

## CPP 16: Focus Session: Frontiers of Electronic-Structure Theory: Correlated Electron Materials II (joint session O/MM/DS/TT/ CPP)

Organizers: Silke Biermann, Ecole Polytechnique, Palaiseau cedex, France; Paul R. Kent, Oak Ridge National Laboratory, USA; Matthias Scheffler, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin (Synopsis provided with part I of this session)

Time: Monday 15:00–17:15

Location: HL 001

CPP 16.1 Mon 15:00 HL 001

**Non-adiabatic Dynamics in Single-Electron Tunneling Devices with Time-Dependent Density Functional Theory** — ●NIKLAS DITTMANN<sup>1,2,3</sup>, JANINE SPLETTSTOESSER<sup>2</sup>, and NICOLE HELBIG<sup>3</sup> — <sup>1</sup>Institute for Theory of Statistical Physics, RWTH Aachen University, Germany — <sup>2</sup>Department of Microtechnology and Nanoscience (MC2), Chalmers University of Technology, Gothenburg, Sweden — <sup>3</sup>Peter-Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich, Germany

The recent advance of various single-electron sources in solid-state setups has sparked interest in the investigation of electronic transport at the single-particle level. In our recent work (N. Dittmann, J. Splettstoesser, N. Helbig, arxiv:1706.04547), we put forward time-dependent density-functional theory to calculate the dynamics of interacting electrons in single-electron tunneling devices. As a physical system, we analyze a single-electron source which is built by a quantum dot tunnel-coupled to a nearby electron reservoir and driven by a time-dependent gate voltage. By using analogies with quantum-transport theory, we extract a time-nonlocal exchange-correlation potential for a Hubbard U on-site interaction on the quantum dot. The time non-locality manifests itself in a dynamical potential step, which we explicitly link to physical relaxation time scales of the electron dynamics. Finally, we discuss prospects for simulations of larger mesoscopic systems.

CPP 16.2 Mon 15:15 HL 001

**Dissipative exchange-correlation functional in QED-TDDFT** — ●CAMILLA PELLEGRINI<sup>1</sup>, ILYA TOKATLY<sup>2,3</sup>, and ANGEL RUBIO<sup>2,4</sup> — <sup>1</sup>Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany — <sup>2</sup>Nano-bio Spectroscopy Group and ETSF Scientific Development Centre, Departamento de Física de Materiales, Universidad del País Vasco UPV/EHU, E-20018 San Sebastian, Spain — <sup>3</sup>IKERBASQUE, Basque Foundation for Science, 48001 Bilbao, Spain — <sup>4</sup>Max Planck Institute for the Structure and the Dynamics of Matter, Luruper Chausse 149, 22761 Hamburg, Germany

Time-dependent density functional theory has been recently extended to treat many-electron systems coupled to quantized electromagnetic modes. Here we discuss the implications of this approach for the theory of open quantum systems. In particular we show that in the limit of continuous spectrum of photon modes, QED-TDDFT naturally leads to time-dependent density functional theory for dissipative systems coupled to the Caldeira-Leggett bath. We consider the application to the Ohmic spin boson model and show that the developed approximation to the exchange-correlation functional describes the natural linewidth of the electronic linear density response function.

CPP 16.3 Mon 15:30 HL 001

**Electric and magnetic response properties of solids from the current density** — ●RUBÉN RODRÍGUEZ FERRADÁS<sup>1</sup>, PINA ROMANIELLO<sup>2</sup>, and ARJAN BERGER<sup>1</sup> — <sup>1</sup>LCPQ, University of Toulouse, France — <sup>2</sup>LPT, University of Toulouse, France

The evaluation of the macroscopic polarization and magnetization of solids is problematic when periodic boundary conditions are used because surface effects are artificially removed. This poses a problem unless surface effects can be reformulated in terms of bulk quantities [1-5]. In this work we show the advantage of calculating electric and magnetic response properties of solids using the current density as basic variable. An efficient approach to calculate the current density is time-dependent current-density-functional theory. We will show results for optical properties of solids using a recently developed functional [6]. We will also discuss how the magnetization can be described within this framework.

[1] F. Kootstra, P.L. de Boeij, and J.G. Snijders, *J. Chem. Phys.* **112**, 6517.

[2] J.A. Berger, P.L. de Boeij, and R. van Leeuwen, *Phys. Rev. B* **71**, 155104 (2005).

[3] P. Romaniello and P.L. de Boeij, *Phys. Rev. B* **71**, 155108 (2005).

[4] J.A. Berger, P. Romaniello, R. van Leeuwen, and P.L. de Boeij, *Phys. Rev. B* **74**, 245117 (2006).

[5] J.A. Berger, P.L. de Boeij, and R. van Leeuwen, *Phys. Rev. B* **75**, 035116 (2007).

[6] J.A. Berger, *Phys. Rev. Lett.* **115**, 137402 (2015)

CPP 16.4 Mon 15:45 HL 001

**Coupling Maxwell's equations to the time-dependent Kohn-Sham equations: near-field effects and electromagnetic backreaction** — ●RENE JESTAEDT<sup>1</sup>, MICAEL OLIVEIRA<sup>1</sup>, ANGEL RUBIO<sup>1,2,3</sup>, and HEIKO APPEL<sup>1</sup> — <sup>1</sup>Max Planck Institute for the Structure and Dynamics of Matter and Center for Free-Electron Laser Science, Germany — <sup>2</sup>Center for Computational Quantum Physics (CCQ), The Flatiron Institute, USA — <sup>3</sup>Nano-bio Spectroscopy Group and ETSF, Universidad del País Vasco, 20018 San Sebastián, Spain

Induced currents in large molecular and condensed matter systems are non-negligible and can affect the conductivity and the optical properties of the system. In the present work, we have implemented the real-time propagation of Maxwell's equations in Riemann-Silberstein representation to use standard unitary propagation techniques in the TDDFT code octopus [1]. The Maxwell and the Kohn-Sham system are coupled via a predictor-corrector method to obtain a self-consistent time-evolution of the total system [2]. Explicitly solving the microscopic Maxwell's equations also allows us to determine the optical properties of the system directly from the Maxwell fields. We show near-field effects of a full Maxwell-matter and matter-Maxwell coupling for plasmon excitations in metallic nanoparticles [2,3] and for ring-currents in organic molecules [2].

[1] Alejandro Varas et al., *J. Phys. Chem. Lett.* **2015**, **6**, 1891-1898 /

[2] R. Jestädt et al., (to be submitted) / [3] X. Andrade et al., *Phys. Chemistry Chem. Physics* **2015**, **17** 31371-31396

CPP 16.5 Mon 16:00 HL 001

**Enhancing excitation energy and charge transfer with strongly correlated light-matter interaction** — ●CHRISTIAN SCHÄFER<sup>1</sup>, MICHAEL RUGGENTHALER<sup>1</sup>, HEIKO APPEL<sup>1</sup>, and ANGEL RUBIO<sup>1,2,3</sup> — <sup>1</sup>Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany — <sup>2</sup>Center for Computational Quantum Physics (CCQ), The Flatiron Institute, 162 Fifth Avenue, New York NY 10010, USA — <sup>3</sup>Nano-bio Spectroscopy Group and ETSF, Departamento de Física de Materiales, Universidad del País Vasco UPV/EHU, San Sebastian, Spain

Förster excitation energy and charge transfer are fundamental processes of chemical reactions and connected to interesting quantities such as correlation. Often this correlation is taken as fixed property of the system.

In the current work, we present how the coupling to cavity photons in a minimal realistic molecular system can drastically alter transfer characteristics, e.g. renders the excitation transfer to be distance independent [1,2]. The photonic interaction can imprint fermionic correlation on arbitrary distances.

The exact real-space description is suited to describe transfer and correlation in a unprejudiced ab-initio picture and allows us to extend our insights beyond common quantum-optical approximations.

[1] X. Zhong et al., *Angew Chem Int Ed Engl.* **56(31)**, 9034 (2017).

[2] M. Sliotsky et al., *PRL* **112**, 076401 (2014).

CPP 16.6 Mon 16:15 HL 001

**Effects of electronic correlations on the magnetic properties of organometallic molecules** — ●SUMANTA BHANDARY and SILKE BIERMANN — Centre de Physique Théorique, Ecole Polytechnique, 91128 Palaiseau, France

The realm of molecular spintronics relies on the external accessibility of molecular magnetic states. In correlated organometallic complexes, a delicate balance between the crystal field, Coulomb repulsion and

dynamical hybridization between metal center and organic ligands dictates the electronic and magnetic properties and often poses challenges for an accurate theoretical modelling. We have employed density functional theory (DFT), the GW approach and Anderson's impurity model (AIM) technique to study the ground state electronic and magnetic properties of transition metal-based porphyrin and phthalocyanine molecules, both in the gas phase [1] as well as while adsorbed on surfaces. Our study reveals that the dynamical correlation effects are important in order to accurately estimate spin-transition energies, magnetic anisotropy energies as well as the ground state electronic configurations in the molecular complexes. We have explored the manipulation of surface molecule interactions to externally influence the electronic and magnetic properties of the molecular system.

[1] S. Bhandary, M. Schüler, P. Thunström, I. di Marco, B. Brena, O. Eriksson, T. Wehling, and B. Sanyal, *Phys. Rev. B* **93**, 155158 (2016).

CPP 16.7 Mon 16:30 HL 001

**Structural, electronic and optical properties of cubic and tetragonal SrTiO<sub>3</sub>: a DFT study 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.

SrTiO<sub>3</sub> (STO) is of fundamental interest as a substrate material in oxide electronics. The bulk undergoes a phase transition from the cubic to a tetragonal structure at T=105 K accompanied by characteristic antiferrodistortive rotations of the TiO<sub>6</sub> octahedra. We present a systematic comparison of the performance of the gradient corrected exchange correlation functional (GGA), the strongly constrained and appropriately normed (SCAN) meta-GGA and the hybrid functional HSE06 with respect to the electronic, structural and optical properties of cubic and tetragonal STO. For the tetragonal structure, SCAN gives a significantly improved description of the structural properties, comparable to HSE06, at a computational cost similar to GGA. The experimental band gap can be reproduced within SCAN with an on-site Hubbard term (+U), whereas within GGA the gap is underestimated even for very high U values. We calculate the optical spectrum for both phases, including many-body effects and excitonic corrections within the GW+Bethe-Salpeter equation approach, and compare this to previous theoretical results for the cubic phase [PRB 87, 235102 (2013)] and experiment [PRB 93, 075204 (2016)]. Funding by the DFG within SFB1242, project C02 is gratefully acknowledged.

CPP 16.8 Mon 16:45 HL 001

**Unveiling the mysterious magnetic state of superconducting iron under pressure** — ●MATTEO D'ASTUTO — Institut NEEL CNRS/UGA UPR2940 25 rue des Martyrs BP 166 38042 Grenoble cedex 9 FRANCE — IMPMC, UMR CNRS 7590, Sorbonne

Universités-UPMC University Paris 06, MNHN, IRD, 4 Place Jussieu, F-75005 Paris, France

Compressed iron undergoes a transition from bcc to hcp crystal structure with a loss of ferromagnetism. The magnetic state of the hcp phase has been debated for many decades and experiments give seemingly contradictory results. Mössbauer measurements find no magnetism, however x-ray emission spectroscopy finds remnant magnetism and Raman mode splitting suggests symmetry breaking due to antiferromagnetism. These paradoxical results are consistent with either a paramagnetic state with spin fluctuations faster than Mössbauer timescales or an antiferromagnetic state, afmII, which is undetectable with Mössbauer spectroscopy. We performed neutron powder diffraction measurements in the hcp phase and do not observe afmII order down to 1.8 K, while confirming the existence of a local magnetic moment in the hcp phase with x-ray emission spectroscopy and find it is intrinsic to this phase (1). This local magnetic moment disappears at 30–40 GPa, exactly the same pressure region where superconductivity disappears.

(1) B. W. Lebert, T. Gorni J.-P. Rueff, S. Klotz, M. Casula, A. Juhin, J. M. Ablett, F. Baudelet, T. Straessle, T. Hansen, A. Polian, P. Munsch, G. Le Marchand, Z. Zhang, M. d'Astuto, article in preparation.

CPP 16.9 Mon 17:00 HL 001

**Frist-principle and experimental characterisation of the electronic properties of CaGaSiN<sub>3</sub> and CaAlSiN<sub>3</sub>: impact of chemical disorder** — ●JAN MINAR<sup>1</sup>, ONDREJ ŠÍP<sup>2</sup>, ROBIN NIKLAUS<sup>3</sup>, JONAS HAUSLER<sup>3</sup>, and WOLFGANG SCGNICK<sup>3</sup> — <sup>1</sup>New Technologies Research Center, University of West Bohemia, Pilsen, Czech Rep., — <sup>2</sup>FZU, Academy of Sciences, Czech Rep — <sup>3</sup>Department of Chemistry, University of Munich, Munich, Germany

We report a detailed investigation of the electronic, mechanical and optical properties of the recently discovered nitridogallosilicate CaGaSiN<sub>3</sub> which has potential as a LED-phosphor host material. We focus on chemical disorder effects, originating from the Ga/Si site, and compared them to those of isostructural CaAlSiN<sub>3</sub>. We calculate the elastic moduli and the Debye temperature in terms of quasi harmonic approximation. Spectral properties like the joint density of states (JDOS) are evaluated and the absorption, reflectance and energy loss function are obtained from the dielectric function. The optical band gap of CaGaSiN<sub>3</sub> from experiment is compared to the electronic band gap in terms of electronic DOS and band structure calculations. All properties are evaluated for different ordering models of Ga/Si while the experimentally observed substitutional disorder is accounted for by utilizing the Coherent Potential Approximation (CPA). We conclude a shrinking of the band gap for both CaGaSiN<sub>3</sub> and CaAlSiN<sub>3</sub> due to atomic disorder, which is unfavorable for potential phosphor applications [1]. R. Niklaus, J. Minar, J. Häusler, W. Schnick, *Physical Chemistry Chemical Physics* 19 (13), 9292 (2017)