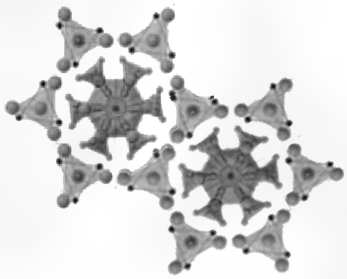
Scientific Report 2013-2019



Sinergia Network

Mott Physics Beyond the Heisenberg Model

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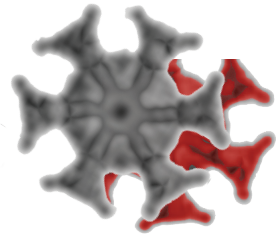
CONTENTS



VISION

The integrated network **Mott Physics Beyond the Heisenberg Model** (MPBH) united researchers across **Switzerland** in a collective effort to explore new aspects of Mott physics, in particular how spin-orbit interactions, in concert with strong electron correlations, lead to **new and exotic quantum materials and phenomena.**

The far-reaching achievements of the network were made possible only by assembling researchers with expertise in many complementary disciplines, who were hosted by the Swiss Federal Institute of Technology Lausanne (EPFL), the Paul Scherrer Institute (PSI), the University of Geneva (UNIGE), the Swiss Federal Institute of Technology Zurich (ETHZ) and the University of Zurich (UniZH).

The **holistic approach** of MPBH enabled us to synthesise and study promising materials by multiple techniques, thereby enabling profound advances in understanding their nature and quantum behaviour.

PAUL SCHERRER INSTITUT PSI

University of Zurich[UZH]

EPFL

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ETH zürich

SUMMARY

The Sinergia network **Mott Physics Beyond the Heisenberg Model** (MPBH) was assembled to unite leading groups in the domains of material synthesis, characterisation and theoretical analysis for a focused and coherent effort to study new magnetic materials beyond the Heisenberg approximation. During the 7-year project duration, we built a strong and multidisciplinary network, which achieved significant new scientific results, demonstrated by:

88 published articles

8 PhD students

15 Postdocs

494 experiments

21 materials synthesised

127 numerical simulations

5 conferences and workshops

NETWORK

The biggest achievement of MPBH was the **uniqueness and complementarity of the network created**. The following scientists contributed strongly to the project with their experience and expertise:

Prof. Henrik M. Rønnow, Head, Laboratory for Quantum Magnetism at the EPFL, is an expert in neutron scattering and low-temperature experimental physics. His research spans a range of quantum phenomena in model magnetic materials and in materials with unconventional electronic properties. Of particular importance to MPBH are his investigations of magnetic correlations in the cuprate high-temperature superconductors, which included discovering field-induced magnetism and the universality of the excitation spectrum. His career-long investigation of quantum effects in different materials realisations of the square-lattice Heisenberg antiferromagnet has recently been boosted by the development and application, in collaboration with colleagues both at EPFL and at PSI, of high-resolution resonant inelastic x-ray scattering (RIXS) techniques, whose successful application forms a core component of MPBH research.

Prof. Frédéric Mila, Head, Chair of Condensed Matter Theory at EPFL, is an expert in the theory of strongly correlated systems. He has worked on model Hamiltonians for high-Tc cuprates, organic conductors, orbitally degenerate systems, and frustrated quantum magnets. He has made seminal contributions to the study of spin-1/2 systems including the two-leg ladder, the kagome Heisenberg antiferromagnet and the Shastry-Sutherland model. He has developed a theory for the spin-orbital-liquid behavior of $LiNiO_2$ and obtained important results for the SU(4)-symmetric spin-orbital Kugel'-Khomskii model in geometries including ladders, square and honeycomb lattices. A contribution of direct relevance to MPBH concerns the two-dimensional Hubbard model on the triangular lattice at intermediate U/t, where his team demonstrated that there is a phase transition to a quantum spin liquid inside the Mott-insulating regime.

Prof. Christian Rüegg, Head, Neutrons and Muons Research Division (NUM) at PSI and Professor, Department of Quantum Matter Physics at the University of Geneva, has made internationally acclaimed studies of the complex order and elementary excitations in prototypical quantum magnets. He specialises in the control of magnetic quantum states by applied pressure and magnetic fields near their intrinsic quantum critical points, and in the quantum statistical models required to described such systems. MPBH has profited from his direct access to the neutron diffraction and spectroscopy instruments at the Swiss Spallation Neutron Source (SINQ). His team drives the technical frontier not only in neutron scattering but also in extreme sample environments (high magnetic fields, high pressure and ultra-low temperatures).

NETWORK

Prof. Johan Chang, Head, Laboratory for Quantum Matter Research at the University of Zurich, is investigating quantum phases emerging from strong electronic interactions. He uses angle-resolved photoemission spectroscopy (ARPES) and RIXS facilities at international synchrotrons, complemented by laboratory-based electrical and thermo-electrical transport measurements, to reveal the electronic structures and properties of correlated systems including high-temperature superconductors, strange metals and materials with density-wave instabilities and electronically driven metal-insulator transitions. He has used high-energy x-ray diffraction to explore quantum phase transitions controlled by an applied magnetic field or hydrostatic pressure. Within MPBH he has also established a new research direction by applying these methods to ruthenium oxide materials.

Dr. Thorsten Schmitt, Head, Spectroscopy of Novel Materials Group at PSI, operates one of the world's most advanced soft x-ray RIXS facilities at the ADRESS beamline of the Swiss Light Source (SLS). He studies correlated-electron materials displaying phenomena including superconductivity, metal-insulator transitions, charge order, magnetic order and low-dimensional magnetism. Dr. Schmitt has developed momentum-resolved soft x-ray RIXS at the SLS as a sensitive probe of the spin, charge, lattice and orbital dynamics in copper- and iron-based superconductors, as well as in quasi-one-dimensional cuprate materials, nickelates, vanadates, ruthenates and iridates.

Prof. Marco Grioni, Head, Laboratory for Electronic Spectroscopy at EPFL, investigates the electronic structure of low-dimensional and strongly correlated materials by two methods. One is high-resolution time-resolved ARPES (tr-ARPES), performed at EPFL and at various synchrotron radiation laboratories, and the other is RIXS. He has applied soft and hard x-ray spectroscopies to study a wide range of transition-metal and intermediate-valence materials and participated in ground-breaking RIXS measurements of magnetic excitations in the paradigmatic Mott insulator NiO and in cuprate superconductors. He also contributed to developing the high-resolution SAXES soft x-ray RIXS spectrometer at the SLS at PSI.

Prof. Janusz Karpinski, Group Leader, Laboratory of Solid-State Physics at ETHZ, has world-leading expertise in single-crystal growth under pressure. He has synthesised exotic structures which could not be achieved otherwise. One of his many original contributions was the Na-doped oxychloride cuprate, a model compound for understanding superconductivity in these materials. Recently, Prof. Karpinski has applied his expertise in high-pressure synthesis to searching for novel iron-based high-temperature superconductors.

NETWORK

Prof. Kazimierz Conder, Group Leader, Solid-State Chemistry group at PSI and Honorary Professor, Department of Materials at ETHZ, is an expert in solid-state chemistry, materials science and crystal growth. He and his collaborators have grown crystals of numerous complex oxide materials using the Traveling Solvent Floating Zone (TSFZ) method, including cuprates, manganates, orthoferrites, cobaltites and vanadates. They have built a unique gas-pressure system with an optical furnace to grow challenging low-dimensional spin systems including $SrCu_2(BO_3)_2$, $BiCu_2PO_6$ and $Sr_{14-x}Ca_xCu_{24}O_{41}$. Prof. Conder's team also uses the Bridgman method, most recently to grow crystals of the new iron-based superconductors ($FeSe_{1-x}Te_x$ and $(K,Rb,Cs)Fe_2Se_2$) and topological insulators (Cu-, Fe- and Mn-doped Bi_2Se_3), and the flux method for growing $Ca_3Ir_4Sn_{13}$.

Dr. Ekaterina Pomjakushina, Group Leader, Solid-State Chemistry Group at the Laboratory for Multiscale Materials Experiments at PSI, is an expert in crystal growth and materials engineering of compounds with novel electronic properties. She took over both the leadership role and all of the crystal-growth facilities from Prof. Conder after his retirement. Her high-quality single crystals have made possible some of the highlighted research performed on the large-scale facilities at PSI and elsewhere. Her major scientific achievements are in the fields of synthesis and study of complex transition-metal oxides, Fe-based superconductors and materials with novel quantum states, including magnetic Weyl semi-metals. Because many of these systems require high oxygen pressures for their synthesis, the new high-temperature, high-pressure optical furnace is a further key addition to the group's crystal-growing capabilities.

Prof. Lázló Forró, Head, Laboratory of Physics of Complex Matter at EPFL, is an expert in electron spin resonance, transport measurements and nanomaterials. In his laboratory, the pressure-dependence of the Mott state has been studied extensively in a large number of compounds, ranging from organics to oxides. A recent highlight is 1T-TaS_2, in which the commensurate CDW system hosts a Mott state, but moderate pressure suppresses this in favour of a robust superconducting state. Prof. Forró also heads a crystal-growth facility within the Institute for Condensed Matter Physics at EPFL, which through the work of Dr. H. Berger has created substantial scientific impact (over 6000 citations spanning several decades) through the production of high-quality crystals. Of particular value to MPBH has been the collaboration of Prof. Forró with Prof. Karpinski to establish a new facility for ultra-high-pressure synthesis and crystal growth at EPFL.

NETWORK
CONFERENCES

An **International Workshop on the topic of Mott Physics Beyond the Heisenberg Model** was held annually between 2012 and 2014, and again in 2016 as part of the Swiss Physical Society meeting.

On **June 25-28, 2012,** during the preparation phase for funding of the Sinergia project, a workshop on the topic was initiated by Profs. Mila, Rüegg and Rønnow and was held at EPFL with 40 participants.

On **October 28-31, 2013,** the MPBH workshop took place at the Centro Stefano Franscini in Monte Verita (Ascona). The number of participants grew to 51, with 22 invited international guest speakers, and the breadth of topics covered by the speakers expanded significantly.

On **October 26-27, 2013,** the Ascona workshop was preceded by an MPBH Training Module, a masters-level class hosted at PSI for over 20 SINERGIA and external PhD students. Two international experts gave preparatory lectures to the students, Dr. G. Khaliullin (MPI-Stuttgurt) presenting theoretical concepts covering coupled spin, orbit and charge (Mott) physics, with emphasis on iridates, and Dr. V. Pomjakushin (PSI) providing practical training in structural refinement for new materials.

On **September 16-18, 2014,** the MPBH workshop took place at Oriel College in Oxford, organised by Profs. A. Boothroyd (Oxford) and D. McMorrow (LCN, London) as well as Profs. Mila, Rüegg and Rønnow. This edition of the workshop had 52 participants including 16 invited international guest speakers, and active discussions and interactions were again widely appreciated by participants at every level.

On **August 23-25, 2016,** the MPBH workshop was organised by Profs. Mila, Rüegg and Rønnow within the Swiss Physical Society annual conference, held in Lugano. Sessions filling three days saw 15 invited presentations and a lively poster session, all attended by over 50 participants.

Oxford, 2014

SCIENTIFIC HIGHLIGHTS

Spin-orbit coupling (SOC) is a central thread in the search for novel physics in quantum materials, and as such formed a guiding theme for MPBH. A particularly promising avenue is the combination of SOC and strong electronic correlations in multi-orbital systems, a scenario realised in heavy transition-metal oxides composed of 4d and 5d elements. Iridium oxides (iridates) such as Sr_2IrO_4 are prime examples of systems where SOC plays a defining role in shaping the Mott-insulating ground state, and in fact for such 5d ions the spin-orbit entanglement essentially outplays the effectiveness of the usually influential crystal field. Of equal interest is the 4d series, which forms the complex regime where SOC and crystal-field energy scales are comparable, and here Ca_2RuO_4 is one focus material that displays a wealth of interesting physical properties surrounding its Mott-insulating ground state; its bilayer analogue, $Ca_3Ru_2O_7$, has also been the subject of intense research due to its puzzling electronic properties. The synergies within MPBH put the network in a unique position to make substantial progress in understanding the orbital physics governing the properties of these materials, and we highlight some of the most significant results in this section.

MATERIALS DISCOVERY

Throughout the funding period of MPBH, we have pursued and produced a large number of materials including doped and undoped cuprates, manganites, cobaltates, iron arsenides, ruthenates, iridates and some mixed-layer systems. Here we focus on an example system that is different from all of these.

$Sr_2Pt_{8-x}As$: in the search for materials with new electronic properties, the family of Pt-based ternary compounds is a rich and as yet little-investigated domain. Superconductivity and other unconventional electronic properties have been reported in all of $SrPt_3P$, SrPtAs, $SrPt_2As_2$, $SrPt_2Ge_2$, $SrPtGe_3$ and $Ca_2Pt_3Si_5$. Still completely unexplored is the Pt-rich compositional range of the Sr-Pt-As phase diagram. The range of stoichiometry we studied includes the theoretical composition $SrPt_3As$, analogous to $SrPt_3P$ (with superconducting critical temperature T_c = 8.4 K). A similar approach led us to the discovery of high-temperature superconductivity in the Fe-based pnictides, when $La[O_{1-x}F_x]FeAs$ was studied because of the known parent superconducting compound LaOFeP (although this has a much lower T_c of 4 K).

Due to the high partial pressure of As, we adopted a high-pressure synthesis technique to prepare $Sr_2Pt_{8-x}As$, which has an incommensurately modulated structure perpendicular to the layers, with the crystal symmetry described by the (3+1)-dimensional super-space group Xmma(00γ) [71]. Pt vacancies (x = 0.706) are unavoidable when using this synthesis route. These vacancies self-organise into lines in the plane of the $PtPt_6$ prism, resulting in an incommensurately modulated structure shown in **Figure 1.**

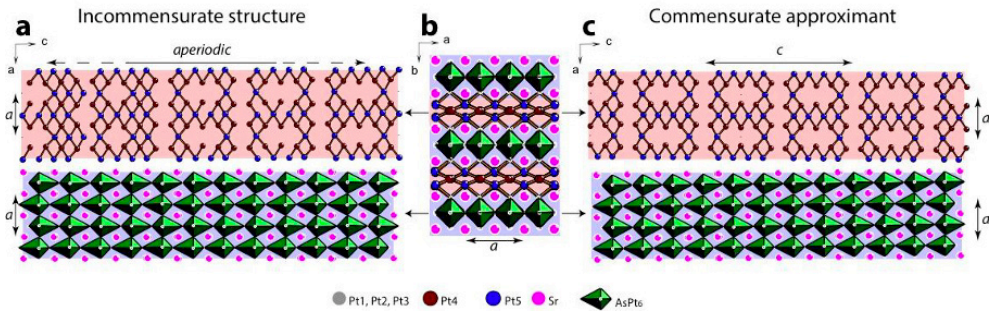

Figure 1. Sketch of the crystal structure of $Sr_2Pt_{8-x}As$.
(a,b) A portion of $Sr_2Pt_{7.294}As$ incommensurately modulated structure with incommensurability vector q = 0.396c*.
(b,c) Commensurate approximant $Sr_2Pt_{7.2}As$ with q = 0.4c* of the $Sr_2Pt_{7.294}As$ composition. Different colours show five different Pt atomic sites.

SCIENTIFIC HIGHLIGHTS

EXPERIMENTS

Mooij correlations in $Sr_2Pt_{8-x}As$. The modulated vacancy distribution in this stoichiometric system introduces scattering centres that influence the electronic structure and transport properties very strongly. The disorder potential is sufficient to suppress superconductivity even if conditions for it were favourable (large electron-phonon coupling, high density of states), but the surprising property of $Sr_2Pt_{7.294}As$ is the nearly flat temperature-dependence of the electrical resistivity **(Figure 2b)** [71]. This behaviour fits the description of Mooij correlations between scattering channels contributing to the electrical resistivity of disordered materials, an interpretation supported by the combination of the modulated structure with the dominant conductivity contributions being due to Pt-*d* electrons. The primary property of strongly disordered systems leading to this behavior is the resulting strong localisation, with the mean free path of the electrons being close to the Mott-Ioffe-Regel limit. A further consequence of the strong disorder is to mix the conductive channels along different axes **(Figure 2a)**, homogenising the resistivity tensor.

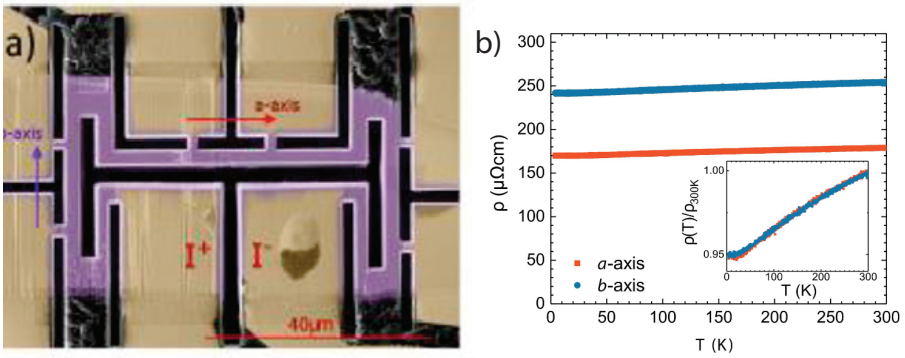

Figure 2. *(a)* Microstructured single crystal (purple in SEM image) for measuring electrical resistivity anisotropy. The starting lamella was extracted following crystallographic directions identified by x-ray diffraction. *(b)* Electrical resistivity measured along the a and b axes.

Metal-insulator transition (MIT) in rare-earth nickelates. We have investigated the MIT and the ground-state electronic structure of $ReNiO_3$ materials (Re = rare earth) using x-ray absorption spectroscopy (XAS) and RIXS at the Ni L_3-edge [33]. A RIXS map across the narrow Ni L_3 resonance of an $NdNiO_3$ thin film **(Figure 3)** reveals an unusual coexistence of bound and continuum excitations. Together with the characteristic signature of the orbital excitations, this allows us to conclude that the electronic ground state contains abundant O 2*p* holes and that the Ni sites assume a $3d^8$ electronic configuration, rather than the conjectured low-spin $3d^7$. Thus we demonstrate unambiguously that the $ReNiO_3$ series are self-doped, negative charge-transfer materials.

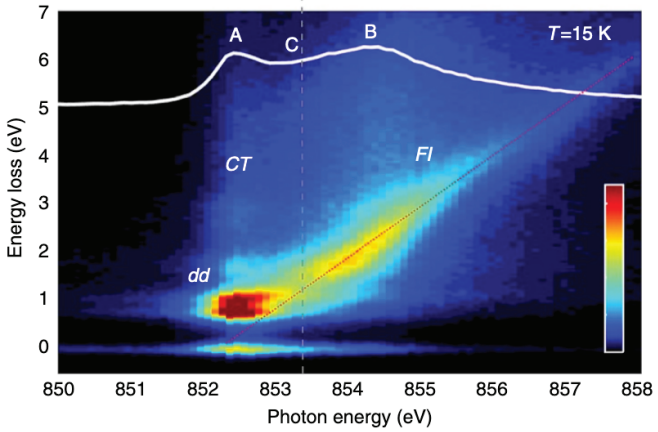

Figure 3. RIXS intensity map measured across the Ni L_3-edge at 15 K. dd, CT and Fl refer to different types of excitation. The grey dashed line indicates the incident energy giving the most pronounced effects due to its proximity to the MIT; the red dotted line is a guide to the eye.

SCIENTIFIC HIGHLIGHTS

Single- and bilayer iridates. We performed a comprehensive ARPES survey of the electronic structure of two square-lattice iridates, single-layer Ba_2IrO_4 (Ba-214) [11] and bilayer $Sr_3Ir_2O_7$ (Sr-327)[10]. In Ba-214, the data reveal a correlated insulator with Mott and spin-orbit physics playing similarly important roles. Data for Sr-327 **(Figure 4)** show two parabolic bands with maxima at the Γ point. These are the bonding (symmetric, S) and anti-bonding (anti-symmetric, AS) combinations of Ir 5*d* states in the two planes of the bilayer, and their splitting, 200 meV, is determined by the interplane interaction. The highest occupied states belong to a different band with a flat maximum (the top of the valence band) at the M point. A tight-binding calculation is sufficient to reproduce the overall features of the data and to estimate the J_{eff} character and symmetry of the states **(Figure 4)**.

For the single-layer case, the Ir 5*d* states are split by the spin-orbit interaction into a filled J_{eff} = 3/2 and a half-filled J_{eff} = 1/2 manifold, in which the Coulomb interaction opens a Mott gap. In Sr-327, the situation is more complex because the inter-layer coupling splits all the bands. The calculation reproduces very well the experimental bilayer splitting of the topmost J_{eff} = 3/2 subband, and suggests a splitting of the J_{eff} = 1/2 band near the M point. Our ARPES data clarify the origin of the gap in the bilayer compound and of the differences with respect to the single-layer compound, and allow an accurate determination of the symmetry character of the wavefunctions.

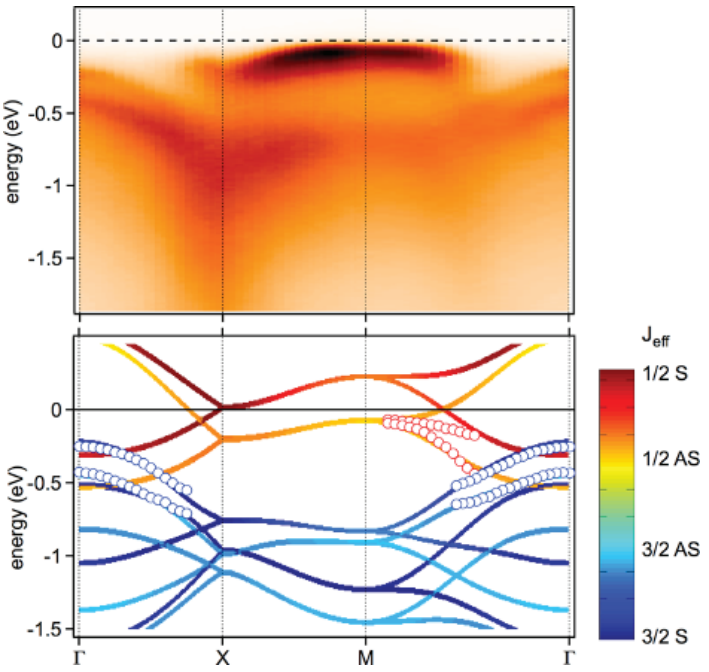

Figure 4. (Top) Experimental ARPES band dispersion of Sr-327. (Bottom) Tight-binding calculation illustrating J_{eff} and the symmetry character of the hybrid Ir 5*d* bands.

Dispersive magnetic and electronic excitations in iridate perovskites. We used O *K*-edge RIXS to study novel J_{eff} = 1/2 Mott physics in the layered perovskite iridates Sr_2IrO_4 and $Sr_3Ir_2O_7$. We observe the single-magnon dispersion branch in both samples **(Figure 5)**, providing conclusive evidence that O *K*-edge RIXS is capable of probing these excitations in 5*d* transition-metal oxides. This is enabled by the hybridisation between the O 2*p* and Ir 5*d* orbitals and the large (0.5eV) spin-orbit coupling in Ir. Taken together with previous Ir L_3 RIXS measurements, our results [70] demonstrate that O *K*-edge RIXS can be used to study single magnons and excitonic quasiparticles over a substantial part of the first Brillouin zone of layered iridates **(Figure 5)**. Because O *K*-edge RIXS has a high energy resolution for iridate materials and usually generates a strong response within a short attenuation length, it is uniquely suited for investigating elementary excitations in 5*d* transition-metal oxides, especially for thin-film samples, heterostructures and superlattices.

SCIENTIFIC HIGHLIGHTS

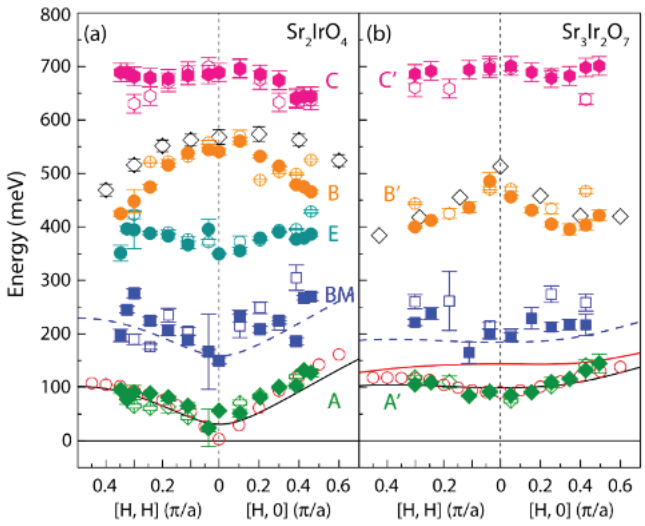

Figure 5. Dispersions of the collective modes in (a) Sr_2IrO_4 and (b) $Sr_3Ir_2O_7$ probed by O K-edge RIXS. Green diamonds, blue squares, cyan circles, orange circles and pink hexagons denote respectively single magnons (A, A'), bimagnons (BM), electron-hole excitons (E), spin-orbit excitons creating magnons (B, B') and spin-orbit excitons (C, C').

Phase diagram and magnetic excitations of $(Sr_{1-x}La_x)_3Ir_2O_7$.

A MIT can be achieved by replacing Sr with La in $(Sr_{1-x}La_x)_3Ir_2O_7$ (electron doping). The robust metallic regime displays exotic electronic properties and may be a platform for realising superconductivity, as well as for understanding the evolution of the ground state and magnetic dynamics. We used resonant elastic and inelastic x-ray scattering (REXS and RIXS) at the Ir L_3 edge to study the magnetic order and excitations of $(Sr_{1-x}La_x)_3Ir_2O_7$ upon electron-doping [52]. The REXS measurements reveal an evolution of the magnetism from three-dimensional (3D) c-axis G-type antiferromagnetic order (AFM) to short range order (SAF) across the MIT, and subsequent 2D SAF deep in the metallic regime, elucidating a rich magnetic phase diagram **(Figure 6a)**. The RIXS results show that the magnons undergo an anisotropic damping with increasing doping, with the large magnon gap being strongly suppressed in the 2D SAF metal **(Figure 6b,c)**. This indicates that the emerging itinerant electrons suppress the AFM by weakening the magnetic couplings and drive the system into a 2D SAF correlated metallic state hosting strong antiferromagnetic fluctuations of the J_{eff} = 1/2 moments. Our results provide a solid experimental basis that will guide future theoretical work on the physics of doping the spin-orbit-induced Mott insulators in the presence of strong interlayer interactions.

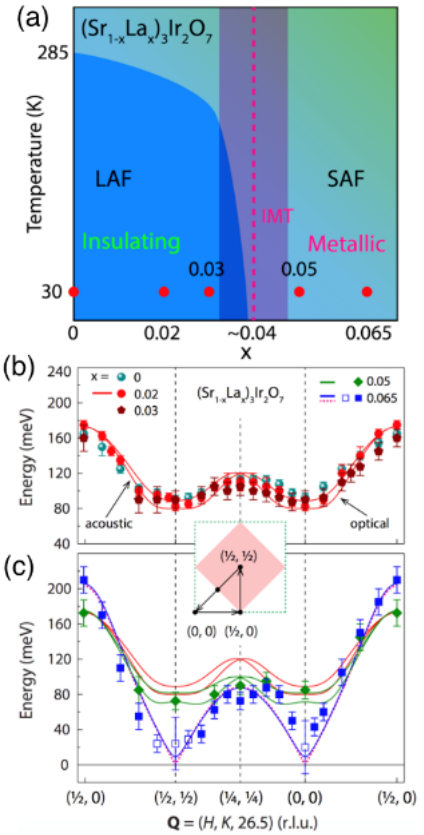

Figure 6. (a) Schematic electronic and magnetic phase diagram of $(Sr_{1-x}La_x)_3Ir_2O_7$. (b,c) Doping-dependence of magnon dispersions.

SCIENTIFIC HIGHLIGHTS

RIXS and XAS on single-layer ruthenates. Motivated by the outstanding questions related to the nature and mechanism of the Mott insulating and superconducting ground states found in Ca_2RuO_4 and Sr_2RuO_4, we studied the orbital physics of these compounds [19]. High-resolution RIXS at the oxygen *K*- edge **(Figure 7)** gave us indirect access to the orbital configurations, and in combination with linear dichroism XAS, **(Figure 8)**, the Ru 4*d*-orbital occupation and excitations were probed through their hybridisation with the O *p*-orbitals. These results were described within a minimal model including the crystal-field splitting and a spin-orbit coupling strength of λ_{so} = 200 meV. Thus, we have demonstrated that the spin-orbit interaction is a significant energy scale in this class of ruthenate materials

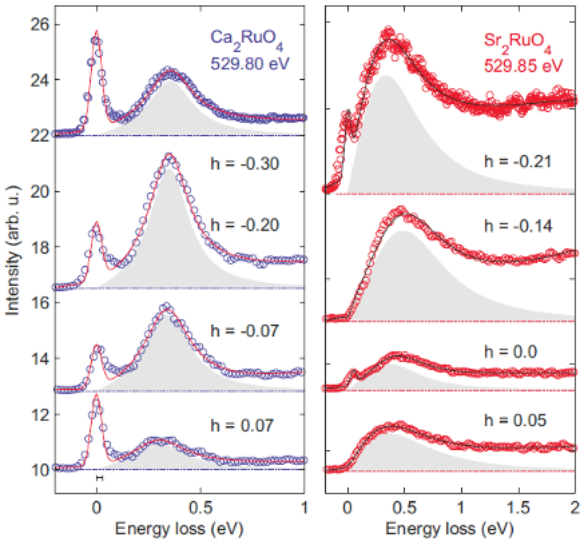

Figure 7. RIXS spectra for different momentum transfers, Q = (h,0) as indicated, on Ca_2RuO_4 (left) and Sr_2RuO_4 (right), recorded using linear-horizontal light tuned to the in-plane oxygen *K*-edge. All spectra have an arbitrary y-axis shift. Solid lines are fits to a Gaussian (elastic line), an anti-symmetric Lorentzian (grey shading) and a quadratic background.

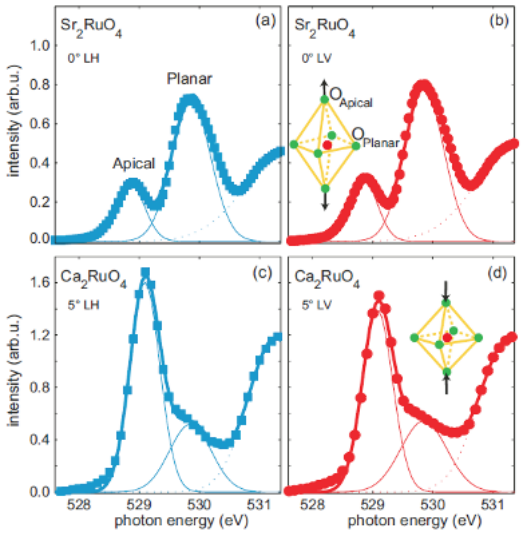

Figure 8. XAS spectra of Sr_2RuO_4 (top) and Ca_2RuO_4 (bottom), recorded using vertically (left) and horizontally (right) polarised light at near-normal incidence. The solid lines are Gaussian fits and a sloping background has been subtracted. Insets show the elongated and compressed RuO_6 octahedron.

ARPES on the Mott insulator Ca_2RuO_4. A paradigmatic case of multi-band Mott physics including spin-orbit and Hund coupling is realised in Ca_2RuO_4. Progress to understand the nature of this Mott-insulating phase has been impeded by the lack of knowledge about the low-energy electronic structure. We performed an ARPES study [60] of the insulating state (at 150 K), finding three different bands, whose orbital character we discussed by comparison with first-principles DFT calculations **(Figure 9)**. The observed bandstructure is not compatible with a single insulating energy scale for all orbitals. A phenomenological Green function incorporating an enhanced crystal field and a spectral gap in the self-energy **(Figure 9c)** can describe the observed band structure on a qualitative level. Further insight is gained from DMFT calculations (**Figure 9d)**: the insulating behavior of the d_{xz} and d_{yz} bands is caused primarily by the Coulomb interaction, while the Hund coupling splits the d_{xy} band, allowing a quantitative estimate of this parameter. These results shed new light on the Mott phase in Ca_2RuO_4 and offer an explanation as to why previous experiments have identified different values for the energy gap.

SCIENTIFIC HIGHLIGHTS

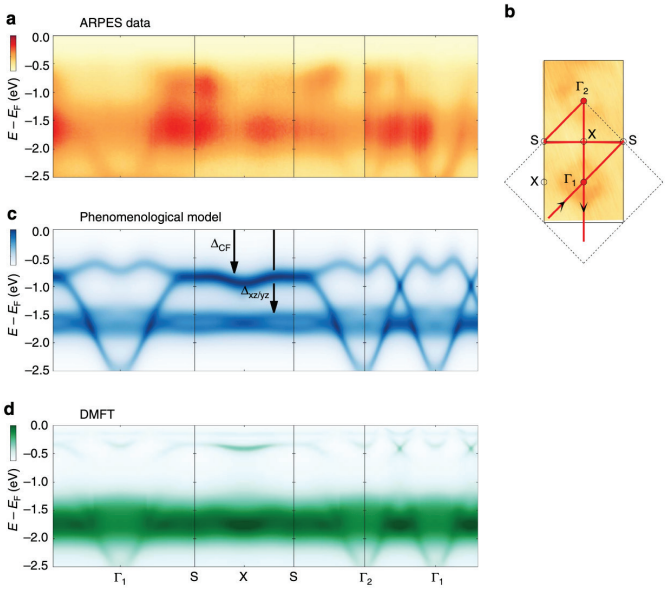

Figure 9. Bandstructure of Ca_2RuO_4 along high-symmetry directions. *(a)* ARPES spectra with 65 eV circularly polarised light. *(b)* Constant-energy map at $E - E_F = -2.7$ eV. *(c)* DFT-derived spectra upon inclusion of a Mott gap, $\Delta = 1.55$ eV, acting between the d_{xz} and d_{yz} bands, and of an enhanced crystal field, $\Delta_{CF} = 0.6$ eV, that shifts the d_{xy} bands. *(d)* DMFT calculation of the spectral function, with Coulomb interaction $U = 2.3$ eV and Hund coupling $J_H = 0.4$ eV.

Ca_2RuO_4 proven to be a band-Mott insulator. We performed a high-resolution oxygen *K*-edge RIXS study of the antiferromagnetic Mott-insulating state of Ca_2RuO_4, finding both low- (80 and 400 meV) and high-energy (1.3 and 2.2 eV) excitations, shown in **Figure 10**, which depend strongly on the incident light polarisation. Guided by theory, we interpreted the low-energy branch as composite spin-orbital excitations, whose presence unveils the intricate interplay of crystal-field splitting and SOC, while the high-energy excitations are intra-atomic singlet-triplet transitions at an energy scale set by the Hund coupling. Thus our results strongly support a spin-orbit coupled band-Mott insulating phase, characterised by orbitally selective coexistence of a band and a Mott gap [76].

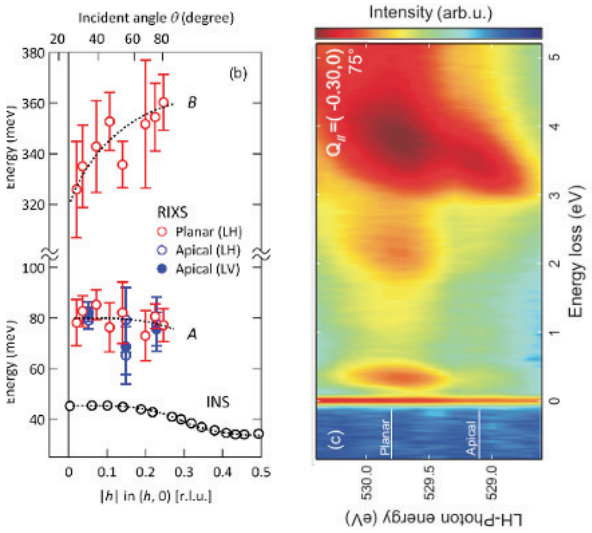

Figure 10. RIXS data and analysis revealing the t_{2g} orbital structure of Ca_2RuO_4.

C_2-symmetric Dirac semimetal uncovered in $Ca_3Ru_2O_7$. Two-dimensional semimetals have been a centre of attention since the realisation of graphene, with the design of Dirac and Weyl semimetals a particular focus. We used ARPES to show how a rotationally symmetry-broken massive Dirac semimetal is realised in $Ca_3Ru_2O_7$ **(Figure 11)** [89]. Typically, Dirac metals are a consequence of the crystal-field environment and can be captured by DFT, but the semimetal state in $Ca_3Ru_2O_7$ emerges from a two-stage electronic transition driven by electron correlations that lie beyond DFT. Its Dirac point and band velocity are consistent with constraints set by quantum oscillation, thermodynamic and transport experiments.

SCIENTIFIC HIGHLIGHTS

While these results advance our understanding of the peculiar fermiology of $Ca_3Ru_2O_7$, the two-stage transition preserves the Brillouin zone, meaning that broken translational symmetries are excluded, and hence the mechanism and symmetry-breaking elements underlying the electronic reconstruction remain to be identified. Our study therefore identifies $Ca_3Ru_2O_7$ as another prominent "hidden-order" problem, in the same category as URu_2Si_2, which also undergoes an electronic transition with no identifiable symmetry-breaking.

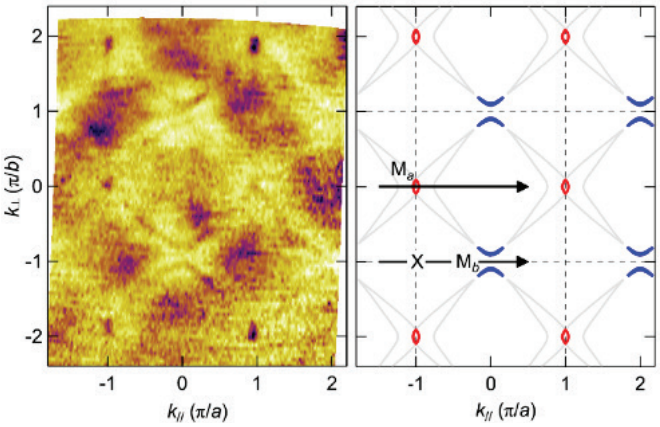

Figure 11. Low-temperature Fermi surface of $Ca_3Ru_2O_7$, displaying a new C_2-symmetric electron pocket.

Electromagnon dispersion in $LiCrO_2$. Inelastic X-ray scattering (IXS) is an ideal tool for measuring collective excitations in solids and liquids with meV energy resolution. Although the cross-section for non-resonant scattering is dominated by phonons, it is possible to probe additional degrees of freedom, such as magnetic fluctuations, that couple strongly to them. We have used the IXS spectrum to observe strong magnon-phonon coupling in $LiCrO_2$, and thus to measure the magnetic correlations throughout the Brillouin zone [50]. We find electromagnons and electric dipole-active two-magnon excitations in the magnetically ordered phase **(Figure 12)** and heavily damped electromagnons in the paramagnetic phase. We anticipate that several frustrated magnets with dominant direct exchange and non-collinear magnetism will show large IXS cross-sections for both magnons and multi-magnon processes.

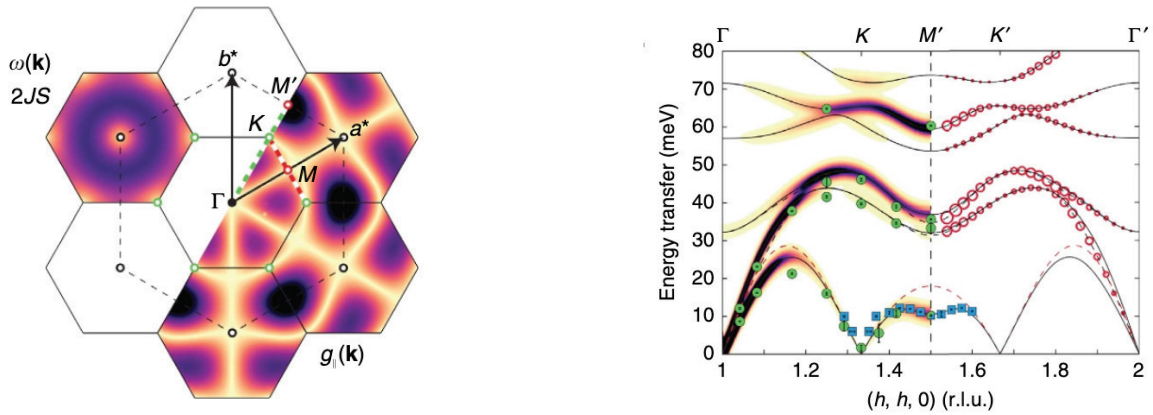

Figure 12. (left) Paths in the Brillouin zone of complementary IXS (red) and INS (green) measurements on $LiCrO_2$. (right) Excitations measured by IXS (green symbols) and INS (blue); the lowest mode is the electromagnon.

SCIENTIFIC HIGHLIGHTS

Entangled Excitations Near Spin-Orbital Quantum Criticality. The spinel $FeSc_2S_4$ is thought to realise a near-critical spin-orbital singlet (SOS) state, where entangled spin and orbital moments fluctuate in a global singlet state on the verge of spin and orbital order. We have performed powder inelastic neutron scattering measurements in both zero field and applied magnetic fields up to 8.5 T to observe the full bandwidth of magnetic excitations [65], as shown in **Figure 13**. We deduce that the key features of the spectrum are fully consistent with spin-orbital triplon excitations of an SOS state: the upward shift of low-energy spectral weight on increasing the field is explained naturally by the entangled spin-orbital character of the magnetic states, and stands in direct contrast to the behaviour of a spin-only singlet ground state, where the spin gap decreases with increasing field.

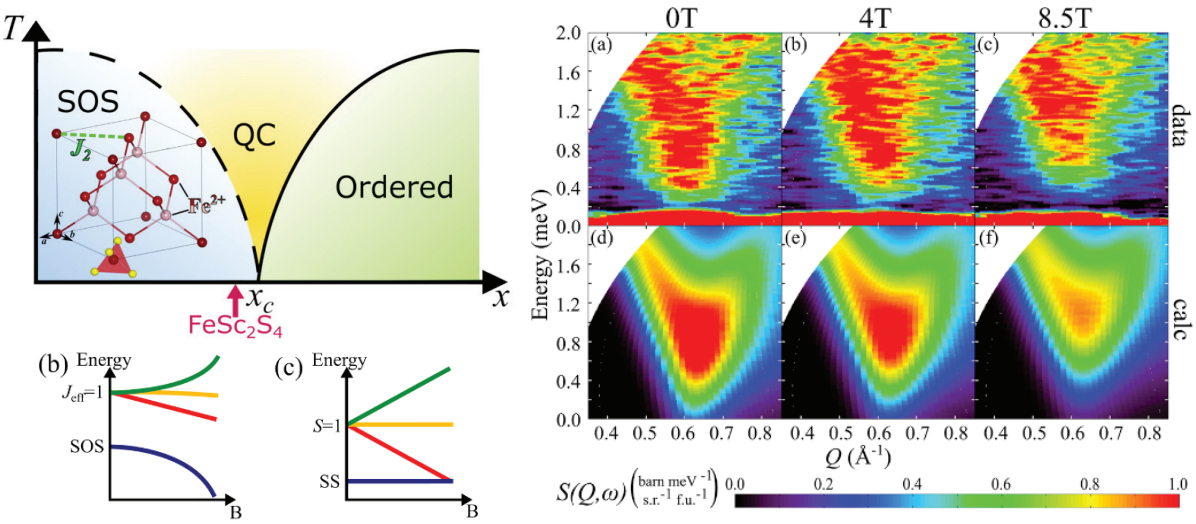

Figure 13. (upper left) Phase diagram of the spin-orbital system as a function of $x = J_2/\lambda$, showing the spin-orbital singlet (SOS), quantum critical (QC) and spin plus orbitally ordered regimes. (lower left) Representation of the evolution of ground- and excited-state energies for SOS (b) and spin-only singlet (c) states. (right) Powder INS spectra of $FeSc_2S_4$ at three different applied magnetic fields.

SCIENTIFIC HIGHLIGHTS

THEORY

Quantum dimer excitations in $Sr_3Ir_2O_7$. Building on the observation that the magnetic excitation spectrum of the bilayer iridate perovskite $Sr_3Ir_2O_7$ is more typical of weakly coupled dimers than of weakly coupled planes, we have performed a detailed investigation of its properties both theoretically, using a bond-operator method, and experimentally, by high-resolution RIXS. Using an experimental geometry with the x-ray wavevector transfer lying predominantly in the iridium oxide bilayer allowed us to reveal the existence of two magnon branches, shown in **Figure 14**, which are clearly separated from one another in energy and are gapped across the entire Brillouin zone. These findings agree with the general predictions of our bond-operator theory, allowing the interaction parameters to be fitted with quantitative accuracy and providing the insight that the lower and upper magnon branches are respectively transverse and longitudinal modes [24].

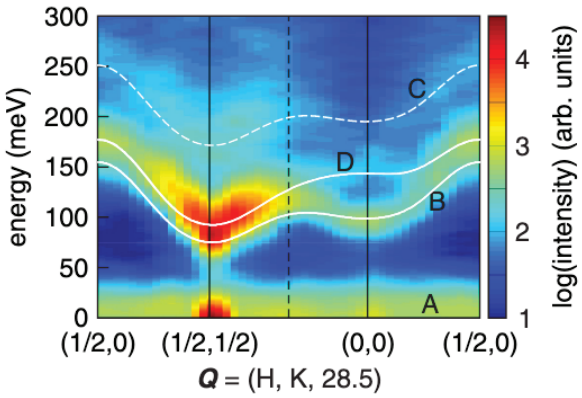

Figure 14. In-plane wavevector-dependence of the RIXS response of $Sr_3Ir_2O_7$. White lines represent the results of model calculations, which in particular discovered and explained Mode D.

Excitation modes of the bilayer Heisenberg model. Motivated by the presence of a single transverse spin excitation in $Sr_3Ir_2O_7$, whereas a bilayer of weakly coupled Heisenberg planes is known to have two spin-wave modes, we have investigated the dynamical spin structure factor of the spin-1/2 Heisenberg model on the square-lattice bilayer using quantum Monte Carlo simulations augmented by higher-order spin-wave theory, bond-operator methods and strong-coupling expansions [31]. We have shown that, on approaching the quantum phase transition at which the spin gap opens, the spin-wave mode antisymmetric with respect to layer inversion evolves smoothly into the gapped triplon mode of the strong-coupling limit, while the symmetric spin-wave mode loses almost all of its intensity. We also find that the amplitude (Higgs) mode is essentially invisible in the dynamical spin structure factor, but that it appears as a distinct feature in the singlet dynamical structure factor that could be probed by light-scattering methods.

IMPACT
PHDS AND POSTDOCS

PhD graduated

Lin Yang, 2016
Sara Fatale, 2017
Claudia Fatuzzo, 2017
Virgile Favre, 2017
Noore E. Shaik, 2019

PhD partially suppported by MPBH

Vera Schnells, 2013
Lakshmi Das, 2017

PhD in progress

Yi Tseng, 2020
Edoardo Martino, 2020
Gianmarco Gatti, 2020

Postdocs and current positions

Paul Olalde Velasco, 2013-2014, Postdoctoral Fellow, Diamond Light Source, UK
Bastien Dalla Piazza, 2014- 2015, Technical Lead, Pix4D, CH
Xing-Ye Lu, 2015-2016, Assistant Professor, Beijing Normal University, CN
Björn Wehinger, 2015-2016, Scientist, Elettra Synchrotrone, IT
Andrew J. Smerald, 2013-2017, Scientist, Max Planck Institute, DE
Sergiy Katrych, 2014, Chemist, Excelsus Structural Solutions, CH
Alun M. Biffin, 2015- 2017, Data Scientist, Van Lanschot Kempen, NL
Katharina Rolfs, 2013-2017, Research Scientist, Paul Scherrer Institute, CH
Diane Lançon, 2017-2018, Postdoctoral Fellow, ETH Zurich, CH
Marton Kollar, 2017-2018, Postdoctoral Fellow, EPF Lausanne, CH
Danu Bimla, 2018, Postdoctoral Fellow, Julius-Maximilians-Universität, DE
Saeed S. Jahromi, 2018, Postdoctoral Fellow, Donostia International Physics Center, ES
Eugenio Paris, 2017-2019, Postdoctoral Fellow, Paul Scherrer Institute, CH
Masafumi Horio, 2017-2019, Postdoctoral Fellow, University of Zurich, CH
Trpimir Ivsic, 2018-2019, Scientist, EPF Lausanne, CH

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OUTLOOK

The **Mott Physics Beyond the Heisenberg Model** network investigated all aspects of the rich physics in materials with strong correlations and strong spin-orbit-coupling, driving a deeper understanding of many profound phenomena. True to its SINERGIA title, research progress in the field has a deep and synergistic dependence on new technology, and MPBH teams developed new methods in time-resolved ARPES, high-resolution RIXS, high-pressure crystal growth and sample preparation by focused ion-beam milling, as well as providing a steady source of new materials. The timing of the MPBH initiative was designed specifically to seize the opportunity of profiting from these advances, and MPBH research and technology has provided essential input for long-term Swiss infrastructure investments, particularly the SLS 2.0 and SINQ upgrade projects at PSI. This also allowed MPBH researchers, in collaboration with other groups around the world, to achieve significant advances in the field of spin-orbit-coupled Mott systems. Even so, by elements and materials investigated the field remains young, leaving vast potential for both fundamental and applied breakthroughs. **The network established, the capabilities developed and the large-scale facilities constructed will position Swiss research at the forefront in cementing scientific and technological gains for the next decade.**

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Many of our achievements also relied critically on large-scale research facilities, particularly the national facilities installed at PSI and in the Federal Institutes of Technology, and we are grateful for the foresight and long-term funding which created these.

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