

DS 6: Frontiers of Electronic-Structure Theory: Focus on the Interface Challenge II (joint session O/TT/DS/ CPP)

Time: Monday 15:00–17:30

Location: H9

DS 6.1 Mon 15:00 H9

Influence of structural deformations on the applicability of the Tamm-Dancoff approximation for organic molecules — •TOBIAS LETTMANN and MICHAEL ROHLFING — Institut für Festkörpertheorie, Westfälische Wilhelms-Universität Münster, 48149 Münster, Germany

When calculating excited state properties of electronic systems within the many-body perturbation theory (MBPT), the Bethe-Salpeter equation (BSE) needs to be solved. This is often done within the Tamm-Dancoff approximation (TDA), neglecting the coupling of resonant and anti-resonant excitations.

It is generally accepted that the TDA is justified for large, extended systems e.g. bulk crystals. However it has been shown that the TDA may no longer hold for small organic molecules [1]. In this talk we discuss the applicability of the TDA for molecules of different sizes and show the transition between the two regimes. We then discuss how the applicability is influenced by deformations of the molecules, in particular by the related conjugation length of the π -system.

[1] B. Baumeier et al., J. Chem. Theory Comput. **8**, 997 (2012)

DS 6.2 Mon 15:15 H9

Momentum-Resolved Electron Energy-Loss Spectroscopy in Oxides from Many-Body Perturbation Theory — •CHRISTIAN VORWERK^{1,2}, CATERINA COCCHI^{1,2}, and CLAUDIA DRAXL^{1,2} — ¹Institut für Physik, Humboldt-Universität zu Berlin, 12489 Berlin, Germany — ²European Theoretical Spectroscopy Facility

Electron energy-loss spectroscopy (EELS) is a powerful tool to investigate the local electronic and structural properties of crystalline materials. The accurate determination of these spectra from first principles requires a reliable description of the electron-hole interaction, screened by the surrounding many-electron system. We perform *ab initio* many-body perturbation theory calculations of EELS through the solution of the Bethe-Salpeter equation (BSE), including the screened non-local interaction between electron and hole. Employing an implementation in the all-electron full-potential package `exciting`, we show results for a wide range of energy loss, from the optical to the hard x-ray region. We study EELS at finite momentum loss \mathbf{q} , from small \mathbf{q} close to the dipole limit to large \mathbf{q} well beyond the first Brillouin zone. This momentum resolution of EELS reveals dipole-forbidden excitations that are invisible in absorption spectroscopy. Our calculations also yield insight into the excitonic dispersion, *i.e.* the excitonic bandstructure. We discuss the effects of momentum loss in the EELS spectra of oxide materials, including CaO, CeO₂, and the wide-gap transparent oxide Ga₂O₃, considering both the optical and x-ray energy-loss range.

DS 6.3 Mon 15:30 H9

Electron-magnon scattering in elementary ferromagnets from first principles: implementation and results — •CHRISTOPH FRIEDRICH, MATHIAS C.T.D. MÜLLER, and STEFAN BLÜGEL — Peter Grünberg Institut and Institute of Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

Propagating electrons and holes can scatter with spin fluctuations and form quasiparticles as a result or more complex many-body states. To calculate this effect, a \mathbf{k} -dependent self-energy describing the scattering of electrons and magnons is constructed from the solution of a Bethe-Salpeter equation for the T matrix. Partial self-consistency is achieved by the alignment of the chemical potentials. We discuss details of the implementation and illustrative results. The renormalized electronic band structures exhibit strong spin-dependent lifetime effects close to the Fermi energy, which are strongest in Fe. The renormalization gives rise to a band anomaly at large binding energies in iron, which results from a coupling of the quasihole with Stoner excitations.

DS 6.4 Mon 15:45 H9

Dielectric function of homogeneous electron gas from Bethe-Salpeter equation — •JAAKKO KOSKELO^{1,2}, MARTIN PANHOLZER^{2,3}, LUCIA REINING^{1,2}, and MATTEO GATTI^{1,2,4} — ¹Laboratoire des Solides Irradiés, École Polytechnique, CNRS, CEA, Université Paris-Saclay, F-91128 Palaiseau, France — ²ETSF — ³Institute for Theoretical Physics, Johannes Kepler University, Linz, Austria — ⁴Synchrotron SOLEIL, France

The homogeneous electron gas (HEG) is one of the most important model systems in condensed matter physics, and it has been subject of a great number of studies. Some properties of HEG such as total energy and static correlation functions can be obtained from quantum Monte Carlo simulations with great accuracy, but for dynamical correlation functions only very few results are available.

Methods based on the Bethe-Salpeter equation (BSE) have been very successful in semiconductors and insulators, but metals have been less studied. In this contribution, we use the BSE in its standard approximations, including a statically screened electron-hole interaction, to study the dielectric function of HEG. We find significant differences in static screening and spectra compared to other approaches. In particular, the BSE in its current approximations fails to reproduce the negative static screening in the low-density HEG, which is related to a so-called ghost exciton. We also use the time-dependent mean-density approximation [1] in order to compare our results to experimental loss spectra of sodium.

[1] M. Panholzer et al, Phys. Rev. Lett. **120**, 166402 (2018).

DS 6.5 Mon 16:00 H9

DFT study of electronic and optical properties of SrTiO_{3- δ} including many-body effects — •VIJAYA BEGUM, MARKUS E GRUNER, and ROSSITZA PENTCHEVA — Faculty of Physics and Centre for Nanointegration (CENIDE), University of Duisburg-Essen, Duisburg.

The electronic and optical properties of SrTiO₃ (STO), a perovskite material of key importance in the field of oxide electronics, are explored in the framework of density functional theory including many-body effects within the GW approximation and excitonic corrections by solving the Bethe-Salpeter equation (BSE). We further analyse the origin of the strong excitonic effects, in particular a peak at ≈ 6.5 eV, by decomposing the BSE eigenvectors obtained from GW+BSE to extract the leading electron-hole pair contribution for the particular BSE eigenstate following the approach of Bokdam *et al.* [Scientific Reports **6**, 28618 (2016)]. Alternatively, we use the model-BSE (mBSE) which utilises a parametrised analytical model for the static screening. For STO, the mBSE spectrum closely reproduces the one from GW+BSE, which allows to reduce the computational effort by circumventing the intermediate time-consuming GW step. We further proceed to describe the effect of oxygen defects on the electronic and optical properties in STO.

Funding by DFG- SFB1242, project C02 is gratefully acknowledged.

DS 6.6 Mon 16:15 H9

Second-order Møller-Plesset perturbation theory and beyond for the band gap and single-particle excitations of solids — •MARIA DRAGOMI¹, SERGEY V. LEVCHENKO^{2,1,3}, IGOR YING ZHANG^{4,1}, and MATTHIAS SCHEFFLER¹ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, DE — ²Skolkovo Innovation Center, Moscow, RU — ³NUST MISIS, Moscow, RU — ⁴Fudan University, Shanghai, CN

Calculations of the fundamental band-gap and the low-energy excitations of solids are still a challenge for electronic-structure theory. The computationally efficient Kohn-Sham (KS) density functional theory (DFT) with the widely used local or semi-local approximations provides a KS band gap which is much smaller than the fundamental gap. Many-body perturbation theory, on the other hand, addresses the fundamental gap directly. We present here an efficient scalable implementation of Møller-Plesset second order perturbation theory (MP2) for quasi-particle energies [1,2]. By solving the Dyson equation of the single-particle Green's function, considering self-energy up to second order, we go beyond MP2. The new approach shows a competitive or even superior performance in comparison to the current state-of-the-art methods such as hybrid functionals and GW approximation, where second order exchange is missing. We present numerical results for the band-gap of a wide range of semiconductors and insulators.

[1] J. Sun and R. J. Bartlett, J. Chem. Phys. **104**, 8553 (1996).

[2] A. Grüneis, et.al, J. Chem. Phys. **133**, 074107 (2010).

DS 6.7 Mon 16:30 H9

Accelerating GW Calculations within the LAPW Frame-

work — ●SVEN LUBECK, ANDRIS GULANS, and CLAUDIA DRAXL — Humboldt-Universität zu Berlin, Germany

The *GW* approach of many-body perturbation theory is an indispensable method for calculating the electronic band structure of solids. Its implementation in computer programs using the linearized augmented plane-wave + local orbital (LAPW+LO) method allows for obtaining numerically precise results. Unfortunately, high precision comes at the price of a large number of LAPWs and LOs. In this work, we accelerate *GW* calculations by optimizing the use of LAPWs and LOs in the computer package `exciting` [1]. On the one hand, we introduce a systematic way of obtaining a minimal set of LOs. On the other hand, we perform a basis transformation from the plane-wave part of the LAPWs to different types of basis functions, exploring the efficiency of numeric atom-centered orbitals, Gaussian type orbitals, and Kohn-Sham orbitals. Presenting band gaps of two exemplary materials, zincblende ZnO and hexagonal monolayer BN, we illustrate that our optimization schemes reduce the computational cost down to values as low as 15% without compromising the precision.

[1] A. Gulans, S. Kontur, C. Meisenbichler, D. Nabok, P. Pavone, S. Rigamonti, S. Sagmeister, U. Werner, and C. Draxl, *J. Phys.: Condens. Matter* **26**, 363202 (2014).

DS 6.8 Mon 16:45 H9

Ab-initio description of transient ion formation of NO on Au(111) — DANIEL CORKEN, NICHOLAS D. M. HINE, and ●REINHARD J. MAURER — Departments of Physics and Chemistry, University of Warwick, United Kingdom

Gaining a fundamental understanding of the interactions of molecules on metal surfaces is essential for the development of novel heterogeneous catalysts. An interesting feature of gas-surface reactions at metal surfaces is that the Born-Oppenheimer approximation breaks down. Vibrationally excited and translationally hot molecules can transfer energy to the electrons of a metal via excitation of electron-hole pairs (EHP). In case of NO on Au(111), [1] this nonadiabatic energy loss is believed to stem from the transient generation of charged ion species at the surface. A computationally feasible and accurate description of such a molecule-metal charge-transfer state represents a challenge and several methods have been proposed. Upon a review of existing experimental evidence, we will present our approach to this problem. We use linear expansion-Delta-Self-Consistent-Field Density Functional Theory (le Δ SCF-DFT) [2] to model the anionic resonance of NO on Au(111). The le Δ SCF-DFT method enforces the electronic configuration of reference molecular states while solving the Kohn-Sham equations self-consistently. By comparison to experiment and

other models, we assess the methods' ability to describe the ground- and excited-states during molecular scattering. We further explore avenues to extract nonadiabatic couplings and to construct model Hamiltonians based on this method. [1] *JCP* 130, 174716, [2] *JCP* 139, 014708;

DS 6.9 Mon 17:00 H9

Luminescence of β -SiAlON:Eu²⁺ phosphors: DFT study — ●SALEEM AYAZ KHAN¹, ONDREJ SÍPŘ¹, ROBIN NIKLAUS², WOLFGANG SCHNICK², and JAN MINAR¹ — ¹University of West Bohemia, Pilsen, Czech Republic — ²LMU Munich, Germany

Highly efficient phosphor-converted light-emitting diodes (pc-LEDs) are popular in lighting and high-tech electronics applications [1]. Among them β -SiAlON:Eu²⁺ stands out as a promising narrow-band green phosphor for white-LEDs applications exhibiting good thermal and chemical stabilities. Photoluminescent properties of this material can be tuned by introducing the disorder at various sublattices. To understand the mechanism behind this effect, we performed a systematic study of electronic structure and photoluminescence properties of β -SiAlON:Eu²⁺. The calculations were done within the *ab-initio* fully relativistic full-potential framework. The disorder was treated by employing both the supercell approach and the coherent potential approximation (CPA). The Stokes shifts were calculated from differences of total energies of the ground and excited states of β -SiAlON:Eu²⁺. The main focus is on monitoring how the Al and O content and Eu²⁺ activator concentrations influence the local β -Si₃N₄ electronic structure and how this may be used to tune photoluminescence properties.

[1] Z. Wang, W. Ye, Iek-H. Chu, and S. P. Ong, *Chem. Mater.*, **28**, 8622 (2016).

DS 6.10 Mon 17:15 H9

Spin fluctuations in itinerant ferromagnets: Computing the dynamic transverse spin susceptibility with TDDFT and PAW — ●THORBJØRN SKOVHUS and THOMAS OLSEN — Technical University of Denmark

We present a numerical scheme for computing the dynamic transverse spin susceptibility using time-dependent density functional theory which allows us to study magnons in itinerant ferromagnets. The scheme is based on a real-space grid implementation of the projected augmented wave method and use a simple plane wave representation of the response function. Employing the adiabatic local density approximation for the exchange-correlation kernel, calculations of the magnon spectra in bulk transition metals iron and nickel are presented. In the context of the present implementation, the influence from the choice of numerical scheme on the violation of the Goldstone theorem is investigated.