

O 21: Electronic Structure and Spin-Orbit Interaction I

Time: Monday 16:00–18:45

Location: H42

O 21.1 Mon 16:00 H42

A non-local density functional by explicitly modeling of the exchange-correlation hole in inhomogeneous systems —

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We develop a non-local density functional by a direct modeling of the shape of exchange-correlation (xc) hole in inhomogeneous systems. The functional is aimed at giving an accurate xc-energy and an accurate corresponding xc-potential even in difficult near-degeneracy situations such as molecular bond breaking. In particular we demand that: (1) the xc hole properly contains -1 electron, (2) the xc-potential has the asymptotic $-1/r$ behavior outside finite systems and (3) the xc-potential has the correct step structure related to the derivative discontinuities of the xc-energy functional. These demands are achieved by screening the exchange hole in such a way that the pair-correlation function is symmetric and satisfies the sum-rule. These two features immediately imply (1) and (2) while the explicit dependence of the exchange hole on the Kohn–Sham orbitals implies (3). For the dissociating hydrogen molecule, we obtain much improved xc holes and thus also improved binding energies at different interatomic distances, as compared to the local density approximation.

O 21.2 Mon 16:15 H42

Description of Rabi oscillations within time-dependent density functional theory —

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Many physical processes stemming from the interaction of light with matter are non-linear in nature. A prototype of such a process is the Rabi oscillation, i.e. the oscillation between the ground state and an excited state when a monochromatic laser with a frequency close to the resonance is applied. It is one of the few analytically solvable cases of non-linear electron dynamics where the population of states changes dramatically in time. Due to the resulting change in the density the description of Rabi oscillations by density functional theory (DFT) suffers from shortcomings that can be traced back to the use of adiabatic approximations which leads to a time-dependent detuning of the resonant frequency. Here, we investigate Rabi oscillations in a one-dimensional model system containing two electrons in a triplet configuration using exact exchange (EXX) and the adiabatic local density approximation (ALDA) within time-dependent DFT. Contrary to the singlet case, where no resonant Rabi oscillations can be observed, we find that the EXX and ALDA calculations for the triplet state show a near-resonant behavior. A small detuning of the laser frequency even results in a resonant Rabi oscillation in the EXX case which hints at the detuning due to the approximate functional being constant in time. We discuss the differences between the singlet and the triplet excitations which lead to a time-dependent or constant detuning, respectively.

O 21.3 Mon 16:30 H42

Double excitations from modified Hartree-Fock equations —

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While doubly excited states have become important in technological applications, e.g. in increasing the efficiency of solar cells, their description using ab initio methods is a great theoretical challenge because double excitations cannot be described in linear response based on a single Slater determinant. We recently developed a modified Hartree-Fock (HF) approximation for calculating singly excited states [1]. In the present work we extend this method further to also allow for the description of doubly excited states. We describe the double excitation as two holes in the subspace spanned from the occupied HF orbitals and two particles in the subspace of virtual HF orbitals. A subsequent minimization of the energy optimizes the representation of both the

holes and the particles in the occupied and virtual subspaces, respectively. We test our method for various atoms and polyene molecules which are known to have excitations with significant double excitation character.

[1] M.Tassi., I.Theophilou, and S.Thanos, *Int. J. Quantum Chem.* 2012 DOI:10.1002/qua.24049

O 21.4 Mon 16:45 H42

Accurate calculation of optical excitations for large systems —

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Many-body perturbation theory is the natural framework for the description of electronic excited state properties. For instance, within MBPT, single-particle excitation energies (as probed by photoemission and STS experiments) are found at the poles of the single particle Green's function. Response functions, describing optical spectra, are calculated via the solution of the Bethe-Salpeter equation (BSE), which explicitly contains the electron-hole interaction. When excitonic effects are neglected, absorption spectra are often in bad agreement with the experiments. The agreement with experiment is restored when the electron-hole interactions are included. Excited states are in this case described as linear combinations of independent-particle transition. Although very accurate, the appealing of the BSE scheme is limited due to its high computational cost. We will show how we can enlarge the scope of BSE calculations. First, we use maximally localized Wannier's functions for reducing strongly the terms which need to be calculated for evaluating the electron-hole interaction. Then, we avoid any explicit sum over empty one-particle orbitals. As a consequence we are able to determine, not only the excitation spectra, but also, directly, each excited state. We validate our scheme by addressing the optical excitations of small isolated molecules. Finally, we will illustrate a few examples where larger systems are investigated.

O 21.5 Mon 17:00 H42

Frequency-dependent electron-hole interaction in the calculation of absorption spectra —

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State of the art calculations of optical spectra rely on the Bethe-Salpeter Equation (BSE) which is usually solved in a static approximation. The inclusion of dynamical effects is however crucial to describe aspects important e.g. in the design of some high efficiency solar devices^[1]. Even if different attempts to perform dynamical-BSE exist^[2,3,4], there is still the need for a more efficient and flexible method that would allow one to cover all aspects for realistic systems.

We devised a two step scheme to include dynamical effects in optical spectra. To this aim, we first evaluate dynamical effects in the independent e-h correlation function as a pre-processing method that relies on the solution of an integro-differential equation^[5]. Then, using this result as input for an appropriate static BSE-like equation, we evaluate the full spectra including dynamical effects. Our results explains when those effects are strong, and when they are instead weakened by interference effects.

[1] For a review see A.J.Nozik, *Chem.Phys.Lett* **457**, 3 (2008)

[2] F.Bechsted et al, *Phys.Rev.Lett.* **78**, 1528 (1997)

[3] D.Sangalli et al, *J.Chem.Phys* **134**, 034115 (2011)

[4] A.Marini and R.Del Sole, *Phys.Rev.Lett.* **91**, 176402-1 (2003)

[5] See also: G.Lani, P.Romaniello, L.Reining *New J.Phys.* **14**, 013056 (2012) and M. Guzzo et al. *Phys.Rev.Lett.* **107**, 166401 (2011)

O 21.6 Mon 17:15 H42

Spin polarization of (001) surface states of the topological crystalline insulator $\text{Pb}_{1-x}\text{Sn}_x\text{Se}$ —

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It has recently been discovered, that the narrow-gap IV-VI semiconductor $\text{Pb}_{1-x}\text{Sn}_x\text{Se}$ undergoes a transition from a normal insulator

into a topological-crystalline-insulating (TCI) state when the band structure is inverted at large enough Sn content and low temperatures [P. Dziawa *et al.*, Nat. Mater. **11**, 1023–1027 (2012)]. In the TCI phase, the mirror symmetry of the crystal warrants the topological protection of metallic surface states in this material [T. H. Hsieh *et al.*, Nat. Commun. **3**, 982 (2012)].

Here, we study the (001) surface states of $\text{Pb}_{1-x}\text{Sn}_x\text{Se}$. Our calculations not only predict metallic surface states with a chiral spin structure for the TCI state but also gapped surface states with nonzero polarization when the system is a normal insulator. For both phases, angle- and spin-resolved photoemission experiments provide conclusive evidence for the formation of these surface states and their chiral spin structure.

O 21.7 Mon 17:30 H42

Spin texture and circular dichroism in photoelectron spectroscopy from the topological insulator Bi_2Te_3 : First-principles photoemission calculations — ●HOSSEIN MIRHOSSEINI¹ and JÜRGEN HENK² — ¹Max-Planck-Institut für Mikrostrukturphysik, Halle, Germany — ²Martin-Luther-Universität Halle-Wittenberg, Halle, Germany

By relativistic first-principles photoemission calculations for the topological insulator Bi_2Te_3 , we study how the spin texture of the Dirac state manifests itself in circular dichroism [1]. On one hand, there are significant modifications of the initial state's spin texture, which are explained by final-state effects and the symmetry of the photoemission set-up. On the other hand, a highly symmetric set-up allows to conclude on the detailed Dirac state's spin texture. Our study supports that circular dichroism in angular distribution successfully complements spin- and angle-resolved photoelectron spectroscopy from topological insulators.

[1] H. Mirhosseini and J. Henk, Phys. Rev. Lett. **109** (2012) 036803.

O 21.8 Mon 17:45 H42

Electronic structure and photoemission of topological insulators — ●JÜRGEN BRAUN, JAN MINAR, and HUBERT EBERT — Dept. Chemie und Biochemie, LMU Universität München, Germany

We discuss calculated spectral features of Bi_2Te_3 , Bi_2Se_3 and Sb_2Te_3 and compare with corresponding experimental data. In detail, the dependence of the CDAD on the photon energy is identified as a final state effect. Furthermore, a complete calculation of the spin-polarization vector including the component perpendicular to the surface is presented. Also the behavior of Rashba split surface resonances which had been observed at higher binding energies is analysed. Last but not least the unoccupied bulk and surface states of these materials are investigated by use of our one-step photoemission theory. The calculations have been performed in the framework of the fully relativistic version of the one-step model that is part of the Munich SPR-KKR program package [1]. To guarantee for a quantitative description of the surface-sensitive spectral features special attention is paid to the image-potential behavior of the surface barrier, which is included as an additional layer in the photoemission formalism.

1. H. Ebert *et al.*, The Munich SPR-KKR package, version 6.3, <http://olymp.cup.uni-muenchen.de/ak/ebert/SPRKKR> (2012).

O 21.9 Mon 18:00 H42

Spin-dependent unoccupied electronic structure of the topological insulator Bi_2Se_3 : An inverse-photoemission study — ●ANNA ZUMBÜLTE¹, CHRISTIAN LANGENKÄMPER¹, ANKE B. SCHMIDT¹, MARKUS DONATH¹, TOBIAS FÖRSTER², PETER KRÜGER², MARCO BIANCHI³, RICHARD HATCH³, PHILIP HOFMANN³, JIANLI MI⁴, and BO BRUMMERSTEDT IVERSEN⁴ — ¹Physikalisches Institut, Universität Münster — ²Institut für Festkörpertheorie, Universität Münster — ³Department of Physics and Astronomy, Aarhus University — ⁴Department of Chemistry, Aarhus University

In recent years, topological insulators have become a focus of photoemission studies. Especially the spin-dependent electronic structure shows still surprising features, e.g. the evolution of Rashba-split quantum well states due to a strong surface band bending. With regard to possible devices and the interpretation of non-equilibrium behavior of topological insulators, the additional knowledge of their unoccupied electronic structure is essential.

In this talk, we present spin-resolved inverse-photoemission measurements on a prototypical topological insulator: Bi_2Se_3 . While for higher energies bulk-derived features seem to dominate the obtained data, several features with distinct spin asymmetries are observable at energies up to 2 eV above the Fermi level. The nature of these will be discussed along with theoretical calculations including a long-ranged potential to simulate a band bending. In particular, these calculations predict an unoccupied W-shaped spin-split surface state which we observe experimentally at 1 eV above the Fermi level.

O 21.10 Mon 18:15 H42

Influence of adsorbates on the electronic structure of the topological insulator Bi_2Se_3 : ab-initio investigations — ●T. FÖRSTER¹, P. KRÜGER¹, M. ROHLFING¹, A. ZUMBÜLTE², C. LANGENKÄMPER², A. B. SCHMIDT², and M. DONATH² — ¹Institut für Festkörpertheorie, Universität Münster, Germany — ²Physikalisches Institut, Universität Münster, Germany

Topological insulators possess helical surface states that are protected by time reversal symmetry. Additionally, traditional surface states exist. The rich physics of these systems is determined by the actual boundary conditions at the surface which can distinctly be affected by adatoms. Using density-functional calculations, we investigate the influence of adsorbates on the electronic structure of the Bi_2Se_3 surface. First, the effect of an adsorbate induced two-dimensional electron gas at the surface is studied by adding a respective long-ranged potential $V(z)$ to the self-consistent one. This simple approach already leads to a down-shift of the Dirac point and the appearance of additional Rashba-like surface states similar to the findings of various experiments. In addition to this, we observe a W-shaped spin-split surface state in the unoccupied part of the band structure. This previously undescribed state is also seen in spin-resolved inverse-photoemission experiments. Secondly, our calculations show that monolayers of hydrogen or potassium adatoms give rise to long-ranged surface potentials and thus also induce the above mentioned features in the band structure. Furthermore, we find surface states that are specific to H and K, respectively.

O 21.11 Mon 18:30 H42

Dirac-cone-like surface state in $\text{W}(110)$: Dispersion, spin texture, and photoemission from first principles — HOSSEIN MIRHOSSEINI¹, ●MARKUS FLIEGER², and JÜRGEN HENK² — ¹Max-Planck-Institut für Mikrostrukturphysik, Halle, Germany — ²Martin-Luther-Universität Halle-Wittenberg, Halle, Germany

We report on a theoretical study of a d_{z^2} surface state at the tungsten (110) surface, addressing in detail the spin-resolved electronic structure as well as photoemission spectroscopy. In agreement with recent experiments [1], this surface state shows a strongly anisotropic dispersion: In the $\bar{H}-\bar{\Gamma}-\bar{H}$ direction of the surface Brillouin zone, it disperses linearly but becomes flattened along the $\bar{N}-\bar{\Gamma}-\bar{N}$ direction. The *ab initio* calculated spin texture agrees with the one derived from a model Hamiltonian; due to two-fold surface symmetry and time-reversal symmetry, the out-of-plane spin polarization vanishes. The photoemission intensities depend sensitively on the polarization of the incident light, because of the orbital composition of the surface state. The photoelectrons become spin-polarized out-of-plane, which is attributed to breaking the time-reversal symmetry by the excitation process.

[1] K. Miyamoto *et alii*, Phys. Rev. Lett. **108** (2012) 066808; *idem*, Phys. Rev. B **86** (2012) 161411(R).