# DS 18: Frontiers of Electronic-Structure Theory: Focus on the Interface Challenge VI (joint session O/DS/CPP/TT)

Time: Wednesday 15:00–17:45                                                      Location: H9

DS 18.1  Wed 15:00  H9

**First-principles quantum transport simulations including strong correlation effects** — •Andrea Droghetti[1], Wilhelm Appelt[2], Liviu Chioncel[2], Milos Radonjić[3], Enrique Muñoz[4], Stefan Kirchner[5], David Jacob[1], Dieter Vollhardt[2], Angel Rubio[6], and Ivan Rungger[7] — [1]University of the Basque Country (Spain) — [2]University of Augsburg (Germany) — [3]University of Belgrade (Serbia) — [4]Pontificia Universidad Católica de Chile — [5]Zhejiang University (China) — [6]Max Planck Institute for the Structure and Dynamics of Matter (Germany) — [7]National Physical Laboratory (UK)

When magnetic molecules are brought into contact with metals the electron-electron interaction leads to the appearance of the correlated Kondo state. In this talk we will present the results of first-principles calculations for the electronic structure and the linear-response conductance of radical molecules adsorbed on metallic surfaces in the Kondo regime [Phys. Rev. B 95, 085131 (2017), Nanoscale 10, 17738 (2018)]. In particular we will outline the methodological approach as implemented in the Smeagol electron transport code and we will benchmark the results against experiments. The method relies in the first place on the combination of Density Functional Theory with the Green's functions technique. We will explain how a molecular devices is projected onto an effective Anderson impurity problem, which is then solved either by continuum time quantum Monte Carlo or numerical renormalization group. Finally, we will describe some work-in-progress aimed at computing transport properties beyond linear-response.

DS 18.2  Wed 15:15  H9

**Density functional theory for transport through correlated systems** — •Stefan Kurth — Univ. of the Basque Country UPV/EHU, San Sebastian, Spain — IKERBASQUE, Basque Foundation for Science, Bilbao, Spain — Donostia International Physics Center DIPC, San Sebastian, Spain

A recently proposed density functional formalism to describe electronic transport through correlated systems in the steady state uses both the density on the junction and the steady current as basic variables. The corresponding Kohn-Sham system features two exchange-correlation (xc) potentials, a local xc potential and an xc contribution to the bias, which are universal functionals of the basic variables.

A recent parametrization of the xc potentials for the single-impurity Anderson model correctly incorporates both the Kondo and Coulomb blockade regimes. It allows for calculation of currents and differential conductances at arbitrary bias and temperature at negligible numerical cost but with the accuracy of sophisticated renormalization group methods. A time-local version of this functional is used to study the Anderson model under the influence of both DC and AC biases. We observe interaction-induced shifts of the photon-assisted conductance peaks, supression of the Kondo plateau at zero temperature and lifting of Coulomb blockade at finite temperature.

DS 18.3  Wed 15:30  H9

**Exact factorization of the many-electron wave function** — •Camilla Pellegrini[1], Antonio Sanna[1], and Eberhard K. U. Gross[1,2] — [1]Max Planck Institute of Microstructure Physics, Weinberg 2, 06120 Halle, Germany — [2]Fritz Haber Center for Molecular Dynamics, Institute of Chemistry, The Hebrew University of Jerusalem, Jerusalem 91904, Israel

The exact factorization approach [1], originally developed for a system of electrons and nuclei, is extended to a system of electrons only. This allows for a two-particle Schroedinger equation, which uniquely defines the exact effective interaction between two electrons in the medium. This interaction differs from the effective interaction, W, used in many-body Green's function techniques. In particular, it is spin-dependent. We illustrate the formalism for the simplest case of exchange interactions only.

[1] A. Abedi, N.T. Maitra, E.K.U. Gross, PRL 105, 123002 (2010).

DS 18.4  Wed 15:45  H9

**Many-body spectral functions from steady state density functional theory** — •David Jacob[1,2] and Stefan Kurth[1,2,3] — [1]Dpto. de Física de Materiales, Universidad del País Vasco UPV/EHU, San Sebastián, Spain — [2]IKERBASQUE, Basque Foundation for Science, Bilbao, Spain — [3]DIPC, San Sebastián, Spain

We present a scheme to extract the true many-body spectral function of an interacting many-electron system from an equilibrium density functional theory (DFT) calculation [1]. To this end we devise an ideal STM-like setup and employ the recently proposed steady-state DFT formalism (i-DFT) which allows to calculate the steady current through a nanoscopic region coupled to two biased electrodes [2]. In our setup one of the electrodes serves as a probe ("STM tip"). In the ideal STM limit of vanishing coupling to the tip, the system to be probed is in quasi-equilibrium with the "substrate" and the normalized differential conductance yields the exact equilibrium many-body spectral function. Moreover, from the i-DFT equations we derive an exact relationship which expresses the interacting spectral function in terms of the Kohn-Sham one. Making use of i-DFT xc functionals that capture Coulomb blockade as well as Kondo physics, the method yields spectral functions for Anderson impurity models in good agreement with NRG calculations. It is thus possible to calculate spectral functions of interacting many-electron systems at the cost of an equilibrium DFT calculation.

**References:** [1] D. Jacob and S. Kurth, Nano Lett. **18**, 2086 (2018) [2] G. Stefanucci and S. Kurth, Nano Lett. **15**, 8020 (2015)

DS 18.5  Wed 16:00  H9

**Magnetic phase transitions induced by pressure and magnetic field: the case of antiferromagnetic USb2** — •Leonid Sandratskii — Max Planck Institute of Microstructure Physics, Halle, Germany

Fascinating phenomena observed under applied pressure and magnetic field are currently attracting much research attention. Recent experiments have shown that application of the pressure or magnetic field to the USb2 compound induce the transformations of the ground-state antiferromagnetic (AFM) up-down-down-up structure to, respectively, ferromagnetic (FM) or ferrimagnetic configurations. Remarkably, the magnetic critical temperature of the FM state, induced by pressure, is more than two times smaller than the Neel temperature of the ground state. We performed density-functional theory (DFT) and DFT+U studies to reveal the origin of the unusual magnetic ground-state of the system and the driving mechanisms of the phase transitions. We investigate both the magnetic anisotropy properties and the parameters of the interatomic exchange interactions. To study pressure-induced effects we carry out calculations for reduced volume and demonstrate that the AFM-FM phase transformation indeed takes place but depends crucially on the peculiar features of the magnetic anisotropy. We also explain why the magnetic field that couples directly to the magnetic moments of atoms leads to the phase transition to the ferrimagnetic state whereas the pressure that does not couple directly to magnetic moments results in the FM structure.

DS 18.6  Wed 16:15  H9

**Charge localization at a weakly coupled molecule-metal system studied by linear expansion Δ-self-consistent field density-functional theory (ΔSCF-DFT)** — •Hadi H. Arefi[1,2], Daniel Corken[3], Reinhard Maurer[3], F. Stefan Tautz[1,2], and Christian Wagner[1,2] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]JARA-Fundamentals of Future Information Technology — [3]Department of Chemistry, University of Warwick, Coventry, United Kingdom

Predicting the charge arrangements at the interface between molecules and metals represents a formidable challenge for semi-local approximations to Density Functional Theory (DFT). This could become even more critical when molecules are only weakly coupled to the metal. Single-molecular devices based on such weak coupling have recently been created by molecular manipulation with a scanning probe microscope (SPM), where a single PTCDA (perylene-tetracarboxylic dianhydride) molecule was placed in a free-standing upright configuration either on a SPM tip [1] or on a pedestal of two adatoms on the Ag(111) surface [2]. There are indications that the mechanism stabilizing these unexpected configurations is linked to an integer charge transfer creating a singly occupied molecular orbital. We use the ΔSCF-DFT method [3] to confine charge on the LUMO of the PTCDA and study

the consequences with vdW-corrected DFT.

[1] C. Wagner et al. Phys. Rev. Lett. 115, 026101 (2015) [2] T. Esat et al. Nature 558, 573 (2018) [3] R. J Maurer, K. Reuter, JCP 139, 014708 (2013)

DS 18.7    Wed 16:30    H9

**Dispersion corrected density functional theory studies on PVDF/hydrated aluminium nitrate composite system** — •Ranjini Sarkar and Tarun Kundu — Indian Institute of Technology, Kharagpur

Electro-active polymer Polyvinylidene fluoride (PVDF) based ferroelectric composites have gained significant technological importance over conventional ceramic ferroelectrics. This article provides quantum chemical description of PVDF/ hydrated aluminium nitrate salt composite system in the light of density functional theory. Four monomer units of pristine $\alpha$ and $\beta$-PVDF, pure Al(NO3)3.9H2O, and PVDF/ Al(NO3)3.9H2O structures are optimized using dispersion corrected exchange correlation functional B3LYP-D and 6-311+G(d,p) basis set. Similar to the experimental findings, the current theoretical investigation also suggests that hydrogen bond interaction between PVDF and the hydrated salt molecule plays the major role for the enhancement of ferroelectric properties in this composite system. Non-covalent interaction phenomenon is elucidated on the basis of natural bond orbital analysis, Bader's quantum theory of atoms in molecules and reduced density gradient analysis. Chemical Reactivity and charge transfer mechanisms are explained using atomic-dipole corrected Hirshfeld population analysis, molecular electrostatic potential plot and frontier molecular orbital analyses, respectively.

DS 18.8    Wed 16:45    H9

**Band-structure effects in vertical layered material heterostructures** — •Nicholas D. M. Hine[1], Gabriel C. Constantinescu[2], Nelson Yeung[1], Siow-Mean Loh[1], José María Escartín[2], Cuauhtemoc Salazar Gonzalez[1], and Neil R. Wilson[1] — [1]Department of Physics, University of Warwick, United Kingdom — [2]Cavendish Laboratory, University of Cambridge, 19 JJ Thomson Avenue, Cambridge CB3 0HE, United Kingdom

Controlling the properties of layered material heterostructures is crucial to the success of devices based on the novel capabilities of 2D materials, yet theoretical insight has been limited by the large system sizes required to study rotated, incommensurate interfaces. We use linear-scaling DFT calculations with non-local vdW functionals to explore large-scale models of heterostructures of interest for device applications. Results will be presented for heterostructures including MoS2/MoSe2, MoSe2/WSe2, and other Transition Metal Dichalcogenide pairings, TMDCs with graphene and hBN substrates, and hBN/Phosphorene. Band-structure changes caused by stacking and rotation of the layers are obtained by unfolding the supercell spectral function into the primitive cells, incorporating spin-orbit coupling. Changes in spectral weight and band-structure between the monolayers and heterostructured interfaces show how lattice mismatch (MoS2/MoSe2) or spacer layers (Phosphorene/hBN/Phosphorene) can allow the component monolayers to retain more independence in heterostructures than in homo-stacks. Finally, applying electric fields allows the behaviour of gated structures to be predicted and explained.

DS 18.9    Wed 17:00    H9

**Global Trends in Calcium-Silicate-Hydrate Phases Identified by Infrared Spectroscopy and Density Functional Theory** — •Mohammadreza Izadifar, Franz Königer, Andreas Gerdes, Christof Wöll, and Peter Thissen — Karlsruhe Institute of Technology (KIT), Institute of Functional Interfaces (IFG), Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

Building and construction industry are at the same time the backbone and the driving force of our modern society. Nearly all our today's technical infrastructure is based on cement-based materials. Detailed,

spectroscopic investigations of model reactions on well-defined mineral substrates under UHV-conditions are largely lacking, thus prohibiting a validation of theoretical methods. Eight different Calcium-Silicate-Hydrate (CSH) phases, namely Tobermorite 14Å, Tobermorite 11Å, and Tobermorite 9Å, Wollastonite, Jaffeite, Jennite, $\gamma$-C2S, and $\alpha$-C2SH, are calculated with the help of Density Functional Theory using the Vienna ab initio simulation package (VASP). First, we take care of the mechanical properties of the material. Our results revealed that Jaffeite, $\gamma$-C2S, and $\alpha$-C2SH have a linear bulk modulus due to the monomer structure of silicate tetrahedra. Tobermorite 14Å and Jaffeite have the lowest and highest bulk modulus, respectively. In the second part, the optimized geometries allow for the precise calculations vibrational eigenmodes and frequencies by the force-constant (FC) approach. The proportions of C/S and H/C are major criteria for the classification of the calculated wavenumber of $\nu$(Si-O) for all phases in our model system.

DS 18.10    Wed 17:15    H9

**Origin of carbon 1s binding energy shifts in amorphous carbon materials** — •Michael Walter[1,4,5], Filippo Mangolini[2], Robert W. Carpick[3], and Michael Moseler[4,5] — [1]FIT, University of Freiburg, Germany — [2]University of Texas at Austin, USA — [3]University of Pennsylvania, USA — [4]Fraunhofer IWM, Freiburg, Germany — [5]Physikalisches Institut, Universität Freiburg, Germany

The quantitative evaluation of the carbon hybridization state by X-ray photoelectron spectroscopy (XPS) has been a surface-analysis problem for the last three decades due to the challenges associated with the unambiguous identification of the characteristic binding energy values of sp$^2$- and sp$^3$-bonded carbon. Here, we compute the binding energy values for model structures of various carbon allotropes, including graphite, diamond, doped-diamond, and amorphous carbon (a-C), using density functional theory (DFT). The large band-gap of diamond allows defects to pin the Fermi level, which results in large variations of the C(1s) core electron energies for sp$^3$-bonded carbon, in agreement with the large spread of experimental C(1s) binding energy values for sp$^3$ carbon. In case of hydrogen-free a-C, the C(1s) core electron binding energy for sp$^3$ carbon atoms is approximately 1 eV higher than the binding energy for sp$^2$-hybridized carbon. However, the introduction of hydrogen hinders the unambiguous quantification of the carbon hybridization state on the basis of C(1s) XPS alone. This work can assist surface scientists in the use of XPS for the accurate characterization of carbon-based materials.

DS 18.11    Wed 17:30    H9

**Mechanically tuned conductivity of graphene grain boundaries from first-principles calculations** — Delwin Perera, •Jochen Rohrer, and Karsten Albe — Institut für Materialwissenschaft, Technische Universität Darmstadt, Germany

Nanocrystalline graphene has recently been shown to have a strong piezoresistivity and strain gauge factors that are notably higher compared to single- or microcrystalline graphene [1]. The origin of the enhanced piezoresistivity in nanocrystalline graphene is still not fully understood, but several theoretical works suggest that grain boundaries are the main cause as these can evoke transport gaps.

In our work we test this assumption with density functional theory based transport calculations of graphene bicrystals. In particular, we extend our analysis of the interplay between grain boundary structure and transport properties [2] by including mechanical strain. We compute transmission functions and current-voltage curves and compare them with tight binding calculations. Our findings suggest that the strain-induced transport gap modulation can be fully described by the response of the bulk graphene band structure towards strain.

[1] Riaz *et al.*, Nanotechnology **26**, 325202 (2015)
[2] Perera *et al.*, Phys. Rev. B **98**, 155432 (2018)