TT 7: Correlated Electrons: Electronic Structure Calculations

Time: Monday 9:30–12:30 Location: H 3025

TT 7.1 Mon 9:30 H 3025

Influence of the ionic coordination on the electronic structure of NiO and CoO calculated with a DFT+DMFT first principles many-body approach — •Daniel Mutter[1], Daniel F. Urban[1,2], Frank Lechermann[3], and Christian Elsässer[1,2] — [1]Fraunhofer IWM, 79108 Freiburg — [2]Freiburg Materials Research Center (FMF), Universität Freiburg — [3]Institut für Theoretische Physik III, Ruhr-Universität Bochum

The electronic structure of first-row transition-metal (TM) oxides is determined by strong electron correlation leading to interesting effects as, e.g., Mott-insulating behavior. Such effects can be related to the high energy excitation features in the spectral functions, which cannot be described by the quasi-particle density of states resulting from density-functional theory (DFT) calculations based on LDA or GGA exchange-correlation functionals. To derive the many-body spectral functions for the two TM oxides NiO and CoO, we applied a combination of DFT and dynamical mean field theory (DFT+DMFT). The influence of the crystal-field coordination of the TM cations by oxygen anions is studied by considering the two compounds in both rock-salt and zincblende structures. In addition to the electron correlation in the TM 3d orbitals, we account for the correlation in the oxygen 2p orbitals by augmenting DFT+DMFT with a self-interaction-correction pseudopotential scheme [1].

[1] F. Lechermann et al., Phys. Rev. B 100 (2019) 115125.

TT 7.2 Mon 9:45 H 3025

Bridging DFT+U and DFT+DMFT: Hartree-Fock approximation and Wannier Projectors — •Alberto Carta[1], Iurii Timrov[2], Peter Mlkvik[1], Alexander Hampel[3], and Claude Ederer[1] — [1]ETH Zurich, Zurich, Switzerland — [2]Paul Scherrer Institut, Villigen, Switzerland — [3]Flatiron Institute (CCQ), New York, USA

Materials exhibiting strong electron-electron interactions are effectively described by combining density functional theory and dynamical mean-field theory (DFT+DMFT), where electronic correlations are captured by mapping the system to a self-consistently determined impurity model.

By solving the impurity model within the Hartree-Fock approximation, the static-mean field limit of DMFT is recovered. In this contribution we show that DFT+DMFT with the Hartree-Fock approximation is equivalent to the well established DFT+U method.

We demonstrate this equivalence by benchmarking DFT+DMFT calculations against DFT+U for various example systems (MnO, NiO, and LuNiO$_3$), using the new integration of maximally localized Wannier functions as Hubbard projectors in DFT+U as implemented in Quantum ESPRESSO.

Finally, the flexibility of our implementation allows us to extend the application of DFT+U beyond the usual atomic-like orbitals. We explore the use of DFT+U on different basis sets, such as frontier orbitals in LuNiO$_3$ or a molecular orbital-like basis in VO$_2$, showcasing the versatility of our approach.

TT 7.3 Mon 10:00 H 3025

Parametrization of the Coulomb interaction matrix with point-group symmetry — •Coraline Letouzé, Guillaume Radtke, Benjamin Lenz, and Christian Brouder — Sorbonne Université, Muséum National d'Histoire Naturelle, UMR CNRS 7590, Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, IMPMC, 75005 Paris, France

In realistic (DFT+DMFT) calculations of correlated materials, the matrix of the partially-screened electron-electron Coulomb interaction is usually approximated in spherical symmetry and parameterized by Slater integrals (or, equivalently, Racah parameters). Few works have considered the real point-group symmetry of the Coulomb matrix.

Here, Coulomb integrals are analyzed by considering both the point-group symmetry of the site occupied by the atom in the crystal or molecule and the permutation symmetries of the orbitals in the integrals. Explicit formulas are provided to calculate all integrals of the interaction tensor in terms of a minimum set of independent ones. The effect of a symmetry breaking is also investigated by describing Coulomb integrals of a group in terms of those of one of its subgroups.

Possible applications of the presented framework include the calculation of solid-state and molecular spectroscopies via multiplet techniques, dynamical mean-field theory, or the *GW* approximation.

[1] Phys. Rev. B **108**, 115137

TT 7.4 Mon 10:15 H 3025

Single-site DFT+DMFT for vanadium dioxide using bond-centered orbitals — •Peter Mlkvik, Nicola A. Spaldin, and Claude Ederer — ETH Zurich, Zurich, Switzerland

We present a combined density-functional theory and single-site dynamical mean-field theory (DMFT) study of vanadium dioxide (VO$_2$) using an unconventional set of bond-centered orbitals as the basis of the correlated subspace. VO$_2$ is a prototypical material undergoing a metal-insulator transition (MIT), hosting both intriguing physical phenomena and the potential for industrial applications. With our choice of correlated subspace, we can investigate the interplay of structural dimerization and electronic correlation in VO$_2$ in a computationally cheaper way than other state-of-the-art methods, such as cluster DMFT. Our approach allows us to treat the rutile and M1 monoclinic VO$_2$ phases on an equal footing and to vary the dimerizing distortion continuously, exploring the energetics of the different phases. The choice of basis presented in this work hence offers a complementary view on the long-standing discussion on the MIT in VO$_2$ and suggests possible future extensions to other similar materials hosting molecular-orbital-like states.

TT 7.5 Mon 10:30 H 3025

Origin of transitions inversion in rare-earth vanadates — •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

The surprising inversion of orbital- and magnetic-order transition with increasing rare earth radius in the ReVO$_3$ series (with Re=La, Pr, Tb, Y and Lu) is poorly understood[1]. Here –using a combination of the LDA+DMFT method and irreducible tensors decomposition[2]– we show that this remarkable behavior results from the competition between rare-earth-specific super-exchange couplings and lattice distortions. For systems with small Re ionic radius, we find that electron-lattice interaction stabilize orbital ordering. Increasing the rare-earth radius modifies the active super-exchange couplings, leading to a strongly enhancement of orbital super-exchange effects and G-type anti-ferromagnetism.[3]

[1] S. Miyasaka, Y. Okimoto, M. Iwama, and Y. Tokura, Phys. Rev. B 68 (2003) 100406(R).

[2] X. J. Zhang, E. Koch, and E. Pavarini, Phys. Rev. B 105 (2022) 115104; Phys. Rev. B 106 (2022) 115110.

[3] X. J. Zhang, E. Koch, and E. Pavarini, Manuscript in preparation (2023).

TT 7.6 Mon 10:45 H 3025

Neural-network-boosted exact diagonalization: A new approach and a new community code — •Pavlo Bilous[1], Louis Thirion[2], and Philipp Hansmann[2] — [1]Max Planck Institute for the Science of Light, Staudtstr. 2, 91058 Erlangen, Germany — [2]Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

The solution of quantum many-body systems (clusters) in truncated finite size Hilbert spaces presents a paradigm in many fields of research ranging from quantum chemistry to condensed matter physics. Whether the considered clusters are of real nature (atoms, molecules) or auxiliary (e.g. as used in dynamical mean-field theory), solving the eigenvalue equation for the Hamiltonian $\hat{H}\Psi = E\Psi$ with exact or numerical diagonalization procedures is severely limited due to the exponential growth of the Hilbert space dimension w. r. t. the single particle quantum numbers (orbitals/sites). However, it turns out that often only a small subset of basis states is sufficient to approximate expectation values of observables. The challenge is to identify this a priori unknown subset: In this talk we present an approach to converge observables like the ground state energy with successive basis extension and basis selection steps which are assisted by a neural-network (NN) classification scheme. Concrete examples show that the NN selection outperforms traditional truncation schemes dramatically and might push the boundaries of such calculations by an order of magnitude. The method is integrated in SOLAX: a new JAX-based parallelized GPU-accelerated Python code which we are currently developing.

15 min. break

TT 7.7 Mon 11:15 H 3025

Dynamical mean-field theory study of the spin-orbit insulator Ba_2IrO_4: Effective low-energy models from first principles — •Léo Gaspard[1], Francesco Cassol[2], Michele Casula[2], Benjamin Lenz[2], and Cyril Martins[1] — [1]Laboratoire de Chimie et Physique Quantiques, Université Toulouse III - Paul Sabatier , Toulouse, France — [2]Institut de Minéralogie de Physique des Matériaux et de Cosmochimie, Sorbonne Université, Paris, France

Iridium-based transition metal oxydes are generally described by a low-energy effective model based on $j_{\text{eff}} = \frac{1}{2}$ and $\frac{3}{2}$ spin-orbitals. Among those, the most famous is the spin-orbit Mott insulator Sr_2IrO_4 for which it is possible to build a $j_{\text{eff}}=\frac{1}{2}$ one band model, establishing a connection with the isostructural high-temperature superconductor copper oxydes. In Iridates, Coulomb interaction and spin-orbit coupling are of the same order of magnitude as the kinetic energy of the electrons. A model based on effective spin-orbitals thus treats spin-orbit coupling and electronic correlations at different levels of approximations. In this work, we carefully extract an effective Hubbard-Kanamori model for Ba_2IrO_4, a sister-compound of Sr_2IrO_4 which does not exhibit structural distortions. Starting from a DFT calculation, we use maximally localized Wannier functions and constrained Random Phase Approximation to parametrize the model. We then solve this model using Dynamical Mean Field Theory ,treating electronic correlations and spin-orbit coupling on equal footing. We will discuss the influence of the choice of the model space (full 5d manifold against t_{2g} manifold) on the computed observables.

TT 7.8 Mon 11:30 H 3025

Dynamical mean-field theory study of the spin-orbit insulator Ba_2IrO_4: the role of spin-orbit coupling in the Mott transition — •Francesco Cassol[1], Léo Gaspard[2], Michele Casula[1], Cyril Martins[2], and Benjamin Lenz[1] — [1]IMPMC, Sorbonne University - CNRS, Paris, France — [2]LCPQ, Université Paul Sabatier Toulouse III - CNRS, Toulouse, France

The discovery of the spin-orbit (SO) induced insulating ground state in Sr_2IrO_4 has triggered intense resarch efforts targeting materials with strong SO coupling. We focus here on Ba_2IrO_4, a sister compound of Sr_2IrO_4 with similar properties. In Ba_2IrO_4, the absence of structural distortions yields pseudo-spin states that are close in energy, casting doubts on the single band $j_{\text{eff}} = 1/2$ picture invoked for Sr_2IrO_4. Its simple structure makes Ba_2IrO_4 also an ideal candidate to systematically study the interplay between SO coupling and Coulomb interactions in the metal-insulator transition. Based on an effective three-band model of Ba_2IrO_4, we investigate the evolution of the Mott transition within dynamical mean-field theory (DMFT). The corresponding phase diagram is studied with respect to the variation of the relevant couplings and temperature. We clarify the topological role of SO coupling and show that the paramagnetic insulating phase is recovered for realistic electron-electron interactions, thus establishing Ba_2IrO_4 as a SO-induced Mott insulator. Comparing our calculations with available angle-resolved photoemission spectra, we finally discuss the limitations of a single-site DMFT treatment and the role of antiferromagnetic fluctuations.

TT 7.9 Mon 11:45 H 3025

Formation of spin-orbital entangled 2D electron gas in layer delta-doped bilayer iridate $La_\delta Sr_3Ir_2O_7$ — •Amit Chauhan[1], Arijit Mandal[2], and B. R. K. Nanda[2] — [1]Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Condensed Matter Theory and Computational Lab, Department of Physics, IIT Madras, Chennai-36, India

The state-of-the-art doping techniques pave the way to engineer non-trivial exotic quantum transport in strongly spin-orbit coupled quantum materials. By performing DFT calculations and formulating a multi-orbital Hubbard model, we predict the formation of a sharply confined spin-orbital entangled two-dimensional electron gas (2DEG) on a δ-doped bi-layer iridate $La_\delta Sr_3Ir_2O_7$. It differs from the conventional way of forming the 2DEG out of polar oxide heterostructures. The electron doping in the otherwise half-filled $J_{eff} = 1/2$ states destroy the Neel state of the IrO_2 layers near to the δ-doped layer, leading to partially filled Ir upper-Hubbard sub-bands which host the spin-orbital entangled electron gas. The gas is confined by a potential well formed by the positively charged LaO layer. The estimated electrical conductivity is giant and is of the order of 10^{19} $Sm^{-1}s^{-1}$. Our study will encourage experimenters to grow δ-doped structures on a wide class of spin-orbit correlated materials to explore the formation of 2DEG which is crucial for future quantum technologies.

[1] A. Chauhan *et al.*, Phys. Rev. Materials **7** (2023) 114409.

TT 7.10 Mon 12:00 H 3025

On the cuprates' universal waterfall feature: evidence of a momentum-driven cross-over — Benjamin Bacq-Labreuil[1], Chafic Fawaz[2], Matteo d'Astuto[2], Silke Biermann[1,3], and •Benjamin Lenz[4] — [1]Ecole Polytechnique, Palaiseau, France — [2]Institut Néel, Grenoble, France — [3]Collège de France, Paris, France — [4]Sorbonne Université, Paris, France

We study two related universal anomalies of the spectral function of cuprates, so called waterfall and high-energy kink features, by a combined cellular dynamical mean-field theory and angle-resolved photoemission study for the oxychloride $Na_xCa_{2-x}CuO_2Cl_2$ (Na-CCOC). Tracing their origin back to an interplay of spin-polaron and local correlation effects both in undoped and hole-doped (Na-)CCOC, we establish them as a universal cross-over between regions differing in the *momentum*-dependence of the coupling and not necessarily in the energy of the related quasiparticles. The proposed scenario extends to doping levels coinciding with the cuprate's superconducting dome and motivates further investigations of the fate of spin-polarons in the superconducting phase.

TT 7.11 Mon 12:15 H 3025

Electron glass phase with resilient Zhang-Rice singlets in $LiCu_3O_3$ — •Armando Consiglio[1], Gianmarco Gatti[2], Edoardo Martino[2], Tobias Hofmann[1], Ronny Thomale[1], Giorgio Sangiovanni[1], Domenico Di Sante[3], Martin Greiter[1], Marco Grioni[2], and Simon Moser[2,4,5] — [1]Institut für Theoretische Physik und Astrophysik and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, Germany — [2]Institute of Physics, Ecole Polytechnique Fédérale de Lausanne, Switzerland — [3]Department of Physics and Astronomy, University of Bologna, Italy — [4]Advanced Light Source, Berkeley, California 94720, USA — [5]Physikalisches Institut and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, Germany

$LiCu_3O_3$ is an antiferromagnetic cuprate where Li substitution disrupts the native arrangement of edge-sharing Cu(II)O and Cu(I) ions. Utilizing angle-resolved photoemission spectroscopy (ARPES) and density functional theory (DFT), we reveal two distinct insulating electronic subsystems arising from Li-induced disorder. The first involves a Cu dz^2/O p_z valence band dispersing on the Cu(I) plane, while the second showcases a resilient Cu $3d_{x^2-y^2}$/O $2p_{x,y}$ derived Zhang-Rice singlet (ZRS) band on the Cu(II)O planes. Li's impact stabilizes the insulating ground state, also inducing a unique 2D electron glass behavior in charge transport, as observed through ARPES. This study underlines that, despite disorder, the Cu(II)-derived ZRS remains largely unaffected from impurity scattering and implies a local segregation of Li and Cu atoms.