O 31: Focus Session: Frontiers of Electronic-Structure Theory: Correlated Electron Materials III (joint session O/MM/DS/TT/CPP)

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(Synopsis provided with part I of this session)

Time: Tuesday 10:30–13:00 Location: HL 001

O 31.1 Tue 10:30 HL 001

Control and prediction of molecular crystal properties by multilevel strategies — •Jan Gerit Brandenburg — London Centre for Nanotechnology, Department of Physics and Astronomy, University College London, 20 Gordon Street, London, U.K.

Computational material science is a dynamic and thriving area of modern scientific research. Approaches based on the fundamental laws of quantum mechanics are now integral to almost any materials design initiative in academia and industry, underpinning efforts such as the Materials Genome initiative or the computational crystal structure prediction [1]. I will present a hierarchy of quantum chemical methods designed for this purpose, in particular targeting molecular crystals and their property prediction. The methods range from high-level diffusion Monte-Carlo (DMC) to London dispersion inclusive DFT, and thus, cover many orders of magnitudes in computational efficiency [2,3]. I will demonstrate the application to the 6^{th} blind test for organic crystal structure prediction. Comparisons to other state-of-the-art methods indicate both success and remaining challenges in the recent method developments [4].

[1] S. L. Price and J. G. Brandenburg, *Molecular Crystal Structure Prediction*; Elsevier Australia, **2017**.

[2] A. Zen, J. G. Brandenburg, J. Klimeš, A. Tkatchenko, D. Alfè, A. Michaelides, **2017**, *submitted*.

[3] J. G. Brandenburg, E. Caldeweyher, S. Grimme, *Phys. Chem. Chem. Phys.* **2016**, *18*, 15519.

[4] A. M. Reilly, et al. *Acta. Cryst. B* **2016**, *72*, 439.

O 31.2 Tue 11:00 HL 001

Advances in first-principles and model spin Hamiltonian simulations of point defects in semiconductors for quantum sensors and computing — •Viktor Ivády — Department of Physics, Chemistry and Biology, Linköping University, 581 83 Linköping, Sweden — Wigner Research Center for Physics, Konkoly-Thege Miklós út 29-33, 1121 Budapest, Hungary

First principles simulations play a key role in understanding the physics of point defects in semiconductors, while model spin Hamiltonian approaches are traditionally used to interpret experimental spin dependent observations and describe the spin dynamics of point defects. The development of novel point defect applications, such as quantum bit (qubit) and single photon emitter applications for quantum information processing and quantum sensing, requires detailed understanding of spin-related couplings and addressability of localized defects states in the bath of delocalized electrons that calls for further development and implementation of theoretical tools. Here, I report on my contribution to this field that covers 1) first principles studies for identification of point defect based qubits and single photon emitters , 2) method development for the description of point defects with correlated electron states, 3) implementation of zero-field-splitting calculation for point defect based qubits, 4) development of model spin Hamiltonian approaches for the simulation of optical dynamic nuclear polarization process (ODNP) of point defects, and 5) spin dynamic simulation of existing point defect qubits. As an outlook, I discuss the requirements toward fully-ab initio point defect spin dynamic simulations.

O 31.3 Tue 11:30 HL 001

Recent advances in first-principles modelling of correlated magnetic materials — •Yaroslav Kvashnin — Department of physics and astronomy, Uppsala University, BOX 516, 75120 Uppsala

Most of modern first-principles electronic structure studies of correlated materials are based on a combination of density functional theory and dynamical mean field theory (DFT+DMFT).

Addressing magnetic materials within DFT+DMFT has certain peculiarities. There are two recipes one can follow: either to account for magnetism within the DFT functional or to introduce it entirely within the self-energy. Both approaches have their flaws and advantages, which are well-known for DFT+U, but are not often discussed for DFT+DMFT. In my talk I will present a systematic comparison of the two methods and demonstrate the evidences favouring the use of non-polarised functionals.

Next, I will demonstrate how the obtained electronic structure information can be used to simulate finite-temperature magnetic properties in real materials. I employ a so-called two-step approach. First, I map the system on a Heisenberg model and extract the effective exchange parameters Jij's from DFT+DMFT. Then the atomistic spin dynamics simulations are used to simulate magnon spectra and predict the magnetic ordering temperatures.

I will demonstrate the power of such an approach by showing a direct comparison with available experimental data for a wide range of different materials.

O 31.4 Tue 12:00 HL 001

A first-principles approach to hot-electron-induced ultrafast dynamics at metal surfaces — •Reinhard J. Maurer — Department of Chemistry, University of Warwick, Gibbet Hill Road, CV4 7AL Coventry, UK

Low-lying electronic excitations in metals, so-called hot electrons, couple efficiently to molecular adsorbate motion. In doing so, they give rise to a number of curious experimental observations. This includes picosecond-scale energy loss of molecular adsorbate vibration, highly inelastic atomic and molecular scattering from metal surfaces, and light-assisted molecular desorption and chemical transformations, recently coined "hot-electron chemistry". In this talk, I will present a first-principles treatment of hot-electron-induced molecular dynamics based on Density Functional Theory that correctly captures the magnitude and mode-specificity of hot-electron mediated adsorbate-substrate energy transfer [1]. Utilizing our efficient all-electron local-orbital implementation of hot-electron-induced frictional forces based on Time-Dependent Perturbation Theory, [2] I will show how we correctly capture vibrational relaxation in large-scale metal-mounted molecular catalysts as well as the energy loss and coupled electron-nuclear dynamics of small molecular adsorbates in both thermal and laser-heated conditions. [3] We scrutinize our approach in comparison to recent Sum-Frequency Generation (SFG) spectroscopy and molecular beam scattering experiments. [1] Phys. Rev. Lett. 116, 217601 (2016) ; [2] Phys. Rev. B 94, 115432 (2017); [3] Phys. Rev. Lett. 118, 256001 (2017);

O 31.5 Tue 12:30 HL 001

Temperature effects in spin-orbit physics from first principles — •Bartomeu Monserrat — University of Cambridge, UK — Rutgers University, USA

The spin-orbit interaction drives a number of physical phenomena, including the band inversion in topological insulators and the spin splitting of electronic bands in inversion asymmetric crystals. In this work, we study the effects of finite temperature on such spin-orbit physics, including both thermal expansion and electron-phonon coupling effects [PRB 92, 184301 (2015)].

First, we describe the temperature dependence of the inverted gap in topological insulators. We find that increasing temperature reduces the topological gap in the Bi2Se3 family of materials, and we predict a temperature-induced topological phase transition in Sb2Se3 [PRL 117, 226801 (2016)].

Second, we study the temperature dependence of the spin splitting of electronic bands in both inversion symmetric and asymmetric crystals. We predict a dynamical spin splitting in centrosymmetric crystals and characterise the associated phenomenology in the cubic perovskite CsPbCl3 [arXiv:1711.06274]. In inversion asymmetric crystals, exemplified by the bismuth tellurohalides, we find that increasing temperature suppresses the static spin splitting arising from the Rashba effect [PRM 1, 054201 (2017)].