TT 5: Nickelates I

Time: Monday 9:30–13:15 Location: H 3007

TT 5.1 Mon 9:30 H 3007

Theory of magnetic excitations in bilayer nickelate superconductor $La_3Ni_2O_7$ — •Steffen Bötzel, Frank Lechermann, Jannik Gondolf, and Ilya Eremin — Ruhr-Universität Bochum, theoretische Physik III, Bochum, Germany

Motivated by the recent reports of high-T_c superconductivity in $La_3Ni_2O_7$ under pressure, we analyzed theoretically the magnetic excitations in the normal and the superconducting state in this compound, which can be measured by inelastic neutron scattering. We show that the bilayer structure of the spin response allows to elucidate the role of the interlayer interaction and the Cooper-pairing in a very efficient way. In particular, we demonstrate the key difference between the potential $s_\pm$ and d-wave gaps, discussed recently, by comparing the corresponding response in the even and odd channels of the spin susceptibility. We show that mostly interlayer driven $s_\pm$ Cooper-pairing produces a single large spin resonance peak in the odd channel near the X point whereas several resonances are predicted for the d-wave scenario.

TT 5.2 Mon 9:45 H 3007

Electronic correlations and superconducting instability in $La_3Ni_2O_7$ under high pressure — Frank Lechermann, •Jannik Gondolf, Steffen Bötzel, and Ilya M. Eremin — Institut für Theoretische Physik III, Ruhr-Universität Bochum, D-44780 Bochum, Germany

Motivated by the report of superconductivity in bilayer $La_3Ni_2O_7$ at high pressure, we examine the interacting electrons in this system. First-principles many-body theory is utilized to study the normal-state electronic properties. Below 100 K, a multi-orbital non-Fermi liquid state resulting from loss of Ni-ligand coherence within a flat-band dominated low-energy landscape is uncovered. The incoherent low-temperature Fermi surface displays strong mixing between Ni-d_{z^2} and Ni-$d_{x^2-y^2}$ orbital character. In a model-Hamiltonian picture, spin fluctuations originating mostly from the Ni-d_{z^2} orbital give rise to strong tendencies towards a superconducting instability with B_{1g} or B_{2g} order parameter. The dramatic enhancement of T_c in pressurized $La_3Ni_2O_7$ is due to stronger Ni-d_{z^2} correlations compared to those in the infinite-layer nickelates.

TT 5.3 Mon 10:00 H 3007

Superconductivity in high-pressure $La_3Ni_2O_7$ — •Pascal Reiss[1], Minu Kim[1], Runze Zhang[1], Pascal Puphal[1], Matthias Hepting[1], Keita Masaki[2], Kentaro Kitagawa[2], Bernhard Keimer[1], and Hidenori Takagi[1,2,3] — [1]Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany — [2]Department of Physics, The University of Tokyo, Bunkyo, Tokyo, Japan — [3]Institute for Functional Matter and Quantum Technologies, University of Stuttgart, Germany

The recent observation of high-temperature superconductivity in $La_3Ni_2O_7$ under high pressures represents a remarkable discovery. However, superconductivity appears very fragile, and reports so far have differed greatly regarding the observation of a zero-resistance state, pressure range required, and the nature of the normal state.

In this talk, we will present our own high-pressure investigations of single crystal and powder $La_3Ni_2O_7$ samples and we will discuss several implications regarding the nature of the superconducting phase.

TT 5.4 Mon 10:15 H 3007

Investigation of Superconducting Phase in Polycrystalline $La_3Ni_2O_{7-\delta}$ — •Runze Zhang[1], Yuichiro Shiroki[1,2], Minu Kim[1], Pascal Reiss[1], and Hidenori Takagi[1,2,3] — [1]Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany — [2]University of Tokyo, Tokyo 113-0033, Japan — [3]Institute for Functional Matter and Quantum Technologies, University of Stuttgart, 70569 Stuttgart, Germany

The recent discovery of superconductivity in Ruddlesden-Popper (RP) nickelate $La_3Ni_2O_7$ under pressure has attracted significant research attention. However, the exact phase that facilitates this superconductivity remains unidentified, partly due to the complexities in managing RP stacking faults in single crystals. To address this issue, we present a comprehensive growth study focusing on identifying the potential superconducting phase using high-quality pure-phase polycrystalline $La_3Ni_2O_{7-\delta}$ samples. The implications on the critical role of oxygen deficiency will be discussed.

TT 5.5 Mon 10:30 H 3007

Correlated electronic structure of $La_3Ni_2O_7$ under pressure — •Viktor Christiansson[1], Francesco Petocchi[2], and Philipp Werner[1] — [1]University of Fribourg, Switzerland — [2]University of Geneva, Switzerland

The report of superconductivity with a record T_c for nickelates in bulk samples of bilayer $La_3Ni_2O_7$ (up to 78 K at pressures above 14 GPa) sets the stage for a continued interest in the nickelate superconductors. An important theoretical task is therefore to formulate a relevant model to describe this system, and to clarify its normal state properties. Here, we discuss the correlated electronic structure of the high-pressure phase for a minimal four-orbital low-energy subspace using different many-body approaches: GW, dynamical mean field theory (DMFT), extended DMFT (EDMFT), and GW+EDMFT.

We focus mainly on the nonlocal correlation and screening effects captured by GW+EDMFT which result in an instability towards the formation of charge stripes for the experimentally reported high-pressure $Fmmm$ structure, with the $3d_{z^2}$ as the main active orbital. While charge ordering has been found experimentally in the low-pressure phase, it is suppressed under application of pressure. We comment on this in relation to the crystal structure and possible rare-earth self-doping, since hole doping suppresses the ordering tendency in our model.

15 min. break

TT 5.6 Mon 11:00 H 3007

Critical role of interlayer dimer correlations in the superconductivity of $La_3Ni_2O_7$ — •Siheon Ryee[1], Niklas Witt[1,2], and Tim Wehling[1,2] — [1]Universität Hamburg, Hamburg, Germany — [2]The Hamburg Centre for Ultrafast Imaging, Hamburg, Germnay

The recent discovery of superconductivity in $La_3Ni_2O_7$ with $T_c \simeq 80\,K$ under high pressure opens up a new route to high-T_c superconductivity. This material realizes a bilayer square lattice model featuring a strong interlayer hybridization unlike many unconventional superconductors. A key question in this regard concerns how electronic correlations driven by the interlayer hybridization affect the low-energy electronic structure and the concomitant superconductivity. Here, we demonstrate using a cluster dynamical mean-field theory that the interlayer electronic correlations (IECs) induce a Lifshitz transition resulting in a change of Fermi surface topology. By solving an appropriate gap equation, we further show that the dominant pairing instability (intraorbital s-wave/interorbital $d_{x^2-y^2}$-wave) is enhanced by the IECs. The underlying mechanism is the quenching of a strong ferromagnetic channel, resulting from the Lifshitz transition driven by the IECs. Our finding establishes the role of IECs in $La_3Ni_2O_7$ and potentially paves the way to designing higher-T_c nickelates.

TT 5.7 Mon 11:15 H 3007

Synthesis of bulk Rare Earth Nickelates from infinite-layer to Ruddlesden-Popper — •Pascal Puphal[1], Vignesh Sundaramurthy[1], Valentin Zimmermann[1], Yu-Mi Wu[1], Y. Eren Suyolcu[1], Björn Wehinger[2], Masahiko Isobe[1], Bernhard Keimer[1], and Matthias Hepting[1] — [1]Max Planck Institut für Festkörperforschung, Heisenbergstraße 1, D-70569 Stuttgart, Germany — [2]European Synchrotron Radiation Facility, 71 Avenue des Martyrs, F-38043 Grenoble, France

Recently, rare-earth nickel oxides have emerged as a new class of unconventional superconductors. Where two types of structures have drawn particular interest. The first type comprises nickelates with the infinite-layer crystal structure which only exists via topochemical synthesis, such as $Nd_{0.8}Sr_{0.2}NiO_2$, showing superconducting transition temperatures T_c up to 20 K. The second type falls in to the Ruddlesden-Popper phase nickelates, where $La_3Ni_2O_7$ under hydrostatic pressure manifests a remarkably high Curie temperature T_C of 80 K. Despite these promising observations, the possibly distinct mechanisms driving the superconductivity in these two types of nickelates are not yet fully understood. In my talk I will show limitations in the doping and ways out of this issue via topochemistry for the first class of nickelates. We recently observed that the second class exhibits multiple crystallographic phases and a pronounced sensitivity to oxygen stoichiometry. Here, I will show how they affect their physical properties and superconducting mechanism.

TT 5.8 Mon 11:30 H 3007

Synthesis and characterization of electron-doped nickelate single crystals — •Vignesh Sundaramurthy, Pascal Puphal, Masahiko Isobe, Bernhard Keimer, and Matthias Hepting — Max-Planck-Institut für Festkörperforschung, Stuttgart, Deutschland

Transition metal oxides with their strong interplay between the lattice, spin and charge degrees of freedom exhibit a plethora of interesting properties. One example are perovskite rare-earth nickelates that show electronic and magnetic ground states, tunable via change of structural parameters and charge carrier doping. Substitution of the rare-earth site by tetravalent Ce ions serves this purpose, potentially leading to novel physical properties. However, the single crystal growth of these highly distorted and electron-doped nickelates is challenging [1], which will be elaborated in this talk. Furthermore, we will present initial results of topotactic reductions of Ce-substituted nickelates, aiming for the realization of single crystals with the infinite-layer structure that are isostructural and isoelectronic to superconducting cuprates.

[1] P. Puphal, V. Sundaramurthy et al., APL Mater. 11 2023) 081107

TT 5.9 Mon 11:45 H 3007

Orbital imaging of Ni3d states in the infinite-layer nickelate $\mathrm{LaNiO_2}$ — •Edgar Abarca Morales[1], Vignesh Sundaramurthy[2], Georg Poelchen[1], Martin Sundermann[1,3], Hlynur Gretarsson[1,3], Pascal Phupal[2], Matthias Hepting[2], Hao Tjeng[1], and Berit Goodge[1] — [1]MPI for Chemical Physics of Solids, Dresden, DE — [2]MPI for Solid State Research, Stuttgart, DE — [3]DESY, Hamburg, DE

The motivation for comparing nickelates and cuprate high-T_c superconductors has been strongly justified by the discovery of superconductivity in doped infinite-layer $\mathrm{NdNiO_2}$ [1], which is isostructural and with the same electron count ($3d^9$) as the cuprate analogue. However, apart from the considerable lower T_c in the nickelate, recent studies have shown that its electronic structure differs significantly from the cuprate [2-4]. Hence, direct observation of the nickelate electronic structure has become of central need. Orbital imaging through non-resonant inelastic x-ray scattering (sNIXS) has been recently used to directly probe the ground state of Ni in NiO [5]. Here we employ sNIXS to reveal the charge density around the Ni-site in $\mathrm{LaNiO_2}$, proving that the 3d hole structure is compatible with having an e_g symmetry. Moreover, we show that sNIXS is ideal to probe the ground state of other nickelates if efforts are taken towards the synthesis of single-crystalline samples.

[1] D. Li et al., Nature 572 (2019) 624
[2] M. Hepting et al., Nat. Mater. 19 (2020) 381
[3] B.H. Goodge et al., PNAS 118 (2021) e2007683118
[4] H. Lu et al., Science 373 (2021) 213
[5] H. Yavas et al., Nat. Phys. 15 (2019) 559

TT 5.10 Mon 12:00 H 3007

Strong orbital- and momentum-dependent correlation effects in an infinite-layer $\mathrm{NdNiO_2}$ — •Evgeny Stepanov[1], Matteo Vandelli[2], Alexander Lichtenstein[2], and Frank Lechermann[3] — [1]École polytechnique, France — [2]Universität Hamburg, Germany — [3]Ruhr-Universität Bochum, Germany

Layered nickel-oxide compounds have garnered significant attention since the discovery of superconductivity in this class of materials. The physical properties of layered nickelates originate from a complex interplay between strong local Coulomb correlations, spatial collective electronic fluctuations, orbital degrees of freedom, and a non-trivial band structure. Until now, even the most advanced numerical calculations could not account for all these important effects simultaneously.

In this talk we present results of accurate many-body D-TRILEX calculations for the layered $\mathrm{NdNiO_2}$ compound at stoichiometry and upon hole doping [arXiv:2311.09983 (2023)] performed in the framework of an ab-initio three-orbital model. Our calculations demonstrate that both spatial collective electronic fluctuations and orbital degrees of freedom are equally important. Considering both these effects in a self-consistent manner results in a strong momentum- and orbital-dependent renormalization of the electronic spectral function. This renormalization favors the formation of the charge density wave ordering, which originates from the intraband correlations within the Ni-d$_{z^2}$ orbital. We also find that the system displays strong antiferromagnetic fluctuations that stem from the Ni-d$_{x^2-y^2}$ orbital.

TT 5.11 Mon 12:15 H 3007

Charge-density wave in infinite-layer nickelates — Tharathep Plienbumrung[1], Jean-Baptiste Moree[2], Andrzej M. Oles[3], and •Maria Daghofer[1] — [1]FMQ, University of Stuttgart, Germany — [2]Waseda University, Tokyo, Japan — [3]Jagiellonian University, Kraków, Poland

We use the constrained random-phase approximation to derive a two-band model for infinite-layer nickelates. We find that a large variety of orbital wave functions lead to similar bands, but take particular care to arrive at a converged solution. The effective model then turns out to feature substantial long-range Coulomb interactions within and between bands. We next turn to the random-phase approximation and a variant of the variational cluster approach to investigate the effective model. In the variational cluster approach, interactions within the cluster are taken into account exactly, while those connecting clusters have to be decoupled in a mean-field approach. We find that the inclusion of inter-site Coulomb interactions tends to suppress long-range antiferromagnetism while favoring a charge-density wave. We find a charge pattern with periodicity of three sites, consistent with recent experimental results.

TT 5.12 Mon 12:30 H 3007

Feshbach resonances in cuprate and nickelate high-Tc superconductors — •Fabian Grusdt[1,2], Annabelle Bohrdt[2,3], Lukas Homeier[1,2], Hannah Lange[1,2,4], Henning Schloemer[1,2], Ulrich Schollwoeck[1,2], and Eugene Demler[5] — [1]Department of Physics and Arnold Sommerfeld Center for Theoretical Physics (ASC), Ludwig-Maximilians-Universität München, Theresienstr. 37, München D-80333, Germany — [2]Munich Center for Quantum Science and Technology (MCQST), Schellingstr. 4, D-80799 München, Germany — [3]University of Regensburg, Universitätsstr. 31, Regensburg D-93053, Germany — [4]Max-Planck-Institute for Quantum Optics, Hans-Kopfermann-Str.1, Garching D-85748, Germany — [5]Institute for Theoretical Physics, ETH Zurich, 8093 Zürich, Switzerland

Experimental advances in solids, quantum simulators, and numerical techniques allow unprecedented microscopic studies of the structure of strongly correlated quantum matter. Taking advantage of these achievements, we present evidence for the existence of a Feshbach resonance in Hubbard and t-J models commonly used to model cuprate and nickelate compounds. In a 2d square lattice, we show that attractive d-wave interactions between magnetic-polaron charge carriers are mediated through coupling to a near-resonant bosonic paired state of two holes. We present a closely related Feshbach picture for the bilayer nickelate superconductors under pressure, described by a mixed-dimensional t-J model. As an outlook we describe how higher critical temperatures can be reached, including in ultracold atom experiments that have previously observed strong pairing mixed dimensions.

TT 5.13 Mon 12:45 H 3007

Probing the pairing symmetry of superconducting nickelates through electron irradiation induced disorder — •Abhishek Ranna[1], Michal Moravec[1,2], Romain Grasset[3], Kyuho Lee[4], Bai Yang Wang[4], Marcin Konczykowski[3], Harold Y. Hwang[4], Andrew P. Mackenzie[1,2], and Berit H. Goodge[1] — [1]Max Planck Institute for Chemical Physics of Solids, Germany — [2]University of St. Andrews, UK — [3]LSI, Ecole Polytechnique, France — [4]Stanford University, USA

Superconducting infinite-layer nickelates were experimentally realized two decades after their theoretical prediction, generating significant interest for being isoelectronic to the $3d^9$ superconducting cuprates. Despite similarities between their phase diagrams, key distinctions in electronic landscapes and hybridizations have been identified. Direct investigations to probe the fundamental superconducting properties using photoemission spectroscopy and single-particle tunneling experiments have so far proved challenging due to the thin film geometry and surface degradation caused during *ex situ* topotactic reduction. We employ high-energy electron irradiation to controllably and systematically induce disorder, thereby examining the impact of pair-breaking defects on the superconductivity which can help elucidate the nature of the superconducting gap symmetry. Here, we present our initial findings on the evolution of electronic properties such as the superconducting transition temperature and residual resistivity with increasing disorder to shed light on the pairing symmetry and electronic behaviour of the infinite-layer nickelates.

TT 5.14 Mon 13:00 H 3007

Strain engineering of superconducting infinite-layer nickelates — •Berit Goodge[1,2], Dan Ferenc Segedin[3], Grace Pan[3], Charles Brooks[3], Julia Mundy[3], and Lena Kourkoutis[1] — [1]Cornell University, Ithaca, USA — [2]Max Planck Institute CPfS, Dresden, Germany — [3]Harvard University, Cambridge, USA

The superconducting monovalent layered nickelates demonstrate a key example of successful materials prediction and design based on proximity to a related system, namely the superconducting cuprates. The metastable nickel valence corresponding to the requisite electronic configuration, however, results in a complex synthetic route to stabilize these compounds involving high-quality thin film synthesis of higher-valent nickelate epitaxial thin films followed by topotactic chemical reduction to the desired square-planar phase. This two-step processes introduces competing epitaxial strain states in the precursor and target phases, which present both immense challenges as well as unique possibilities to leverage these strain interactions for bespoke tuning of the resulting film structure. Here we discuss both the limits to and possibilities for strain engineering in superconducting nickelates.