O 41: Focussed Session: Frontiers of Electronic Structure Theory IV (jointly with HL and TT)

Time: Wednesday 10:30–13:30 Location: H36

Topical Talk   O 41.1   Wed 10:30   H36
Challenges in data-intensive computational materials design: methodology and infrastructure. — •Boris Kozinsky — Robert Bosch Research and Technology Center, Cambidge, MA, USA

First-principles high-throughput screening of novel materials requires simultaneously inexpensive and accurate predictive computations of key properties. The first and most difficult challenge is the selection of the appropriate descriptors that are relevant to the material performance, and formulating the computational strategy. We will present examples of the computational design process in the fields of materials screening in batteries, catalysis and ferroelectrics. In each case, the critical issue is the selection of practical methods and validation using available data and higher-level models.

The second challenge is the need to establish a materials* informatics infrastructure able to automatically prepare and execute calculations on large classes of materials, to monitor calculations, and to store, retrieve and analyze complex data. We accomplish this by integrating storage databases with grid-enabled computational workflow into a powerful flexible environment adaptable to diverse purposes. We will discuss the requirements and possible use cases of such infrastructure elements. Together with collaborators, we are developing and making available this open-source software platform named AIDA (*Automated Infrastructure and Database for Atomistic design*) to make computational design efforts faster, easier, and fully integrated with automatic data collection and community sharing.

     O 41.2   Wed 11:00   H36
DFT+U(ω): A simplified approach for dynamical Hubbard corrections to DFT: — •David D. O'Regan and Nicola Marzari — Theory and Simulation of Materials, EPFL, Switzerland.

Numerous successful techniques have been developed to date, such as DFT+U, in which the screened Coulomb interactions, underestimated by approximate density functionals, are described more accurately via a mapping onto the Hubbard Hamiltonian. Charge screening is a dynamical process, generally, and so to fully realise the capability of such methods for improving optical and quasiparticle spectra, the Hubbard U describing these interactions must gain a frequency dependence.

We introduce a simple and inexpensive approach, named DFT+U(ω), and readily implementable within an existing DFT+U or constrained-DFT code, in which the dynamical U tensor appropriate to the rotationally-invariant DFT+U functional is computed and used to correct DFT or static DFT+U spectra perturbatively. The rotationally-invariant DFT+U(ω) self-energy interpolates between static DFT+U and GW. We recast the density-functional linear-response approach for the static U, where it is defined as an energy curvature, within the language of many-body perturbation theory. Here, its dynamical generalisation, and its relationship to methods such as constrained RPA, becomes readily apparent. A plasmon-pole type model is used for the inverse dielectric function, whereby low-energy parameters are computed using the appropriately renormalised density-functional linear-response, and high-energy parameters are inexpensively approximated via independent-particle RPA or ALDA.

     O 41.3   Wed 11:15   H36
Effective onsite interactions for materials with strong non-local Coulomb interactions — •Malte Schüler[1], Alexander Lichtenstein[2], Mikhail I. Katsnelson[3], and Tim Wehling[1] — [1]Institut für Theoretische Physik, Universität Bremen, D-28359 Bremen, Germany — [2]1. Institut für Theoretische Physik I, Universität Hamburg, D-20355 Hamburg, Germany — [3]Radboud University Nijmegen, NL-6525 AJ Nijmegen, The Netherlands

sp-Electron systems and low-dimensional materials often comprise strong local Coulomb interaction and non-local Coulomb interaction at the same time. Here we report on a method to map a generalized Hubbard model with non-local Coulomb interaction to an effective Hubbard model with strictly local Coulomb terms U^*. With the examples of graphene and silicene we show that the non-local Coulomb interaction can reduce the effective local interaction up to a factor of 2. The U^* model is defined by a variational principle with respect to the free energy. In this framework, obtaining the effective interaction requires non-local charge correlation functions for various parameters of the effective Hubbard model, which are calculated by the determi-

nant quantum Monte Carlo method. The temperature dependence of the effective interaction is discussed.

     O 41.4   Wed 11:30   H36
First-principles calculation of Hubbard U parameters for half-metallic ferromagnets — •Ersoy Sasioglu, Christoph Friedrich, and Stefan Blügel — Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

Correlation effects play an important role in the electronic structure of half-metallic ferromagnets (HM-FMs). They give rise to non-quasiparticle states above (or below) the Fermi energy at high temperatures [1], which reduce the spin polarization and as a consequence the efficiency of the spintronics devices. Employing the constrained random-phase approximation (cRPA) [2] within the full-potential linearized augmented-plane-wave (FLAPW) method [3], we have calculated the strength of the effective Coulomb interaction (Hubbard U) between localized $3d$ electrons in a series of HM-FMs like zincblende MnAs, half- and full-Heusler alloys NiMnSb and Co_2MnSi, respectively. The obtained Hubbard U parameters lie between 2.5 and 4.5 eV, being smallest for MnAs (Mn-$3d$) and largest for Co_2MnSi (Co-$3d$). The small value of U in the former can be attributed to the efficient screening of the As p electrons. For HM full-Heusler compounds the obtained U values are comparable to the ones in elementary $3d$ transition metals, while for half-Heusler compounds the U is a bit smaller.
[1] M. I. Katsnelson *et al.*, Rev. Mod. Phys. **80**, 315 (2008).
[2] E. Şaşıoğlu *et al.*, Phys. Rev. B **83**, 121101(R) (2011).
[3] http://www.flapw.de

     O 41.5   Wed 11:45   H36
Magnetic Spectroscopies with DFT + Hubbard (U,V) — •Emine Kucukbenli[1], Davide Ceresoli[2], and Nicola Marzari[1] — [1]Theory and Simulation of Materials, École Polytechnique Fédérale de Lausanne (CH) — [2]CNR-ISTM Institute of Molecular Science and Technology, Milan (IT)

Hubbard U corrections to exchange-correlation functionals, introduced to deal with correlated electrons, have been shown to greatly improve the accuracy of DFT calculations of transition-metals, thanks to their ability to restore piecewise linearity of energy as a function of occupations and thus correct self-interaction errors. In addition, U is not a fitting parameter but can be calculated ab initio, using linear-response (LR) formulations.

Nevertheless, transition-metal complexes that display both covalent and ionic character are poorly described by DFT+U. Recently, the addition of an intersite Hubbard V is suggested to restore the accuracy of DFT+U for these cases, while V can be obtained ab initio as well.

In this study we combine DFT+U+V with the gauge-invariant projector augmented wave (GIPAW) method, and calculate magnetic spectroscopic properties of systems with transition metals. We have been implementing this combination in Quantum ESPRESSO package, both for LR and the recently introduced converse approach, that uses a much simpler Berry-phase calculation of the orbital magnetization. We then examine the performance of U+V corrections in determining the structural properties and hyperfine interaction parameters of small transition-metal molecules and complex organometallic systems.

     O 41.6   Wed 12:00   H36
The magnetization of periodic solids from time-dependent current-density-functional theory. — •Arjan Berger[1,4], Nathaniel Raimbault[1,2], Paul de Boeij[3], and Pina Romaniello[2,4] — [1]Laboratoire de Chimie et Physique Quantiques, Université Paul Sabatier, IRSAMC, CNRS, Toulouse, France — [2]Laboratoire de Physique Théorique, CNRS, Université Paul Sabatier, IRSAMC, Toulouse, France — [3]Scientific Computing and Modeling, Amsterdam, The Netherlands — [4]European Theoretical Spectroscopy Facility

The evaluation of the macroscopic 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. For example, in case of the macroscopic polarization one can express the contribution of the charge density accumulated at the surface in terms of the bulk cur-

rent density through the continuity equation. Therefore one can work in the framework of time-dependent current-density functional theory to efficiently calculate the macroscopic polarization [1,2]. In this presentation we will study how also the magnetization can be described within this framework.

[1] F. Kootstra, P. L. de Boeij, and J. G. Snijders, J. Chem. Phys. 112, 6517 (2000).

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

O 41.7   Wed 12:15   H36

Structure, charge order, phonons and IR spectra of magnetite — •CHARLES PATTERSON — School of Physics, Trinity College Dublin, Dublin 2, Ireland.

The structure and charge order of magnetite (Fe_3O_4) below the Verwey transition have been contentious issues for over 70 years. An x-ray refinement for the full 112 atom, Cc space group crystal structure of magnetite was reported only recently [1]. Previous refinements were hampered by multiple domain twinning in samples, whereas the recent study was performed on a micron-sized sample with two domains. Here we report hybrid density functional theory (DFT) calculations for the crystal structure, charge order, vibrations and IR spectra of magnetite in the Cc (112 atom) and $P2/c$ (56 atom) unit cells. Charge order in the Cc structure is found to consist of Fe trimerons, both in experiment [1] and calculations.

[1] M. S. Senn, J. P. Wright and J. P. Attfield, Nature 481, 173 (2012).

O 41.8   Wed 12:30   H36

Crystalline and Magnetic Anisotropy of the 3d Transition-Metal Oxides — •ANDREAS SCHRÖN[1], CLAUDIA RÖDL[1,2], and FRIEDHELM BECHSTEDT[1] — [1]Institut für Festkörpertheorie und -optik, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany — [2]Laboratoire des Solides Irradiés, École Polytechnique, CNRS, CEA-DSM, 91128 Palaiseau, France

The 3d transition-metal oxides (TMOs) are subject of debate since many decades due to their extraordinary properties, such as the formation of an antiferromagnetic ordering AFM2 below their Néel temperature. Many studies, both experimental and theoretical, focus only on MnO and NiO, where the crystalline anisotropy is solely driven by exchange striction along the unique symmetry axis in the [111] direction and where the magnetic anisotropy is explained in terms of magnetic dipole interactions. In the other TMOs, FeO and CoO, however, orbital magnetization and spin-orbit interaction play an additional, yet crucial role for both crystalline and magnetic anisotropy.

We present density-functional theory (DFT) studies including an on-site interaction U of the crystalline and magnetic anisotropy of the electronic systems with non-collinear spins. The influence of the (semi-)local description of exchange and correlation (XC) by means of the local density approximation (LDA) and generalized gradient approximation (GGA) on the orbital moments in FeO and CoO and the implications on the aforementioned properties is investigated. We discuss the quenching of the orbital magnetization due to the gradient corrections.

O 41.9   Wed 12:45   H36

Electronic Structure and Magnetic interactions in 5d Ir oxide compounds — •VAMSHI MOHAN KATUKURI[1], VIKTOR YUSHANKHAI[2], RADU COLDEA[3], LIVIU HOZOI[1], and JEROEN VAN DEN BRINK[1] — [1]Institute for Theoretical Solid State Physics, IFW Dresden, Helmholtzstr. 20 01069 Dresden, Germany — [2]Joint Institute for Nuclear Research, Joliot-Curie 6, 141980 Dubna, Russia — [3]Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom

We investigate the correlated d-level electronic structure and magnetic interactions of 5d Ir oxide compounds by fully *ab initio* quantum-chemical many-body calculations on finite embedded clusters. The

wave-function quantum-chemical methods provide a promising alternative to density-functional-based approaches to the electronic structure of solids. The computed d-d excitations in square-lattice, honeycomb, pyrochlore, and chain-like iridates compare well with recent RIXS (resonant inelastic x-ray scattering) data. We also perform a detailed analysis of the relativistic spin-orbit wave functions and compute observables such as the $\langle \mathbf{L} \cdot \mathbf{S} \rangle$ ground-state expectation value of the spin-orbit operator. The latter is in principle accessible from x-ray absorption and provides information on the role of $t_{2g}-e_g$ couplings in the ground-state wave function and on the strength of non-cubic fields that lift the degeneracy of the t_{2g} levels. As concerns to the magnetic structure, we find, in honeycomb lattice structures, A_2IrO_3, the magnetic interactions strongly deviate from the proposed Kitaev-Heisenberg model, due to low-symmetry crystal fields.

O 41.10   Wed 13:00   H36

Bulk electronic structure of the diluted magnetic semiconductor GaMnAs through hard x-ray angle resolved photoemission — •JAN MINAR[1], IGOR DIMARCO[2], J. BRAUN[1], H. EBERT[1], A.X. GRAY[3], and CH. FADLEY[3] — [1]University of Munich, Munich, Germany — [2]University of Upsalla, Upsalla, Sweden — [3]UC Davis, Davis, USA

A detailed understanding of the origin of the magnetism in diluted magnetic semiconductors is crucial to their development for applications. Using hard X-ray angle-resolved photoemission [1] at 3.2 keV, we investigate the bulk electronic structure of the prototypical diluted magnetic semiconductor GaMnAs, and the undoped reference system GaAs [2]. The fully self-consistent combination of LSDA and dynamical mean field theory (DMFT) [3,4] and its combination with the one-step model of photoemission has been used to explain the experimental findings. Distinct differences are found between angle-resolved, as well as angle-integrated, valence spectra of GaMnAs and GaAs, in good agreement with theory. In addition to the standard LSDA based calculations the LSDA+DMFT approach shows an important effect of electronic correlations on the states close to the Fermi level. The combination of LSDA+DMFT and corresponding the Monte-Carlo simulations indicates an origin of ferromagnetism in GaMnAs and provides us a rather unifying picture of this controversial material.

[1]A. Gray et al., J. Minar et al., Nat. mat. 10, 759 (2011) [2] A. Gray, J. Minar et al., Nat. mat. 11, 957 (2012) [3] J. Minar, J. Phys.: Cond. Mat. (Topical Review) 23, 253201 (2011)

O 41.11   Wed 13:15   H36

Magnetic state of pyrochlore $Cd_2Os_2O_7$ emerging from strong competition of ligand distortions and longer-range crystal anisotropy — •NIKOLAY BOGDANOV[1], REMI MAURICE[2], IOANNIS ROUSOCHATZAKIS[1], JEROEN VAN DEN BRINK[1], and LIVIU HOZOI[1] — [1]IFW Dresden, Germany — [2]Groningen University, The Netherlands

We investigate the correlated d-level electronic structure of $Cd_2Os_2O_7$, a spin $S=3/2$ pyrochlore, by fully *ab initio* quantum-chemical many-body calculations on finite embedded clusters. The wave-function quantum-chemical methods provide a promising alternative to density-functional-based approaches to the electronic structure of solids. We describe the local Os d^3 multiplet structure, the precise mechanism of second-order spin-orbit coupling and zero-field splitting (ZFS), and determine the parameters of the effective spin Hamiltonian, i.e., the single-ion anisotropy, nearest-neighbor Heisenberg exchange as well as the Dzyaloshinskii-Moriya interactions.

The results indicate that local ligand distortions and the anisotropic Cd-ion coordination strongly compete, rendering the magnetic interactions and ordering crucially depend on these geometrical features. Without trigonal distortions a large easy-plane magnetic anisotropy develops. Their presence, however, reverses the sign of the ZFS and causes a large easy-axis anisotropy ($D \simeq -6.8$ meV), which in conjunction with the antiferromagnetic exchange interaction ($J \simeq 6.4$ meV) stabilizes an all-in/all-out magnetic order. The competition uncovered here is a generic feature of 227 pyrochlore magnets and opens new perspectives on the basic magnetism in these materials.