waals heterostructures (vdWH) where the individual 2D lay-
ers are held together by weak van der Waals forces leading to atom-
ically well-defined interfaces. This fascinating scenario opens up the
possibility of designing heterostructures with tailored electronic or op-
tical properties. I will give a general introduction to the electronic
properties of 2D materials, including characteristic features of their
dielectric screening and collective excitations with special emphasis on
the challenges related to their ab-initio description. I will show how
the dielectric function of a given 2D material can be controlled by em-
bedding it into a vdWH, and how this in turn can be used to control
the band structure, exciton binding energies or the plasmon dispersion
in 2D materials.

HL 56.2   Wed 11:00   GER 38
**Charge and energy transport at the nanoscale: A DFT per-
spective** — ●Florian G. Eich, Fabio Covito, and Angel Rubio
— Max Planck Institute for the Structure and Dynamics of Matter,
Luruper Chaussee 149, D-22761 Hamburg, Germany

Understanding the interplay between charge and energy transport at
the nanoscale paves the way for novel thermoelectric devices, which
may prove useful for the development for sustainable energy sources.
However, concepts, such as heat flow, temperature and entropy are
only well-established at the macroscopic level for slow dynamics. This
raises the question about whether these concepts can be employed for
small length and short time scales. We will present our recent efforts
to use a time-dependent density-functional theory framework, dubbed
nonequilibrium thermal DFT, in order to generalize temperature and
heat or energy flow to the microscopic regime. To this end we will
highlight the analogy of the formally exact microscopic equations of
motion for charge density and energy density in thermal DFT to the
macroscopic equations of motion of hydrodynamics. Furthermore, we
will present first result using our approach to compute transient energy
energy currents induced by a temperature gradient and show that in
the steady-state limit persistent temperature oscillations develop.

HL 56.3   Wed 11:15   GER 38
Conductance of aromatic and antiaromatic molecular circuits
— Narendra P. Arasu and ●Héctor Vázquez — Inst. of Physics,
Academy of Sciences of the Czech Rep., CZ

Molecular structures with delocalized conjugated orbitals play an es-
sential role in molecular transport due to their high conductance and
small attenuation factors. While much work has been done on aro-
matic molecules, some studies have shown that conductance actually
decreases with aromaticity [1].

In this talk I will discuss the effect of (anti)aromaticity on conduc-
tance. I will show results of first-principles transport calculations for
an aromatic-antiaromatic pair of molecules and compare with exper-
iment. Conductance is calculated using DFT and NEGF including
corrections to the DFT level positions. The corrected conductance
values are in very good agreement with experiment. We find that the
conductance of the antiaromatic molecule is much higher than that of
its aromatic counterpart. Calculations show this to be a consequence
of the smaller HOMO-LUMO gap of the antiaromatic complex as well
as on the molecular level alignment at the junction [2].

[1] W. Chen, H. Li, J.R. Widawsky, C. Appayee, L. Venkataraman,
and R. Breslow, J. Am. Chem. Soc. 136 918 (2014).

[2] S. Marqués-González, S. Fujii, J.-Y. Shin, H. Shinokubo, N.P.
Arasu, H. Vázquez and M. Kiguchi, (to be submitted).

HL 56.4   Wed 11:30   GER 38
**Current-induced cooling of Carbene-based molecular junc-
tions: role of electrodes structure** — ●Giuseppe Foti and Héc-
tor Vázquez — Institute of Physics, Czech Academy of Sciences
Cukrovarnicka 10, Prague 6

In this talk I will present our first principles calculations based on
density functional theory (DFT) plus Nonequilibrium Green's func-
tions (NEGF) of the current-induced heating and cooling dynamics
of a series of Carbene-based molecular junctions [1]. I will show how
the atomistic details of electrode terminations have a strong impact
on the heating dynamics of the junctions and how they can maxi-
mize the cooling of the system. In the cases where the molecule is
attached to blunt leads and the electronic coupling to bulk states is
strong the cooling efficiency of the most active vibrational modes de-
creases monotonically as bias increases. This results in the heating of
the junction. On the other hand, when the molecule is connected to
sharp electrode terminations such as chain-like structures, which can
be formed experimentally when the metal-molecule bond is mechani-
cally strong, and the electronic coupling to electrode states is weak, the
cooling efficiency shows a non-monotonic behavior. It first decreases
as a function of voltage but then increases at relatively high biases,
effectively cooling down the junction [2]. These results reveal the im-
portant role of the atomistic structure of metal-molecule interface in
the current-induced damping of localized molecular vibrations.

[1] Foti, G.; Vázquez, H. Nanotechnology 2016, 27, 125702.
[2] Foti, G.; Vázquez, H. submitted

HL 56.5   Wed 11:45   GER 38
**DFTB-based recursive Green's function algorithms for elec-
tron transport in quasi-1D systems** — ●Fabian Teichert[1,2,4],
Andreas Zienert[3,4], Jörg Schuster[4], and Michael Schreiber[2]
— [1]Dresden Center for Computational Materials Science (DCMS),
Dresden, Germany — [2]Institute of Physics, Technische Universität
Chemnitz, Chemnitz, Germany — [3]Center for Microtechnologies
(ZfM), Technische Universität Chemnitz, Chemnitz, Germany —
[4]Fraunhofer Institute for Electronic Nano Systems (ENAS), Chem-
nitz, Germany

Within the last decades, quantum transport theory and density func-
tional theory have become very important for predicting the electronic
properties of new materials and future electronic devices.

We focus on the problem of improving quantum transport algorithms
for large quasi-1D systems which are enormously time-consuming to-
day. We combine the density functional tight binding (DFTB) ap-
proach with the recursive Green's function formalism (RGF), which
is very effective for such systems. First, we show how to improve the
RGF for the case of randomly distributed real defects. For this, we use
the steps of the renormalization decimation algorithm (RDA), which
is part of the electrode calculation. Second, we show how to improve
the calculation of the surface Green's functions of electrodes which
have a long unit cell. Here, we employ the decimation technique to
reduce the dimensionality of the periodic Hamiltonian matrix, leading
to effective matrices, which are treated by the RDA. Finally, we apply
these algorithms to carbon nanotubes and present our results.

HL 56.6   Wed 12:00   GER 38
**Conditions for formation of two-dimensional electron gas at
the LaFeO$_3$/SrTiO$_3$** — ●Igor Maznichenko[1], Sergey Ostanin[1],
Arthur Ernst[2], Ingrid Mertig[1,2], Katayoon Mohseni[2], Hol-
ger L. Meyerheim[2], Eberhard K.U. Gross[2], Pengfa Xu[3], Wei
Han[3], Philip M. Rice[3], Jaewoo Jeong[3], Mahesh G. Samant[3],
and Stuart S.P. Parkin[2,3] — [1]Institut für Physik, Martin-Luther-
Universität Halle-Wittenberg, D-06099 Halle, Germany — [2]Max-
Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle,
Germany — [3]IBM Almaden Research Center, 650 Harry Road, San
Jose, California 95120, USA

The formation of a conducting two dimensional electron gas (2DEG)
at the interface between two insulating oxide layers was explained the-
oretically for atomically and chemically abrupt interfaces via polar
discontinuity.

Here we show that a 2DEG is formed at the interface between thin
layers of lanthanum ferrite, LaFeO$_3$ (LFO), that are more than 3 unit
cells thick, when grown epitaxially on SrTiO$_3$ (STO) (001). The in-
terface property highly depends on the surface property of TiO$_2$ ter-
minated STO. The interface is conducting if the STO is not annealed
in an oxygen environment prior to the LFO growth, while insulating
if the STO is annealed.

First principles calculations reveal that a 2DEG should be realized for an ideal interface but that modest chemical intermixing suppresses it. These calculations also show that the presence of oxygen vacancies supports 2DEG formation due to electronic doping.

HL 56.7 Wed 12:15 GER 38

Thermal Renormalization of the Electronic Structure: Trends across Chemical and Structural Space — ●Honghui Shang[1], Christian Carbogno[1], Patrick Rinke[2], and Matthias Scheffler[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin — [2]Aalto University, Helsinki, Finland

Advances in electronic structure theory now allow us to compute the renormalization of the electronic structure due to thermal nuclear motion from first principles [1]. In this work, we present a systematic computational assessment of this renormalization for 82 octet binaries in both the zincblende and the rocksalt structure. After validating our computational approach that is based on finite-differences [2] and Fröhlich-type corrections [3] for polar materials, we discuss and analyze the observed trends: For instance, we find that most materials exhibit the expected band-gap reduction upon temperature increase; however, some materials (e.g. CuCl and CdO) do not follow this trend and exhibit the opposite behavior. We discuss the underlying electronic mechanism as well as its dependence on the chemical composition and structure of the material. In this context, also the sensitivity of such calculations with respect to the chosen basis set and exchange-correlation functional (LDA, PBE, HSE06) are critically investigated.
[1] F. Giustino, arXiv:1603.06965 (2016).
[2] G. Antonius, *et al. Phys. Rev. Lett.* **112**, 215501 (2014).
[3] J. P. Nery and P. B. Allen, *Phys. Rev. B* **94**, 115135 (2016).

HL 56.8 Wed 12:30 GER 38

Spin-wave excitations and electron-magnon scattering from 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

We study the spin excitations and the electron-magnon scattering in bulk Fe, Co, and Ni within the framework of many-body perturbation theory as implemented in the full-potential linearized augmented-plane-wave method. Starting from the GW approximation we obtain a Bethe-Salpeter equation for the magnetic susceptibility treating single-particle Stoner excitations and magnons on the same footing. 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 theorem and present an approach that implements the magnetic susceptibility using a renormalized Green function instead of the non-interacting one, leading to a substantial improvement of the Goldstone-mode condition [1]. Finally, we employ the solution of the Bethe-Salpeter equation to construct a self-energy that describes the scattering of electrons and magnons. The resulting renormalized band structures exhibit strong spin-dependent lifetime effects close to the Fermi energy. We also see kinks in the electronic bands, which we attribute to electron scattering with spatially extended spin waves.
[1] Müller *et al.*, Phys. Rev. B **94**, 064433 (2016).

HL 56.9 Wed 12:45 GER 38

Charged supercells revised: Small Polarons in Oxides with proper account for long-range polarization — ●Sebastian Kokott, Sergey V. Levchenko, and Matthias Scheffler — Fritz-Haber-Institut der MPG, Berlin, DE

Formation of small polarons (excess charges localized within one unit cell) often determines charge mobility and optical absorption in oxide materials. In this work, we address two important challenges in the DFT description of small polarons: sensitivity to the errors in exchange-correlation (XC) treatment and finite-size effects in supercell calculations. The polaron properties are obtained using a modified neutral potential-energy surface (PES) [1]. Using the hybrid HSE functional and considering the whole range $0 \leq \alpha \leq 1$ of exact exchange, we show that the modified PES model significantly reduces the dependence of the polaron level and binding energy in MgO and TiO_2 on the XC treatment. It does not eliminate the dependence on supercell size. Based on Pekar's model [2], we derive the proper long-range behavior of the polaron and a correction that allows to obtain the polaron properties in the dilute limit (tested for supercells containing up to 1,000 atoms). The developed approach reduces drastically the computational time for exploring the polaron PES, and gives a consistent description of polarons for the whole range of α. It allows us to find a self-trapped hole in MgO that is noticeably more stable than reported previously.—[1] B. Sadigh *et al.*, PRB **92**, 075202 (2015); [2] S.I. Pekar, ZETF **16**, 335 (1946). This work received funding from the Leibniz ScienceCampus "GraFOx".