

CPP 47: Focused Session: Frontiers of Electronic Structure Theory - Non-equilibrium Phenomena at the Nano-scale VI (original: O, joined by CPP)

Time: Thursday 10:30–13:15

Location: TRE Ma

Topical Talk

CPP 47.1 Thu 10:30 TRE Ma
Localization at the edge of 2D topological insulator by Kondo impurities — ●BORIS ALTSHULER¹, IGOR ALEINER¹, and VLADIMIR YUDSON² — ¹Physics Department, Columbia University, New York, NY 10027, USA — ²Institute for Spectroscopy, Russian Academy of Sciences, Troitsk, Moscow 142190, Russia

Recent interest to the topological insulators [1] is inspired by the fact that their boundaries host gapless electronic excitations, which are extended and make the system conductive even in the presence of a potential disorder. 1D edge of a 2D topological insulator is predicted to have perfect conductance ($2e^2/h$): right and left moving electrons carry opposite spins and potential disorder cannot flip spins and thus causes neither back-scattering nor the usual 1D localization.

What if there are localized spins coupled to the edge electrons? It turns out that the conductivity is still perfect provided that this coupling conserves the z-projection of the total spin of the impurities and electrons. Magnetic anisotropy violates this conservation and causes the backscattering even at $T=0$, i.e. an arbitrary small density of the spins with arbitrary weak anisotropy of the coupling leads to Anderson localization of the edge states in long enough samples [3]. The conclusion follows from the mapping of the electron-spin coupling to the well-studied problem [2] of disordered Luttinger liquid.

1. M.Z. Hasan and C.L. Kane, Rev. Mod. Phys. 82, 3045 (2010).
2. T.Giamarchi and H.J.Schulz, Phys. Rev. B 37, 325 (1988).
3. B.L.Altshuler, I.L.Aleiner, V.I. Yudson Phys. Rev. Lett 111, 086401 (2013)

CPP 47.2 Thu 11:00 TRE Ma
Multiple Exciton Generation in Si and Ge Nanoparticles with high pressure core structures — ●STEFAN WIPPERMANN¹, MARTON VÖRÖS², DARIO ROCCA³, ADAM GALI⁴, GERGELY ZIMANYI², and GIULIA GALLI² — ¹Max-Planck-Institute for Iron Research, Düsseldorf — ²University of California, Davis — ³Universite de Lorraine, Nancy — ⁴Budapest University of Technology and Economics

Multiple exciton generation (MEG) in semiconductor nanoparticles (NPs) is a promising path towards surpassing the Shockley-Queisser limit in solar energy conversion efficiency. Recent studies demonstrate MEG to be more efficient in NPs than in the bulk, including Si. However, the increased efficiency is observed only on a relative energy scale in units of the gap: quantum confinement (QC) effects believed to be responsible for efficient MEG in NPs, also increase their optical gap, swiftly shifting the MEG threshold beyond the solar spectrum.

We present density functional and many body perturbation theory calculations of the electronic, optical, and impact ionization properties of Si and Ge nanoparticles (NPs) with core structures based on high-pressure bulk Si and Ge phases. Si and Ge particles with a BC8 or ST12 core structure exhibit significantly lower optical gaps and multiple exciton generation (MEG) thresholds, and an order of magnitude higher MEG rate than diamondlike ones of the same size (1).

- (1) S. Wippermann et al., Phys. Rev. Lett. 110, 046804 (2013)

CPP 47.3 Thu 11:15 TRE Ma
Advanced time-evolution method for optical absorption spectra calculations — ●TOBIAS SANDER and GEORG KRESSE — Computational Materials Physics, University of Vienna, Sensengasse 8/12, 1090 Vienna, Austria

The Green's function formalism from many-body perturbation theory gives access to electronic structure calculation within the quasiparticle picture, as well as provides for calculating optical absorption spectra. Within the traditional ansatz [1], a Bethe-Salpeter like equation for the polarizability is solved. This requires to diagonalize an in general non-hermitian and complex matrix (BSE matrix). Usually, the off-diagonal elements of the BSE matrix are neglected and this is referred to as Tamm-Dancoff approximation. The computational effort can be reduced by using the time-evolution ansatz [2] which avoids the matrix diagonalization. We present a method based on the time-evolution algorithm, that finally avoids storing and diagonalizing the BSE matrix. This leads to a reduction of the scaling w.r.t the system size N from N^5 to N^3 . Finally, we present first results for typical systems.

- [1] S. Albrecht, L. Reining, R. Del Sole, G. Onida, PRL 80, 4510

(1998)

- [2] W. G. Schmidt, S. Glutsch, P. H. Hahn, F. Bechstedt, PRB 67, 085307 (2003)

CPP 47.4 Thu 11:30 TRE Ma
New starting point for the calculation of optical properties — ●IGOR RESHETNYAK^{1,2} and LUCIA REINING^{1,2} — ¹Laboratoire des Solides Irradiés, École Polytechnique, CNRS, CEA-DSM, F-91128 Palaiseau, France — ²European Theoretical Spectroscopy Facility (ETSF)

The Bethe-Salpeter Equation based on Hedin's GW approximation to the self-energy is a powerful approach for describing electron-hole interactions in optical properties and photo-absorption spectra. However, in its current formulation it is both computationally heavy and displays cancellation effects not accounted for analytically. We discuss the sources of these cancellations and the possibility of putting them forward explicitly. We furthermore assess alternative formulations and sets of approximations to the BSE. For each of them we examine its behavior on model systems as well as their computational applicability. Finally we suggest possible directions for further investigations.

CPP 47.5 Thu 11:45 TRE Ma
Electron-Energy Loss and Inelastic X-ray Scattering of CuO from First Principles — ●CLAUDIA RÖDL, FRANCESCO SOTTILE, MATTEO GATTI, and LUCIA REINING — Laboratoire des Solides Irradiés, Ecole Polytechnique, CNRS, CEA-DSM, 91128 Palaiseau cedex, France and European Theoretical Spectroscopy Facility (ETSF)

Even though the strongly correlated transition-metal oxide CuO has many fields of application (potential absorber material in photovoltaic devices, pigment in glass and ceramics, building block of cuprate superconductors,...), surprisingly little is known about its electronic excitations from a theoretical point of view. The band gap and all electronic excitations in its vicinity are governed by the intricate interplay between itinerant $O\ 2p$ and localized $Cu\ 3d$ electrons. Complex many-body effects, that are still not well understood nowadays, determine the screening of the electron-electron interaction.

Electron-energy loss and inelastic x-ray scattering experiments yield direct access to the wave-vector- and frequency-dependent loss function $-\text{Im}\ \varepsilon_{\mathbf{G}\mathbf{G}'}^{-1}(\mathbf{q}, \omega)$, and, hence, to the screened Coulomb interaction W . We use time-dependent density-functional theory (TDDFT) to calculate the loss spectrum of CuO and discuss the occurring $d-d$ and plasmon excitations. This allows us, by comparing theory and experiment, to assess the quality of the screened Coulomb interaction which is a key quantity for many-body approaches, for instance, GW and Bethe-Salpeter calculations.

CPP 47.6 Thu 12:00 TRE Ma
Optical Spectra from Molecules to Solids: Insight from Many-Body Perturbation Theory — ●CATERINA COCCHI and CLAUDIA DRAXL — Humboldt-Universität zu Berlin, Institut für Physik and IRIS Adlershof, Berlin, Germany

The spurious long-range behavior of time-dependent (TD) density functional theory (DFT) is a well known source of error in describing bound excitons in solids. Remarkably, TD-DFT is often able to capture the optical features of isolated systems, even with the most simple exchange-correlation kernels, like the TD local density approximation. With the example of molecular crystals, we aim at solving the puzzle when and why TD-DFT can be relied on. We answer this question by confronting TD-DFT with many-body perturbation theory (GW and Bethe-Salpeter equation), which is the most accurate methodology to describe optical excitations in solids. Our results are obtained with the all-electron code "exciting" (<http://exciting-code.org>), where all the quantities entering the two formalisms are treated on the same footing [1]. In-depth analysis allows us to identify the shortcomings of TD-DFT in predicting the excitonic spectra of extended systems and to understand when this methodology is capable of providing correct results.

- [1] S. Sagmeister and C. Draxl, Phys. Chem. Chem. Phys. 11, 4451 (2009)

CPP 47.7 Thu 12:15 TRE Ma

Relativistic Solar Cells — ●PAOLO UMARI¹, EDOARDO MOSCONI², and FILIPPO DE ANGELIS² — ¹Dipartimento di Fisica e Astronomia, Università di Padova, via Marzolo 8, I-35131 Padova, Italy — ²Computational Laboratory for Hybrid/Organic Photovoltaics (CLHYO), CNR-ISTM, Via Elce di Sotto 8, I-06123, Perugia, Italy

Hybrid AMX₃ perovskites (A=Cs, CH₃NH₃; M=Sn, Pb; X=halide) have revolutionized the scenario of emerging photovoltaic technologies. Indeed, a rapid evolution led, very recently, up to 15% efficient solar cells. CH₃NH₃PbI₃ has so far dominated the field, while the similar CH₃NH₃SnI₃ has not been explored for photovoltaic applications, despite the reduced band-gap. Replacement of Pb by the more environment-friendly Sn would facilitate the large uptake of perovskite-based photovoltaics. Despite the extremely fast progress, the materials electronic properties which are key to the photovoltaic performance are relatively little understood. Here we develop an effective GW method incorporating spin-orbit coupling which allows us to accurately model the electronic, optical and transport properties of CH₃NH₃SnI₃ and CH₃NH₃PbI₃, opening the way to new materials design. The different CH₃NH₃SnI₃ and CH₃NH₃PbI₃ properties are discussed in light of their exploitation for solar cells, and found to be entirely due to relativistic effects.

CPP 47.8 Thu 12:30 TRE Ma

Solar nanocomposites with complementary charge extraction pathways for electrons and holes: Si embedded in ZnS — ●STEFAN WIPPERMANN¹, MARTON VÖRÖS², ADAM GALI³, FRANCOIS GYGI², GERGELY ZIMANYI², and GIULIA GALLI² — ¹Max-Planck-Institute for Iron Research, Düsseldorf — ²University of California, Davis — ³Budapest University of Technology and Economics

We propose that embedding silicon nanoparticles (NP) into amorphous, non-stoichiometric ZnS leads to promising nanocomposites for solar energy conversion. Using *ab initio* molecular dynamics simulations we show that upon high temperature amorphization of the host chalcogenide, sulfur atoms are drawn to the NP surface. We found that the sulfur content may be engineered to form a type II heterojunction, with complementary charge transport channels for electrons and holes, and that sulfur capping is beneficial to lower the nanoparticle gap, with respect to that of NPs embedded in oxide matrices. Our analysis was conducted using density functional theory with local and hybrid functionals and many body perturbation theory at the GW level.

CPP 47.9 Thu 12:45 TRE Ma

Ultraviolet photo-emission spectroscopies from Koopmans-compliant functionals — ●NGOC LINH NGUYEN¹, GIOVANNI BORGHI¹, ANDREA FERRETTI², ISMAILA DABO³, and NICOLA MARZARI¹ — ¹Theory and Simulations of Materials, École Polytechnique

Fédérale de Lausanne, Station 12, 1015 Lausanne, Switzerland. — ²Centro S3, CNR-Istituto Nanoscienze, I-41125 Modena, Italy — ³Department of Materials Science and Engineering, The Pennsylvania State University, University Park, USA.

We study the photo-electron properties of organic photovoltaic molecules using Koopmans-compliant functionals [1] as well as the Perdew-Zunger self-interaction correction (PZ-SIC) [2] to density-functional theory. A simple method for simulating ultraviolet photo-emission spectra (UPS) of molecules has been implemented. It is based on a plane-wave approximation for the final states to account for the spectra intensities. Our calculations show that Koopmans-compliant functionals provide ionization potentials and electron affinities closely comparable with those obtained by many-body perturbation theory (GW). In addition, we find that UPS spectra computed imposing the Koopmans' condition on the PZ-SIC functional are in remarkable agreement with experimental results.

Refs: [1] I. Dabo, A. Ferretti, N. Poilvert, Y. Li, N. Marzari, and M. Cococcioni, *Phys. Rev. B* 82, 115121 (2010); [2] J. P. Perdew and A. Zunger, *Phys. Rev. B* 23, 5048 (1981).

CPP 47.10 Thu 13:00 TRE Ma

Self-consistent dynamical embedding in real space — ●Wael Chibani¹, XINGUO REN^{1,2}, PATRICK RINKE¹, and MATTHIAS SCHEFFLER¹ — ¹Fritz Haber Institute of the Max Planck Society, Berlin, Germany — ²Key Laboratory of Quantum Information, USTC, Hefei, China

Density-functional theory with its local-density (LDA) and generalized gradient approximations (GGA) is known to fail for localized states. To extend *ab initio* approaches to this domain, we have devised an embedding scheme that facilitates the treatment of the physically important part of a system with electronic structure methods, that are computationally too expensive for periodic systems, whereas the rest of the periodic system is treated with computationally less demanding approaches, i.e. LDA/GGA, in a self-consistent manner. Our scheme is based on the concept of dynamical mean-field theory (DMFT) [1]. However, in contrast to the original DMFT formulation for correlated model Hamiltonians, we consider here the unit cell as embedded cluster in an *ab initio* way, that includes all electronic degrees of freedom. The performance of our scheme is demonstrated by treating the embedded region with hybrid functionals for simple bulk systems, e.g. Si or NiO. The total energy and the density of states converge rapidly with respect to the computational parameters and approach their bulk limit with increasing cluster size. By treating the embedded region with GW we were able to improve the band gap using only a small cluster. The effect of self-consistency in GW for the embedded region will also be addressed. [1] A. Georges *et al.*, *Rev. Mod. Phys.* **68**,14 (2006)