TT 21: Correlated Electrons: Electronic Structure Calculations

Time: Tuesday 9:30–11:45   Location: HSZ 201

TT 21.1   Tue 9:30   HSZ 201

Revealing electronic correlations in YNi_2B_2C using photoemission spectroscopy — •Aki Pulkkinen[1], Geoffroy Kremer[2], Vladimir Strocov[3], Frank Weber[4], Ján Minár[1], and Claude Monney[5] — [1]New Technologies-Research Center, University of West Bohemia, Pilsen, Czech Republic — [2]Département Physique de la Matière et des Matériaux, Institut Jean Lamour, Université de Lorraine/CNRS, France — [3]Paul Scherrer Institut, Swiss Light Source, Villigen, Switzerland — [4]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, Karlsruhe, Germany — [5]Département de Physique and Fribourg Center for Nanomaterials, Université de Fribourg, Fribourg, Switzerland

YNi_2B_2C is an intermetallic borocarbide superconductor with a complex electronic band structure that has a very strong Ni character near the Fermi energy. We present density functional theory (DFT) and one-step model of photoemission results for YNi_2B_2C and compare them to experimental soft x-ray angle resolved photoemission spectroscopy (SX-ARPES) measurements. We show that moderate electron correlations have to be included, using dynamical mean field theory (DFT+DMFT) applied to the Ni d-states, to reach the best agreement between the experimental and theoretical SX-ARPES spectra. The one-step model calculations allow us to identify the effect of DFT+DMFT on the energy bands observed in the SX-ARPES measurements.

TT 21.2   Tue 9:45   HSZ 201

Distortion and pressure induced phase transitions in double perovskite La_2CoTiO_6 — •Sromona Nandi[1], Ashis K. Nandy[2], and Rudra Sekhar Manna[1] — [1]Department of Physics, Indian Institute of Technology Tirupati, Tirupati 517619, AP, India — [2]School of Physical Sciences, National Institute of Science Education and Research, An OCC of Homi Bhabha National Institute, Jatni-752050, India

We study the density-functional theory spin-polarized electronic structure calculations for a double perovskite La_2CoTiO_6 where the magnetic moment comes from the Co^{2+} as Ti is in 4^+ state. Due to the rotation and volume mismatch between the TiO_6 and CoO_6 octahedra ($\angle$Co-O-Ti is 151–153°), the system is distorted at ambient pressure. The Co-moments align antiferromagnetically at low temperature and show an insulating transport property with a band gap of 1 eV, consistent with the experiment [1]. With the removal of distortion, i.e., ideal undistorted structure ($\angle$Co-O-Ti is 180°) shows a metallic behavior. Such metallic state can also be achieved with the application of hydrostatic pressure on the distorted structure accompanied by the quenching of Co-moment in the metallic phase which can be understood by the shifting of the exchange splitting of the Co 3d orbitals at the Fermi level. In addition, atomic projected density of states analysis of the Co sublattices shows an interesting half-metallic like intermediate state which could be interesting for spintronics applications.

[1] K. L. Holman *et al.*, J. Solid State Chem. **180**, 75 (2007)

TT 21.3   Tue 10:00   HSZ 201

Oxygen vacancies at the origin of pinned moments in oxide interfaces: the example of tetragonal $CuO/SrTiO_3$ — •Benjamin Bacq-Labreuil[1], Benjamin Lenz[2], and Silke Biermann[3] — [1]Institut Quantique, Université de Sherbrooke, Sherbrooke, Canada — [2]Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, Sorbonne Université, Paris, France — [3]Centre de Physique Théorique, Ecole Polytechnique, Palaiseau, France

Obtaining an accurate theoretical description of the emergent phenomena in oxide heterostructures is a major challenge. Recently, intriguing paramagnetic spin and pinned orbital moments have been discovered by x-ray magnetic circular dichroïsm measurements at the Cu $L_{2,3}$-edge of a tetragonal $CuO/SrTiO_3$ heterostructure. Using first principles calculations, we propose a scenario that explains both types of moments [1], based on the formation of oxygen vacancies in the TiO_2 interface layer. We show the emergence of a paramagnetic 2D electron gas hosted in the interface CuO layer. It is invisible at the Ti $L_{2,3}$-edge since the valence of the Ti atoms remains unchanged. Strong structural distortions breaking both the local and global fourfold rotation C_4 symmetries at the interface lead to the in-plane pinning of the Cu orbital moment close to the vacancy. Our results, and in

particular the pinning of the orbital moment, may have implications for other systems, especially monoxide/dioxide interfaces with similar metal-oxygen bond length and weak spin-orbit coupling.

[1] B. Bacq-Labreuil, et al, arXiv:2210.15084 (2022).

TT 21.4   Tue 10:15   HSZ 201

New superexchange paths due to synergetic breathing and hopping in corner-sharing cuprates — •Nikolay A. Bogdanov[1], Giovanni Li Manni[1], Sandeep Sharma[1,2], Olle Gunnarsson[1], and Ali Alavi[1,3] — [1]Max Planck Institute for Solid State Research, Stuttgart, Germany — [2]Department of Chemistry, University of Colorado Boulder, Boulder, CO, USA — [3]University Chemical Laboratory, Cambridge, UK

We present *ab initio* quantum chemistry calculations of the nearest-neighbour superexchange antiferromagnetic spin coupling J for two cuprates, Sr_2CuO_3 and La_2CuO_4. Good agreement with experimental estimates is obtained for both systems. We also find that J increases substantially as the distance between Cu and apical O is increased.

Analysis of the correlated wavefunctions together with extended superexchange models shows that there is an important synergetic effect of the Coulomb interaction and the O–Cu hopping. When an extra electron hops into Cu 3d orbital, Coulomb interaction leads to the orbital breathing (expansion), which on its turn reinforces electron hopping. This correlated breathing-enhanced hopping mechanism is a new ingredient in superexchange models. Our analysis shows that suppression of the described mechanism leads to a drastic reduction in the antiferromagnetic coupling, indicating that it is of primary importance in generating the strong interactions.

TT 21.5   Tue 10:30   HSZ 201

DFT with corrections for an efficient and accurate description of strong electron correlations in NiO — •Julian Gebhardt[1,2] and Christian Elsässer[1,2,3] — [1]Fraunhofer IWM, 79108 Freiburg — [2]Cluster of Excellence livMatS, University of Freiburg — [3]Freiburg Materials Research Center, University of Freiburg

An efficient and accurate description of the electronic structure of a strongly correlated oxide like NiO has been notoriously difficult. Here, we study the capabilities and limitations of two frequently employed correction schemes, a DFT+U on-site correction and a DFT+1/2 self-energy correction. While both methods individually are unable to provide satisfactory results, in combination they provide a very good description of all relevant physical quantities. Since both methods cope with different shortcomings of common DFT methods (using local-density or generalized-gradient approximations), their combination is not mutually dependent and remains broadly applicable. The combined approach retains the computational efficiency of DFT calculations while providing significantly improved predictive power.

TT 21.6   Tue 10:45   HSZ 201

$LaVO_3$: A true Kugel-Khomskii system — •Xuejing Zhang[1], Erik Koch[1,2], and Eva Pavarini[1,2] — [1]Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany — [2]JARA High-Performance Computing, 52062 Aachen, Germany

Almost 50 years ago, Kugel' and Khomskii (KK) showed in a classic paper that, in strongly correlated materials, orbital ordering can arise from pure superexchange interactions [1]. It can, however, also result from the crystal-field splitting via a lattice distortion, i.e., from electron-lattice coupling. Despite the intensive search, it has been hard to find an undisputed realization of a KK system. We identify that the t_{2g}^2 perovskite $LaVO_3$, in its orthorhombic phase, is a rare case of a system hosting an orbital-ordering KK phase transition rather than being controlled by the Coulomb-enhanced crystal-field splitting [2]. We find that, as a consequence of this, the magnetic transition is close to (and even above) the superexchange driven orbital-ordering transition, whereas typically magnetism arises at much lower temperatures than orbital ordering. To explore the effects of crystal-field splitting and filling, we compare to YVO_3 and t_{2g}^1 titanates. In all these materials the crystal field is sufficiently large to suppress the KK phase transition [3].

[1] K. I. Kugel' and D. I. Khomskii, Zh. Eksp. Teor. Fiz. **64**, 1429 (1973) [Sov. Phys. JETP **37**, 725 (1973)]
[2] X. J. Zhang, E. Koch, E. Pavarini, Phys. Rev. B **106**, 115110

(2022)

[3] X. J. Zhang, E. Koch, E. Pavarini, Phys. Rev. B **102**, 035113 (2020)

TT 21.7   Tue 11:00   HSZ 201

Orbital-selective metal-insulator transitions in the presence of strong magnetic fluctuations — •Evgeny Stepanov — CPHT, CNRS, École polytechnique, Institut Polytechnique de Paris, 91120 Palaiseau, France

Orbital-selective phenomena that can be realised in materials attract enormous interest. A prominent example is an orbital-selective Mott phase, where itinerant and localised electrons live in different orbitals of the same material. Since its theoretical prediction, the orbital-selective Mott transition has been intensively studied by the state-of-the-art theoretical methods that are based on local approximations to electronic correlations, namely the dynamical mean-field theory and the slave-spin approach. Nevertheless, the existence of the orbital-selective Mott phase in realistic materials is still heavily debated and has not yet been realised experimentally.

In this talk, I will show that consistently taking into account non-local magnetic fluctuations completely changes the physical picture in the Hubbard-Kanamori model, where the orbital-selective Mott transition was predicted in the framework of the local theories. I will show that upon lowering temperature the considered system undergoes the Néel transition to an ordered antiferromagnetic phase before it experiences the orbital-selective Mott transition. Importantly, the former occurs simultaneously for all orbitals, which eliminates the orbital selectivity from the metal-insulator transition. The possibility to realise an orbital selective Néel transition will also be discussed.

TT 21.8   Tue 11:15   HSZ 201

Quantum chemical study on cobalt(II) in honeycomb lattices based on multi-configurational approaches — •Thorben Petersen and Liviu Hozoi — Leibniz IFW Dresden, 01069 Dresden, Germany

Quantum Spin Liquids (QSLs) are fascinating materials that display quantum entanglement and host unconventional fractionated excitations. While QSL behavior was found in Cu(II) $3d^9$ cuprates, $4d^5$ RuCl$_3$, $5d^5$ iridates and triangular-lattice $4f^{13}$ oxides, we here explore $3d^7$ Co(II) compounds such as Na$_2$Co$_2$TeO$_6$, which form spin-orbit-entangled $J_{\text{eff}} = 1/2$ pseudo-spin moments at Co(II) sites that potentially realize Kitaev-magnetism. In this study, we apply multi-configurational quantum-chemistry methods like CASSCF and beyond to model the $t_{2g}^5 e_g^2$ magnetic sites in these honeycomb oxides by carefully designing an embedded cluster model to represent the extended bulk lattice. With this, we quantify the extent of the $3d$-site multiplet structure and spin-orbit coupling associated with the $^4T_{2g}$ ground state term. Moreover, our model will be validated against experimental magnetization data and first impressions on the $3d^7$-$3d^7$ intersite couplings will be given.

TT 21.9   Tue 11:30   HSZ 201

Calculation of atomic forces for correlated materials, preliminary results — •Dorota Gotfryd[1], Robinson Outerovitch[1], Marc Torrent[1], Ambroise van Roekeghem[2], and Bernard Amadon[1] — [1]CEA, DAM, DIF 91297 Arpajon CEDEX, France — [2]CEA, LITEN 38054 Grenoble, France

Atomic forces are crucial ingredientis for phonon spectra or molecular dynamics calculations. Within Abinit package, one can currently perform such calculations for weakly or strongly correlated systems, obtaining forces via density functional theory (DFT) or DFT + U methods, correspondingly.

We are working on extending the existing implementation of atomic forces in Abinit to DFT+DMFT (dynamical mean-field theory). Such application would allow for the calculation of forces close to metal/Mott insulator transition where the details of electronic correlations matter and DFT + U approximation is often insufficient for capturing the physics.

We derive the atomic forces for DFT + DMFT as functional derivatives of the free energy functional over the atomic positions and transform them to the projector-augmented wave (PAW) approximation of DFT using Wannier functions as correlated orbitals. We show that the DFT and DFT + DMFT versions of the forces differ by just two terms in the PAW language and we discuss the details of the implementation.