First-principles calculations of Hubbard $U$ parameters for half-metallic ferromagnets — Ensoy Sashoglu, 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 spintronic 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$_2$MnSi, respectively. The obtained Hubbard $U$ parameters lie between 2.5 and 4.5 $eV$, being smallest for MnAs (Mn-3d) and largest for Co$_2$MnSi (Co-3d). The small value of $U$ in the former can be attributed to the efficient screening of the $p$ 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.


HL 54.4 Wed 11:45 H36 Magnetic Spectroscopies with DFT + Hubbard ($U$V) — Emine Kucukbenli, Davide Ceriotti, and Nicola Marzari — Theory and Simulation of Materials, École Polytechnique Fédérale de Lausanne (CH) — 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 inter-site 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.

HL 54.6 Wed 12:00 H36 The magnetization of periodic solids from time-dependent current-density-functional theory — Arjan Berger, Nathaniel Raimhaut, Paul de Boer, and Pina Romaniello — 1Theory and Simulation of Materials, École Polytechnique Fédérale de Lausanne (CH) — 2Department of Chemistry and Physics Quantiques, Université Paul Sabatier, IRSAMC, CNRS, Toulouse, France — 3Laboratoire de Physique Théorique, CNRS, Université Paul Sabatier, Toulouse, France — 4Scientific Computing and Modeling, Amsterdam, The Netherlands — 5European 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.


HL 54.7 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 (Fe₃O₄) 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 P2₁/c (56 atom) unit cells. Charge order in the Cc structure is found to consist of Fe trimers, both in experiment [1] and calculations.


HL 54.8 Wed 12:30 H36
Crystalline and Magnetic Anisotropy of the 3d Transition-Metal Oxides — ●Andreas Schröd1, Claudia Röd1,2, and Friedhelm Bechtold — 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 on MnO and NiO, where the crystalline anisotropy is solely driven by exchange splitting 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.

HL 54.9 Wed 12:45 H36
Electronic Structure and Magnetic interactions in 5d Ir oxide compounds — ●Yamshi Mohan Katukuri1, Viktor Yushankhai2, Radu Coldea3, Liviu Hozoi1, and Jeroen van den Brink2,3 — 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 recentRIXS (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 (L,S) 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 t₂g–e₉ couplings in the ground-state wave function and on the strength of non-cubic fields that lift the degeneracy of the t₂g levels. As concerns to the magnetic structure, we find, in honeycomb lattice structures, A₁gIrO₃, the magnetic interactions strongly deviate from the proposed Kitaev-Heisenberg model, due to low-symmetry crystal fields.

HL 54.10 Wed 13:00 H36
Bulk electronic structure of the diluted magnetic semiconductor GαMnAs through hard x-ray angle resolved photoemission — ●Jan Minar1, Igor di Marco2, J. Braun3, H. Ebert1, A.X. Gray1, and Ch. Fadley3 — 1 University of Munich, Munich, Germany — 2 University of Upsala, 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 GαMnAs, 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 GαMnAs 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 GαMnAs and provides us a rather unifying picture of this controversial material.


HL 54.11 Wed 13:15 H36
Magnetic state of pyrochlore Cd₂O₃:O₂⁺ emerging from strong competition of ligand distortions and longer-range crystal anisotropy — ●Nikolay Bogdanov1, Remi Mauzerolle2, Ioannis Rousochatzakis3, Jeroen van den Brink3, and Liviu Hozoi1 — 1 Technische Universität Dresden, Germany — 2 University of Strasbourg, France — 3 Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom.

We investigate the correlated d-level electronic structure of Cd₂O₃:O₂⁺, 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 d5 multiply-occupied, 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 ≃ –6.8 meV), which in conjunction with the antiferromagnetic exchange interaction (J ≃ 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.