MM 12: Frontiers of Electronic Structure Theory: Focus on Topology and Transport

Time: Monday 15:45–17:45   Location: H51

MM 12.1 Mon 15:45 H51

Mechanism of Li intercalation/deintercalation into/from the surface of LiCoO$_2$ — •Ashkan Moradabadi and Payam Kaghazchi — Institut für Chemie und Biochemie, Freie Universität Berlin, Takustr. 3, 14195 Berlin, Germany

LiCoO$_2$ is the most commonly used cathode material in Li-ion batteries. In this work, we have investigated atomic and electronic structures, magnetic properties, formation energies, and energy barriers for the diffusion of Li in single vacancies, divacancies, and missing rows in bulk and surface of LiCoO$_2$. Our GGA-PBE results indicate that there is almost no energy barrier for the Li-ion deintercalation from the surface layer. Energy barrier for the Li-ion intercalation is also very small. However, we find that Li hopping in PBE+U is accompanied by electron hopping between nearby transition metal ions. Therefore a PBE+U barrier, which is for both Li hopping and charge hopping, is higher than the corresponding PBE barrier [1]. This study has implications in understanding the role of the surface in the rate capability of nanostructured LiCoO$_2$ cathodes of Li-ion batteries.

[1] Ashkan Moradabadi and Payam Kaghazchi, Mechanism of Li intercalation/deintercalation into/from the surface of LiCoO$_2$, Phys. Chem. Chem. Phys., 2015, 17, 22917-22922.

MM 12.2 Mon 16:00 H51

Potential-dependent mechanism of Li diffusion in Li$_2$S — •Ashkan Moradabadi[1,2] and Payam Kaghazchi[1] — [1]Institut für Chemie und Biochemie, Freie Universität Berlin, Takustr. 3, 14195 Berlin, Germany — [2]Institut für Materialwissenschaft, Fachgebiet Materialmodellierung, Technische Universitat Darmstadt, Jovanka-Bontschits-Str. 2, 64287 Darmstadt, Germany

Li-S batteries are promising candidates for large-scale applications such as electrical vehicles. However, the measured discharge capacity is often less than the theoretical one [1,2]. This is mainly due to the slow diffusion of Li through Li$_2$S shells formed on S$_8$ cores, which leads to an incomplete conversion of S$_8$ cores to Li$_2$S (the final product of lithiation of S$_8$). In the present work, using density functional calculation, we have investigated mechanism of Li diffusion in Li$_2$S. At low cell voltages (< 0.93 V), Li diffusion occurs via an exchange mechanism with a high energy barrier of 0.45 eV. However at higher cell voltages, Li diffusion takes place via a vacancy mechanism with a lower energy barrier of 0.27 eV. Our findings can explain the capacity fading in Li-S batteries at high operation rates.

[1] Liang, X.; Hart, C.; Pang, Q.; Garsuch, A.; Weiss, T.; Nazar, L. F.; A highly efficient polysulfide mediator for lithium-sulfur batteries. Nature Communications, 2015, 6, 5682.

[2] Wang, L.; Wang, Y.; Xia, Y.; A high performance lithium-ion sulfur battery based on a Li$_2$S cathode using a dual-phase electrolyte. Energy Environ. Sci. 2015, 8, 1551.

MM 12.3 Mon 16:15 H51

Extremely high magnetoresistance in topological insulator candidate LaBi — •Nitesh Kumar, Chandra Shekhar, and Claudia Felser — Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany.

Lanthanum monopnictides (LaX, X=N, P, As, Sb, Bi) have recently been predicted to be topological insulators, except LaN which is a topological semimetal. Inspired from this report we have studied the transport properties of LaBi. It has a simple rock salt-type structure with alternate La and Bi atoms arranged in all three directions. Temperature variation of resistivity at different magnetic fields follows Kohler's rule. Resistivity follows almost a parabolic relation with magnetic field without saturation, exhibiting a huge magnetoresistance (1.5×10^5% at 2 K and 9 T). By employing two band model we calculate carrier density and mobility of electrons and holes which suggests that LaBi is a compensated system. We believe this to be responsible for high unsaturated MR in LaBi. We observe excellent Shubnikov-de Haas (SdH) oscillations starting from around 3T. We also analyse the angle and temperature dependence of these oscillations.

MM 12.4 Mon 16:30 H51

VOTCA-STP - Multi Scale Modeling of Spin Transport in Organic Semiconductors — •Erik R. McNellis, Shayan Hemmatiyan, Amaury Melo Souza, Sebastian Müller, and Jairo

Sinova — Johannes Gutenberg University, Mainz, Germany

Organic molecules present a range of unique and highly attractive properties in solid state technology applications. So also in spintronics, where the weak but highly tailorable spin-orbit coupling in light elements offers spin lifetimes of unparalleled length and controllability.

1st-principles theoretical modeling stands to provide a crucial perspective on the emerging field of spin transport in organic semiconductors. Comprehensive modeling of relevant systems is challenging, with several of the spin transport mechanisms in traditional solid state materials non-existent or strongly modified in organics.

We are developing a multi-scale modeling framework for spin transport in bulk organic materials, based on the VOTCA toolkit for charge transport in the same. The core component is a semi-classical kinetic Monte-Carlo model, with input parameters calculated using 1st-principles theory.

The scope, capabilities of and particular challenges for this development will be presented along with possible extensions to e.g. 'spinterfaces', where spin currents are manipulated by tailoring of an inorganic / organic solid interface, as well as a perspective on the potential ramifications for experimental work in the field.

15 min. coffee break

MM 12.5 Mon 17:00 H51

High-pressure and nonlinear elastic response of solids: Example of carbon allotropes — •Pasquale Pavone, Rostam Golesorkhtabar, Stefan Kontur, and Claudia Draxl — Humboldt-Universität zu Berlin, Physics Department and IRIS Adlershof, 12489 Berlin, Germany

As prototype materials showing strong nonlinear elastic behaviour, diamond and, more recently, layered carbon allotropes have attracted much attention. However, even the nonlinear elasticity of diamond is not completely clarified: Experimentally, nonlinear elastic constants of diamond were investigated only recently [1], showing significant discrepancies with theoretical results. Furthermore, the standard ab-initio reference calculation for diamond [2] is nowadays about 30 years old and needs to be updated in the light of current development of theory, numerical algorithms, and available computer power. Using the full-potential all-electron package **exciting** [3], we perform a systematic ab-initio investigation of the nonlinear elastic properties of diamond, graphene monolayers, as well as simple-hexagonal and hexagonal graphite. We develope an extension of the **ElaStic** tool [4] for the determination of third-order elastic constants. From these results the pressure dependence of linear elastic constants is obtained and connected to dynamical quantities like the mode Grüneisen parameters.

[1] J.M. Lang et al., Phys. Rev. Lett. **106**, 125502 (2011).
[2] O.H. Nielsen, Phys. Rev. B **34**, 5808 (1986).
[3] A. Gulans et al., J. Phys.: Condens. Matter **26**, 363202 (2014).
[4] R. Golesorkhtabar et al., Comp. Phys. Commun. **184**, 1861 (2013).

MM 12.6 Mon 17:15 H51

Calculations of temperature dependent resistivity for transition metals from the first principles — •David Wagenknecht[1,2], Ilja Turek[1,2], and Karel Carva[1] — [1]Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University in Prague; Ke Karlovu 3, 12116 Prague 2, Czech Republic — [2]Institute of Physics of Materials, Academy of Sciences of the Czech Republic; Žižkova 22, 61662 Brno, Czech Republic

The temperature dependence of electrical resistivity is studied from the first principles. Properties of late transition metals have been calculated using the linear muffin-tin orbital (LMTO) method with the coherent potential approximation (CPA). The influence of non-zero temperature has been described by a frozen lattice disorder – atoms were moved from the positions on an ideal lattice and different temperatures are then given by the magnitudes of the random displacement vectors. Dependence of the physical quantities on the parameters of the displacements (like random and non-random directions of the displacement vectors) has been observed and taken into account during discussion of the results. Special attention has been paid to an influence of spin-orbit interaction on the final resistivity, as well as to comparison with other *ab initio* calculations and experimental data;

the obtained results agree reasonably well with those of other authors. Derived analytical modifications of the LMTO potential functions and the numerical codes can be now used to calculate relevant physical properties of different materials.

MM 12.7   Mon 17:30   H51

Ab Initio Molecular Dynamics Study of Conjugated Polymer Systems: The Elusive Localization of the Polaron — •Håkan W. Hugosson[1], Amina Mirsakiyeva[1], and Anna Delin[1,2] — [1]Department of Materials och Nano Physics, KTH Royal Institute of Technology, Stockholm, Sweden. — [2]Ångstrom Laboratory, Uppsala University, Uppsala, Sweden.

The thermoelectric conjugated polymer poly(3,4-ethylenedioxythiophene), or PEDOT, contains a carbon backbone consisting of alternating short and long carbon bonds. Therefore there are two isomeric states: aromatic and quinoid. Charge injection or the presence of charged doping agents leads to the formation of localized charge in the conjugated polymer - a so-called polaron. This polaron induces a localized structural distortion (a shift from the aromatic form towards the quinoid) in the conjugated carbon backbone.

Self-localized polarons in conjugated carbon systems have been found using semi-empirical or HF-theory, but formerly never using DFT with local or gradient corrected functionals (e.g. LDA/BLYP). Self-localization has been seen using DFT and long range hybrid functionals with partial exact exchange included.

Using modern ab initio molecular dynamics methods based on DFT we have studied PEDOT and its charge carrying polarons. A localized polaron is now found when studying the time-averaged changes in bond-distances and also in snap-shots for the frontier orbitals for long oligomers (12 monomers).