MM 62: Methods in Computational Materials Modelling IV: Method development

Time: Thursday 15:45–17:45   Location: H53

MM 62.1   Thu 15:45   H53

Functional renormalization and mean-field approach to multi-band systems with spin-orbit coupling: Application to the Rashba model with attractive interaction — Giulio Schober[1], •Kay-Uwe Giering[1], Michael Scherer[1], Carsten Honerkamp[2], and Manfred Salmhofer[1] — [1]Heidelberg University — [2]RWTH Aachen University

The functional renormalization group (RG) in combination with Fermi surface patching is a well-established method for studying Fermi liquid instabilities of correlated electron systems. In this article, we further develop this method and combine it with mean-field theory to approach multi-band systems with spin-orbit coupling, and we apply this to a tight-binding Rashba model with an attractive local interaction. The spin dependence of the interaction vertex is fully implemented in a RG flow without SU(2) symmetry, and its momentum dependence is approximated in a refined projection scheme. In particular, we discuss the necessity of including in the RG flow contributions from both bands of the model, even if they are not intersected by the Fermi level. As the leading instability of the Rashba model, we find a superconducting phase with a singlet-type interaction between electrons with opposite momenta. While the gap function has a singlet spin structure, the order parameter indicates an unconventional superconducting phase, with the ratio between singlet and triplet amplitudes being plus or minus one on the Fermi lines of the upper or lower band, respectively. We expect our general approach to be useful for an unbiased theoretical description of the low-temperature properties of spin-based materials.

MM 62.2   Thu 16:00   H53

Advanced Brillouin zone sampling for first-principles calculations — •Philip Hasnip and Matthew Probert — University of York, York, UK

In quantum mechanical materials models such as density functional theory (DFT), the material's wavefunction is expressed as a Bloch function multiplied by a phase factor exp{ik.r}, where k is drawn from the material's first Brillouin zone. Solutions to the quantum equations exist for all k and many quantities, including the total energy, require an integration over all k; these integrals are performed numerically on a finite mesh of sampling 'k-points'.

The sampling density required for accurate integration cannot be known a priori, and varies from material to material; metallic states require particularly high k-point densities in order to reproduce the Fermi surface accurately, yet metallicity is an emergent property of the simulation and cannot be known reliably beforehand.

In this work we discuss the common methods for performing this k-point integration, highlighting the problems encountered in real simulations, and present alternative methods which ameliorate these difficulties. In particular we present a fast estimate for the k-point sampling error which may be used dynamically to guide simulations. The method will be illustrated with results using the Castep DFT program.

MM 62.3   Thu 16:15   H53

The electronic structure of transition metal phthalocyanines: the influence of electron correlation — •Iulia Emilia Brumboiu, Soumyajyoti Haldar, Johann Lüder, Olle Eriksson, Heike C. Herper, Barbara Brena, and Biplab Sanyal — Department of Physics and Astronomy, Uppsala University, SE-75120 Uppsala, Sweden

Transition metal phthalocyanines (TMPcs) have been in the focus of scientific research as possible candidates for applications in spintronics and molecular electronics. These types of applications make use of the molecular spin and usually involve the adsorption of the TMPc on various substrates. In the framework of density functional theory, the description of the molecule adsorbed on a surface is not straightforward, especially due to the presence of the metal d electrons. A compromise between accuracy and computational cost is achieved by combining DFT with a Hubbard term exclusively acting on the correlated electrons. Many studies on TMPcs adsorbed on surfaces make use of this approach, but the choice of U is mainly based on previously reported values for similar systems. We provide a detailed analysis of the influence of U on the electronic structure of five TMPcs (Mn-, Fe-, Co-, Ni- and CuPc). By comparing the calculated valence band to photoelectron spectra and by computing the Hubbard term from

linear response, we show that U is different for every phthalocyanine. For each molecule, we provide a range of U values that show good agreement with the experiments. These values represent a safe starting point for DFT+U calculations of physisorbed TMPcs.

MM 62.4   Thu 16:30   H53

Hybrid-functional calculations of iron defects in ferropericlase — •Sebastián Alarcón Villaseca, Sergey V. Levchenko, and Matthias Scheffler — Fritz-Haber-Institut der MPG, 14195 Berlin, DE

The accurate description of transition-metal-containing defects in crystals presents a challenge for DFT with standard LDA and GGA functionals. Although replacing a fraction α of the (semi-)local exchange by the exact exchange (EX) in hybrid DFT reduces the self-interaction error, α remains a parameter depending on the target property. Here, the dependence of the formation energies of Fe_{Mg} substitutional defects in ferropericlase (Mg,Fe)O - the second most abundant mineral in Earth's lower mantle - on α in the Heyd-Scuseria-Ernzerhof hybrid functional (HSE) ($0 \leq \alpha \leq 1$, $\alpha = 0$ corresponds to PBE) is explored, and the results are compared to CCSD(T) embedded cluster calculations. We find that HSE($\alpha = 0.48$) reproduces the formation energy obtained with CCSD(T) and satisfies the constraint that the DFT HOMO equals the ionization energy as calculated by G_0W_0@HSE(α). Periodic models with converged unit-cell size are then used to calculate the atomic and electronic structure of Fe_{Mg} substitutional defect in MgO. The $O_h \rightarrow D_{4h}$ symmetry lowering (compressed FeO_6 octahedron) derived from experiments [1] is found for all α values (with HSE($\alpha = 0.25$) lattice parameter). However, the negative Δt_{2g} splitting of the Fe $3d$ states is obtained only for $\alpha \geq 0.4$. The calculated Δt_{2g} at these α values indicate a dynamic Jahn-Teller effect [1].
[1] T. Hauptricht et al., Phys. Rev. B 82, 035120 (2010).

15 min. coffee break

MM 62.5   Thu 17:00   H53

First-principles simulations of solid state NMR spectral parameters for metals with the GIPAW method — •Ary Ferreira, Karsten Reuter, and Christoph Scheurer — Technische Universität München

Solid-state nuclear magnetic resonance (NMR) spectroscopy is a powerful tool for the characterization of a number of industry-relevant metallic materials like bulk metallic glasses or promising metallic anode materials in secondary lithium-ion batteries. For insulating solid materials, NMR chemical (or orbital) shifts can be successfully predicted from first-principles by all-electron density-functional theory calculations using localized basis sets in a cluster approach. Alternatively, the Gauge-Including Projector-Augmented Wave (GIPAW) method allows the use of periodic boundary conditions, plane waves and pseudopotentials. In both cases, the effect of the applied magnetic field on the electronic structure is simulated considering only the charge nature of electrons. However, for metals, the measured NMR shifts are dominated by their spin nature, resulting in the so-called Knight shifts. We address this limitation by implementing an extension of the GIPAW method to metals proposed in [1] within the framework of the open-source Quantum Espresso package. We report a set of preliminary computations of NMR spin and orbital shifts for bulk metallic Ag, Cu, and Cs, for which experimental reference values are available in the literature. The encouraging results pave the way towards the application of the GIPAW method to real-world metallic systems. [1] Mayeul d'Avezac et al., Phys. Rev. B **76**, 165122 (2007).

MM 62.6   Thu 17:15   H53

Efficient "On-the-Fly" Calculation of Raman Spectra from Ab-Initio Molecular Dynamics: Application to Lithium-Sulfur Batteries — •Pouya Partovi-Azar[1], Thomas D. Kühne[2], and Payam Kaghaychi[1] — [1]Physical and Theoretical Chemistry, Freie Universität Berlin, Takustr. 3, D-14195 Berlin, Germany — [2]Department of Chemistry, University of Paderborn, Warburger Str. 100, D-33098 Paderborn, Germany

We present a novel computational method to accurately calculate Raman spectra from first principles. Together with an extension of the second-generation Car-Parrinello method to propagate maximally lo-

calized Wannier functions along with the nuclei, a speed-up of one order of magnitude can be obtained.

We have used this method to investigate the Raman spectra of $(Li_2S_4)_n$, $n = 1, 4, 8$ clusters which are believed to be the last intermediates in the $S_8 \rightarrow Li_2S$ transition in lithium-sulfur (Li-S) batteries during the discharge cycle. However, it is not fully established whether or not any other products coexist with the Li_2S crystal in the discharged state. In this study, we have observed a clear evidence of $Li_2S_4 \rightarrow Li_2S_2$ transition by investigating the systematic changes in the simulated Raman spectra as the cluster size increases. In line with recent experiments, we found that the Raman-active sulfur-sulfur stretching mode at 440 cm^{-1} can be considered as a signature of covalent bonding between two and more sulfur atoms per formula. We have also demonstrated that the transition is mainly due to the strong electrostatic interactions between polar Li_2S_4 monomers.

MM 62.7   Thu 17:30   H53

Anisotropic magnetoresistance from the surface states of disordered topological insulators — •Henry Legg and Achim Rosch — Institut für Theoretische Physik, Universität zu Köln, D-50937 Cologne, Germany

In the presence of time-reversal symmetry electrons on the surface of a topological insulator cannot backscatter from disorder. A magnetic field parallel to the surface lifts this protection mechanism and enables backscattering which is predominantly in the direction parallel to the magnetic field. As a result the anisotropy of magentoresistance parallel and perpendicular to **B** is a sensitive probe of the loss of topological protection when time-reversal symmetry is broken.

Using a self-consistent T-matrix approximation we demonstrate how an in-plane magnetic field dramatically alters both the density of states and the resistivity of a topological insulator's surface. Our results are compared to experiments where the strong dependence on gate voltages provides an especially clear experimental signature of the scattering mechanism.