TT 30: Complex Oxides

Time: Wednesday 9:30–13:00 Location: HSZ 204

TT 30.1 Wed 9:30 HSZ 204

Planar superconducting resonators on $SrTiO_3$ and $KTaO_3$: GHz properties of quantum paraelectrics — Vincent T. Engl, Nikolaj G. Ebensperger, Lars Wendel, Marius Tochtermann, Ilenia Neureuther, Ishan Sarvaiya, Cenk Beydeda, and •Marc Scheffler — 1. Physikalisches Institut, Universität Stuttgart, Stuttgart, Germany

$SrTiO_3$ is a common substrate for perovskite thin films, and it hosts a superconducting interface when covered with $LaAlO_3$. Recently, similar superconducting interfaces based on $KTaO_3$ were realized. Any cryogenic electronics application using $SrTiO_3$ or $KTaO_3$ faces the quantum-paraelectric nature of both materials: upon cooling to temperatures of a few K, their very large real part ϵ_1 of the dielectric function complicates high-frequency device design, while the imaginary part ϵ_2 indicates rather high losses for $SrTiO_3$ at GHz frequencies.

Overcoming the challenges of the high ϵ_1, we present coplanar superconducting Nb microwave resonators on $SrTiO_3$ and $KTaO_3$ substrates, which we operate at temperatures down to 25 mK. For $SrTiO_3$ with $\epsilon_1 \approx 20000$ at low temperatures, we couple the coplanar resonator to a conventional 50 Ω feedline using a distant-flip-chip geometry.[1,2] With loaded quality factors exceeding 800 for $SrTiO_3$ and 8000 for $KTaO_3$ we advance the prospects of these quantum paraelectrics towards applications in oxide-based superconducting quantum circuitry.

[1] L. Wendel *et al.*, Rev. Sci. Instrum. **91**, 054702 (2020)
[2] V. T. Engl *et al.*, arXiv:1911.11456 [cond-mat.supr-con]

TT 30.2 Wed 9:45 HSZ 204

Revsisiting magnetic and orbital ordering in V_2O_3 — Louis-Victor Schäfer and •Maria Daghofer — Institut für funktionelle Materie und Quantentechnologien, Universität Stuttgart

We revisit the correlated bands of V2O3 using the variational cluster approximation. Starting from various sets of one-particle parameters discussed in the literature, double counting of correlations turns out to be relevant here, as also reported in the literature. Settling onto consistent parameters, we then find that a careful treatment of inter-layer dimers is crucial to reproduce the experimentally observed magnetic ordering. Additionally, bonds in the planes orthogonal to the dimers affects orbital densities. Antiferromagnetic ordering is the found to go in hand with orbital symmetry breaking within the e_g sector. We find any models that reproduce the measured a_{1g} orbital densities also leads to active orbital degrees of freedom above the Neel transition.

TT 30.3 Wed 10:00 HSZ 204

Direct imaging of valence orbitals using hard x-ray photoelectron spectroscopy — •Daisuke Takegami[1], Laurent Nicolaï[2], Yuki Utsumi[1], Anna Meléndez-Sans[1], Daria A. Balatsky[1], Cariad-A. Knight[1], Connor Dalton[1], Shao-Lun Huang[1], Chi-Sheng Chen[1], Li Zhao[1], Alexander C. Komarek[1], Yen-Fa Liao[3], Ku-Ding Tsuei[3], Ján Minár[2], and Liu Hao Tjeng[1] — [1]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany — [2]University of West Bohemia, Pilsen, Czech Republic — [3]National Synchrotron Radiation Research Center, Hsinchu, Taiwan

It was hypothesized already more than 40 years ago that photoelectron spectroscopy should in principle be able to image atomic orbitals. If this can be made to work for orbitals in crystalline solids, one would have literally a different view on the electronic structure of a wide range of quantum materials. Here, we demonstrate how hard x-ray photoelectron spectroscopy can make direct images of the orbitals making up the band structure of our model system, ReO3 [1]. The images are energy specific and enable us to unveil the role of each of those orbitals for the chemical bonding and the Fermi surface topology. The orbital image information is complementary to that from angle-resolved photoemission and thus completes the determination of the electronic structure of materials.

[1] D. Takegami *et al.*, Phys. Rev. Res. 4, 033108 (2022)

TT 30.4 Wed 10:15 HSZ 204

Orbital imaging of the spin state transition in $LaCoO_3$ — Brett Leedahl[1], Daisuke Takegami[1], Martin Sunderamnn[1,2], Hlynur Gretarsson[1,2], Alexander Komarek[1], Arata Tanaka[3], Maurits Haverkort[4], and •Liu Hao Tjeng[1] — [1]MPI-CPfS, Dresden, Germany — [2]DESY/PETRA-III, Hamburg, Germany — [3]Department of Quantum Matter, Hiroshima University, Japan — [4]Institute for Theoretical Physics, Heidelberg University, Germany

Here we have investigated the Co 3*d* orbital occupation in $LaCoO_3$ across the spin state transition using the recently developed x-ray-based orbital-imaging method. The images collected allow for a direct determination of the amount of the Co t_{2g} and e_g holes. We find that, at the lowest temperatures, the low-spin state with the nominally $t_{2g}^6 e_g^0$ electron configuration *does* have holes in the t_{2g} subshell which we attribute to the presence of spin-orbit interaction. The hole amount, however, sets limits to the minimum energy gap between the lattice-frozen low-spin and high-spin states. At high temperatures, we find that the high-spin state occupation is about *half* (!) of the value reported in the most recent literature. Implications for the ab-initio modeling of the spin-state transition process are discussed.

TT 30.5 Wed 10:30 HSZ 204

Directional ballistics in ultra-pure delafossite metals — •Michal Moravec[1,2], Graham Baker[3], Maja D. Bachmann[1,2], Philippa H. McGuinness[1,2], Markus König[1], Seunghyun Khim[1], and Andrew P. Mackenzie[1,2] — [1]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany — [2]School of Physics and Astronomy, University of St Andrews, St Andrews, United Kingdom — [3]University of British Columbia, Vancouver, Canada

Studying electrical transport in extremely pure materials has led to the discovery of novel phenomena. The delafossite metals, with their two-dimensional electronic structure and mean free paths in excess of 20 μm, are a recent addition to the ultra-pure materials class. Here we study electrical transport in bars of varied width in delafossite metals $PdCoO_2$ and $PtCoO_2$. The bar-like structures are sculpted in two crystal orientations using the focused ion beam and sequentially narrowed across the width corresponding to the mean free path. We observe a cross-over from Ohmic to ballistic transport and gradual increase in resistivity anisotropy between the two orientations, as the channel is narrowed. This anisotropy is not allowed by the symmetry of the bulk crystal lattice; it is an example of symmetry lowering due to the imposition of shapes of finite size and the Fermi surface anisotropy [1]. We also complement our measurements with numerical solutions to the Boltzmann equation including the ARPES Fermi surface parametrisation. We obtain a good qualitative agreement with resistivity the data.

[1] M.D. Bachmann et al. Nat. Phys. 18, 819 (2022)

TT 30.6 Wed 10:45 HSZ 204

Electronic structure of the Fe^{2+} compound $FeWO_4$: a combined experimental and theoretical X-ray photoelectron spectroscopy study — •Simone G. Altendorf[1], Daisuke Takegami[1], Anna Meléndez-Sans[1], Vanda M. Pereira[1], Chang-Yang Kuo[1,2,3], Chun-Fu Chang[1], Chien-Te Chen[2], Masato Yoshimura[2], Ku-Ding Tsuei[2], Antoine Maignan[4], Marcus Schmidt[1], Arata Tanaka[5], and Liu-Hao Tjeng[1] — [1]Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany — [2]National Synchrotron Radiation Research Center, Hsinchu 30076, Taiwan — [3]Department of Electrophysics, National Yang Ming Chiao Tung University, Hsinchu 30010, Taiwan — [4]CRISMAT, Normandie Univ, ENSICAEN, UNICAEN, CNRS, Caen 14000, France — [5]Quantum Matter Program, Hiroshima University, Higashi-Hiroshima 739-8530, Japan

Iron tungstate ($FeWO_4$) is one of the rare oxide compounds with a nearly pure divalent iron valence state. We investigate the electronic structure of $FeWO_4$ experimentally by X-ray photoelectron spectroscopy. Using various photon energies, cross-section effects allow to identify the individual contributions of the Fe and W states to the valence band. A comparison with band structure and full atomic multiplet configuration interaction calculations yields important insights into the correlations in the material.

The research in Dresden is partially supported by the DFG through SFB 1143.

TT 30.7 Wed 11:00 HSZ 204

Imaging the orbital-switching in Ti_2O_3 across the metal-insulator transition — •Paulius Dolmantas[1], Chun-Fu Chang[1], Martin Sundermann[1,2], Brett Leedahl[1], Andrea Amorese[1],

Hlynur Gretarsson[1,2], Alexander Komarek[1], Arata Tanaka[3], Maurits W. Haverkort[4], and Liu Hao Tjeng[1] — [1]MPI-CPfS, Dresden, Germany — [2]DESY/PETRA-III, Hamburg, Germany — [3]Department of Quantum Matter, Hiroshima University, Japan — [4]Institute for Theoretical Physics, Heidelberg University, Germany

Using the recently developed x-ray-based *orbital-imaging* method, we have been able to observe a switching of the Ti 3*d* orbital occupation across the metal-insulator transition in Ti_2O_3. We find that in the insulating state only the a_{1g} of the t_{2g} subshell is occupied, while in the metallic state also the e_g^π gets filled at the expense of the a_{1g}. The insulator-metal transition may then be viewed as a transition from a solid of isolated Ti-Ti c-axis singlet dimers into a solid of electronically partially broken dimers, where the Ti ions acquire additional hopping in the a-b plane via the e_g^π channel. These findings have implications for the ab-initio modeling of the metal-insulator transition.

15 min. break

TT 30.8 Wed 11:30 HSZ 204

Effect of cation dynamics on polar instabilities of perovskite derived structures — •Rinis Ferizaj[1], Florian Büscher[1], Peter Lemmens[1], Hiroshi Kageyama[2], and Tong Zhu[2] — [1]IPKM, TU Braunschweig — [2]Energy Chemistry, Univ. Kyoto, Japan

Perovskite derived structures have a huge variability in structural, electronic and magnetic properties. This makes them leading candidates for todays search for novel energy materials. Using Raman scattering we probe phonon dynamics in such material and focus to the cation dynamics to better understand transitions into different polar/nonpolar low temperature phases.

We acknowledge support by DFG GrK 1952/2, Metrology for Complex Nanosystems-NanoMet, and DFG EXC-2123 QuantumFrontiers - 390837967.

TT 30.9 Wed 11:45 HSZ 204

Anomalous magnetotransport in $SrIrO_3$ (111) films — •Ji Soo Lim[1], Merit Spring[1], Martin Kamp[1], Louis Veyrat[1,2], Axel Lubk[2], Bernd Büchner[2], Michael Sing[1], and Ralph Claessen[1] — [1]Physikalisches Institut and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, Würzburg, Germany — [2]Leibniz Institute for Solid State and Materials Research and Würzburg-Dresden Cluster of Excellence ct.qmat, Dresden, Germany

Complex oxides provides an inexhaustible playground for the study of interactions between charge, spin, orbital, and lattice degrees of freedom. In particular, spin-orbit coupling can be of comparable magnitude to electron correlations in 5d iridates, leading to exotic quantum phases such as spin liquids and correlated topological semimetals [1]. Perovskite $SrIrO_3$ (SIO) films grown along the <111> direction exhibit a buckled honeycomb lattice, and are predicted to be topological crystalline insulators with a relatively large band gap [2].

Here we report on the epitaxial growth of ultrathin SIO films on $SrTiO_3$ (111) that display a twinned perovskite-like superlattice structure with a periodicity of 3 unit cells and the observation of unusual magnetotransport properties, namely the anomalous Hall effect. We observe a sign change of the magnetoresistance and hysteresis loops with increasing coercive fields below 10 K. By comparing with theoretical results, we discuss the origin of the unusual transport behavior.

[1] J. Chakhalian et al., APL Mater. 8, 050904 (2020)
[2] P. M. Gunnink et al., J. Phys.: Condens. Matter 33, 085601 (2021)

TT 30.10 Wed 12:00 HSZ 204

Magnetoelectric coupling of terbium orthotantalates — •Xiaotian Zhang[1], Nicola Kelly[2], Cheng Liu[1], Sian Dutton[1], and Siddharth Saxena[1] — [1]Cavendish Laboratory, University of Cambridge, Cambridge, United Kingdom — [2]Department of Chemistry, University of Oxford, Oxford, United Kingdom

Quantum multiferroic materials form a new and emerging area of physics where one expects to find emergence of novel quantum phases induced by subtle coupling between spin and charge degrees of freedom at low temperatures. Experimental study of such phenomena is limited by the lack of model materials where magnetism and dielectric properties can be tuned using magnetic fields at low temperatures.

In a recent breakthrough, we found that $TbTaO_4$ exhibits enhancement in dielectric response below 2 K on application of magnetic field, indicating magnetoelectric coupling. Previously, using susceptibility and heat capacity measurements we showed that $TbTaO_4$ orders at $T_N = 2.25$ K; powder neutron diffraction (PND) was used to solve the magnetic structure, which is A-type antiferromagnetic.

These rare-earth tantalates $LnTaO_4$ (Ln = Y, La-Lu) are of wide interest as a result of their luminescent, proton-conducting, oxide-ion-conducting and dielectric properties. In addition, in the monoclinic M polymorph of the tantalates with Ln = Nd-Er, the magnetic Ln^{3+} ions are arranged on an elongated diamond lattice. Materials with such a magnetic lattice have the potential for unusual magnetic behaviour owing to the interplay of the crystal electric field with the (possibly competing) J1 and J2 interactions.

TT 30.11 Wed 12:15 HSZ 204

$S = 1$ dimer system $K_2Ni(MoO_4)_2$: A candidate for magnon Bose-Einstein condensation — •Benjamin Lenz[1], Bommisetti Koteswararao[2], Silke Biermann[3,4,5], Panchanan Khuntia[6,7], Michael Baenitz[7], and Swarup K. Panda[8] — [1]IMPMC, Sorbonne Université, Paris, France — [2]IIT Tirupati, Tirupati, India — [3]CPHT, Ecole Polytechnique, Palaiseau, France — [4]Collège de France, Paris, France — [5]Lund University, Lund, Sweden — [6]IIT Madras, Chennai, India — [7]MPI for Chemical Physics of Solids, Dresden, Germany — [8]Bennett University, Greater Noida, India

Dimerized quantum magnets provide a unique possibility to investigate the Bose-Einstein condensation of magnetic excitations in crystalline systems at low temperature. Here, we model the low-temperature magnetic properties of the recently synthesized spin $S = 1$ dimer system $K_2Ni(MoO_4)_2$ and propose it as a candidate material for triplon and quintuplon condensation. Based on a first-principles analysis of its electronic structure, we derive an effective spin dimer model that we first solve within a mean-field approximation to refine its parameters in comparison to experiment. Finally, the model is solved by employing a numerically exact quantum Monte Carlo technique which leads to magnetic properties in good agreement with experimental magnetization and thermodynamic results. We discuss the emergent spin model of $K_2Ni(MoO_4)_2$ in view of the condensation of magnetic excitations in a broad parameter regime. Finally, we comment on a geometrical peculiarity of the proposed model and discuss how it could host a supersolid phase upon structural distortions.

TT 30.12 Wed 12:30 HSZ 204

The crucial influence of side groups on magnetic superexchange - a modification of the Goodenough-Kanamori-rules — Dijana Milosavljevic[1], Oleg Janson[2], Stefan-Ludwig Drechsler[2], and •Helge Rosner[3] — [1]Max-Planck-Institut für Mikrostrukturphysik, 06120 Halle, Germany — [2]IFW Dresden, 01069 Dresden, Germany — [3]Max-Planck-Institut für Chemische Physik fester Stoffe, 01187 Dresden, Germany

According to the famous Goodenough-Kanamori-Anderson rules, the key structural feature that determines the magnetic exchange coupling constant for superexchange in magnetic insulators is the magnetic ion-ligand-magnetic ion bond angle. Here, we demonstrate that this angle is not the only determining factor. An at least equally important influence on the exchange coupling has the presence of side groups attached to the ligands. Applying density functional calculations and subsequently derived realistic parameters for a multiband tight-binding model, we provide a quantitative analysis for the example case of edge-sharing Cu-O chains with bond angles near 90 degrees. We find that a single parameter, the difference in onsite-energies of the ligand orbitals parallel and perpendicular to the Cu-O chain, is at least as important as the bond angle for sign and size of the superexchange. This parameter strongly depends on the position of side groups outside the superexchange pathway. For a fixed bond angle, changes of a side group position, only, can cause changes in the superexchange of several hundred Kelvin and thus dramatic changes in the magnetic properties.

TT 30.13 Wed 12:45 HSZ 204

A consistent microscopic magnetic model of the edge-sharing chain cuprate $CuSiO_3$ — •Dijana Milosavljevic[1,2], Andrei Gippius[3], Oleg Janson[4], Michael Baenitz[1], Yurii Prots[1], Johannes Richter[5], and Helge Rosner[1] — [1]MPI for Chemical Physics of Solids, Dresden — [2]MPI of Microstructure Physics, Halle — [3]Moskow, Russia — [4]IFW, Dresden — [5]MPI Physics of Complex Systems, Dresden

A unique place in the family of low-dimensional magnets occupies the Cu^{2+} compounds featuring a series of square-planar CuO_4 plaquettes connected via their edges, forming in this way a chain. The edge-shared plaquettes in Li_2CuO_2 show ferromagnetic ordering [1], whereas similar chains with increased Cu-O-Cu bond angle bridging neighboring Cu atoms in $CuGeO_3$, undergo a spin-Peierls transition [2]. A compound

with a Cu-O-Cu angle essentially identical to the one in Li_2CuO_2 and isostructural to the famous Spin-Peierls system $CuGeO_3$ is spin-1/2 cuprate $CuSiO_3$ [3]. In a combined experimental-theoretical approach, we derive a new magnetic model of $CuSiO_3$, consistent with all reported experimental results.

[1] E.L.M Chung et al., Phys. Rev. B **68**, 144410 (2003)
[2] M. Hase et al., Phys. Rev. Lett. **70**, 3651 (1993)
[3] H. H. Otto et al., Z. Kristallogr. **214**, 558 (1999)