A selection of recent RIXS articles in the Journal of Synchrotron Radiation

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IRIXS Spectrograph: an ultra high-resolution spectrometer for tender RIXS
J. Bertinshaw *et al.*


Development of the Soft X-ray AGM-AGS RIXS beamline at the Taiwan Photon Source
A. Singh *et al.*

*J. Synchrotron Rad.* (2020). 27, 1235–1239

$hv^2$-concept breaks the photon-count limit of RIXS instrumentation
K. J. Zhou, S. Matsuyama and V. N. Strocov

*J. Synchrotron Rad.* (2020). 27, 979–987

Saturation and self-absorption effects in the angle-dependent 2p3d RIXS spectra of Co$^{3+}$
R.-P. Wang *et al.*


A design of RIXS spectrometer for spatial- and time-resolved spectroscopy
Y.-D. Chuang *et al.*

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**I21 Resonant Inelastic X-ray Scattering**

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Energy: 0.20 - 3.0 keV
Talks - Abstracts
Intermediate-energy resonant inelastic x-ray scattering (IRIXS) with photon energies in the “tender” x-ray regime (2.5-5 keV) has recently been established at beamline P01 at the PETRA-III synchrotron [1,2]. The energy resolution of the IRIXS instrument is currently ~30 meV at the Ru L-edge, comparable to the most advanced soft x-ray RIXS instruments. We will highlight recent experiments on collective spin excitations as well as spin-state transitions and spin-orbit excitons in a variety of ruthenium compounds [3-7], and outline perspectives for future IRIXS studies of 4d-electron compounds and heterostructures.

The work discussed in this talk is based on collaboration with many scientists, who are listed in the publications below. Financial support from the European Research Council Advanced Grant 669550 (Com4Com) is gratefully acknowledged.

References
Exotic magnetism in honeycomb ruthenium compounds: 
Critical role of spin-orbit coupling

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Ruthenium compounds have been the central focus of materials research thanks to their rich phenomena and functionalities, including exotic superconductivity and efficient catalytic activity. The orbital degrees of freedom in the partially-filled \(t_{2g}\) orbitals and the Hund’s rule interactions between the electrons play a key role in realizing these properties. However, little attention has been paid to the role of spin-orbit coupling (SOC) in the 4\(d\) transition metal compounds. In this talk, I will address exotic magnetism in Ru-based honeycomb Mott insulators where SOC plays a crucial role, and how RIXS can be utilized to understand them. We used Ru \(L_3\)-edge RIXS enabled by the new intermediate-energy resonant inelastic x-ray scattering (IRIXS) spectrometer [1].

I will first introduce the magnetic excitation spectra of a microcrystal of SrRu\(_2\)O\(_6\) [2], which exhibits an antiferromagnetic order at an exceptionally high transition temperature of 563 K. We observed of antiferromagnetic magnon dispersion, based on which we determined the spin Hamiltonian. Our observation of single magnon by the tender x-ray RIXS opens broad prospects for the magnetism research in 4\(d\) transition metal compounds.

Next I introduce our study of a Kitaev spin liquid candidate \(\alpha\)-RuCl\(_3\) [3]. Due to the strong frustrations between the bond-dependent pseudospin interactions, the excitation spectrum is expected to contain a continuum. Based on the momentum dependence of RIXS intensity in conjunction with theoretical modelling, we have determined the pseudospin Hamiltonian that precisely reproduces the zigzag magnetic order at low temperature and its quick disappearance above \(T_N\).

References
The presence of Mott insulating states in single-layer $\text{Sr}_2\text{IrO}_4$ and double-layer $\text{Sr}_3\text{Ir}_2\text{O}_7$ with relatively delocalized Ir 5d orbitals with much larger bandwidth ($W$) than on-site Coulomb interaction ($U$) (i.e. $W \gg U$) was unexpected. The insulating phase in these iridates can neither be described by usual band theory nor by solely considering the $U/W$ ratio. The missing player for reconciling this unusual behavior is the spin-orbit (SO) interaction, which in 5d transition metal oxides is at least one order of magnitude larger than in 3d transition metal oxides. In layered iridates, $\text{Ir}^{4+}$ accommodates five electrons in the $t_{2g}$ levels, which establish by cooperation of SO, U and the crystal field interactions an exotic Mott insulating ground state composed by an electron in a $J_{\text{eff}}=1/2$ orbital. Resonant inelastic x-ray scattering (RIXS) is a unique spectroscopic tool for measuring the full spectrum of low-energy elementary excitations with bulk and element-sensitivity. For iridates, RIXS at the Ir L- [1] and O K-edges [2] give detailed and complementary insight in magnons, spin-orbit excitons and charge-transfer excitations.

The nature of the highly spin-orbit coupled Mott state of $\text{Sr}_2\text{IrO}_4$ suggests the ground state as well as the collective entangled spin and orbital excitations to be strongly dependent on the lattice degree of freedom. For this reason, $\text{Sr}_2\text{IrO}_4$ provides an ideal platform for controlling the physical properties of a correlated material by inducing local lattice distortions. We use epitaxial strain to modify the Ir-O bond geometry and perform momentum-dependent RIXS, both at Ir L- and O K-edges, to unveil the response of the low energy elementary excitations [3]. By applying tensile strain, we observe a large softening of the spin(-orbital) wave dispersion along the [h,0] direction and a simultaneous hardening along the [h,h] direction. This evolution entails a strain-driven crossover from anisotropic to isotropic interactions between the magnetic moments. We also show how the charge excitations are coupled to the lattice in $\text{Sr}_2\text{IrO}_4$. To this end, using O K-edge RIXS, we unveil the evolution of a dispersive electron-hole pair excitonic mode which shifts to lower (higher) energies upon compressive (tensile) strain, manifesting a reduction (increase) in the size of the charge gap. We show that this behavior originates in the modified hopping elements between the $t_{2g}$ orbitals induced by strain. Our work highlights the central role played by the lattice in determining both the spin-orbital interactions as well as the charge gap of $\text{Sr}_2\text{IrO}_4$ and confirms epitaxial strain as a promising route towards the control of the ground state of complex oxides in the presence of high spin-orbit coupling [3].

References
Bad Metal Phase in Charge Frustrated Pyrochlore Cd$_2$Re$_2$O$_7$

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Cd$_2$Re$_2$O$_7$ is pyrochlore bad metal that shows a near temperature independent resistivity above $T_{\text{HO}}=205$ K [1-3]. Below $T_{\text{HO}}$, Cd$_2$Re$_2$O$_7$ experiences a “hidden order” phase transition accompanied with a lattice inversion symmetry-breaking. Here we combine optical conductivity, Re L-edge resonant inelastic x-ray scattering and DFT+DMFT calculations to show that the bad metal phase in Cd$_2$Re$_2$O$_7$ is induced by the charge frustration of the pyrochlore lattice, where the destructive quantum interference yields localized electronic wave function and flat band near the Fermi level.

References
Quantum fluctuations of charge order induce phonon softening in a superconducting cuprate


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Quantum phase transitions play an important role in shaping the phase diagram of high-temperature cuprate superconductors. These cuprates possess intertwined orders which interact strongly with superconductivity. However, the evidence for the quantum critical point associated with the charge order in the superconducting phase remains elusive. Here we show the short-range charge orders and the spectral signature of the quantum fluctuations in La$_{2-x}$Sr$_x$CuO$_4$ near the optimal doping using high-resolution resonant inelastic X-ray scattering. On performing calculations through a diagrammatic framework, we discovered that the charge correlations significantly soften several branches of phonons. These results elucidate the charge order in cuprates, providing evidence for the existence of quantum critical point and discommensurations associated with charge order.
The electronic structure of layered high $T_c$ cuprate superconductors may be understood in terms of a hybridization between the Cu $3d_{x^2-y^2}$ and O $2p_z$ orbitals, and a strong on-site Coulomb repulsion between electrons on the Cu sites [1–4]. When holes are introduced, they reside preferentially in the so-called “charge-transfer band” which is composed primarily of O orbitals [5]. In contrast, doped electrons enter the upper Hubbard band and primarily reside on the Cu orbitals [4]. Despite this asymmetry in the electronic structure, charge order, a complex phase of periodically modulated charge-carrier density, is ubiquitous in both the electron- and hole-doped cuprates [6]. Surprisingly, a more widely observed mode of collective charge-density oscillation, the acoustic plasmon, has been rather elusive for the cuprates. In contrast to ordinary metals, where long-range Coulomb interactions give rise to isotropic long-wavelength optical-like gapped plasmons, out-of-phase oscillations of charges in neighboring planes of layered electron gases, form acoustic plasmons, whose energy tends to zero for small in-plane wave vectors [7]. Confinement of the doped charges to CuO$_x$ planes and poor screening of out-of-plane Coulomb interactions by intervening dielectric blocks, suggest that acoustic plasmons should also be present in the cuprates [8–10]. While acoustic plasmons were recently observed in electron-doped cuprates using Cu $L_3$ resonant inelastic x-ray scattering (RIXS) [11,12], the situation in hole-doped cuprates, has remained inconclusive. In this work, we show that acoustic plasmons are indeed present in hole-doped cuprates from O K-edge RIXS study of La$_{1.84}$Sr$_{0.16}$CuO$_4$ and Bi$_2$Sr$_1$La$_{4-δ}$CuO$_{6+δ}$. The excitations have a strong out-of-plane dispersion as expected for plasmons in layered systems. The discovery of acoustic plasmons in the hole-doped systems remarkably illustrates the universal existence of low-energy collective excitations besides phonons and spin fluctuations across the cuprate phase diagram. The observed acoustic plasmons are predominantly associated with the O orbitals in these systems. Our results will therefore stimulate more studies of doped-hole charge dynamics, taking into account the three band model in the cuprates [2,3].

References
Doping and temperature evolution of charge density fluctuations in two families of high-$T_c$ superconductors

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Charge order, i.e. the incommensurate modulation of electron density in the CuO$_2$ planes, might be the key for explaining the mysterious properties of high-$T_c$ superconductors (HTS). Charge density waves (CDW) have been indeed found in all the cuprate families, and their interplay with superconductivity is proved by many experimental evidences [1]. However, this phenomenology is present in underdoped samples and below the pseudogap temperature only, casting doubts about their actual role in shaping the transport properties of cuprates either in the normal or superconducting states. By taking advantage of the superior sensitivity of modern Resonant Inelastic X-ray Scattering (RIXS), we have discovered another form of charge modulation, we named charge density fluctuations (CDF), in the Y123 family [2]. They are very short-ranged -can be viewed as precursors of CDW- and pervade a large area of the phase diagram, being present in a broad doping range and persisting at temperatures exceeding not only the previously defined onset of CDW, but also the pseudogap temperature $T^*$, and surviving up to room temperature and beyond. Several recent RIXS studies have shown that this occurrence is universal for cuprates, since it has been observed in all the families of investigated compounds, including those with the low temperature locking of spin and charge modulations [3]. In view of these experimental results, the temperature-doping phase diagram gets profoundly reshaped: in order to explain the cuprate phenomenology, one cannot ignore the presence of these pervasive high temperature charge fluctuations. Notably, several experiments have shown that these modulations are dynamical, i.e. they are characterized by finite energies in the meV range. Because of their properties, several theoretical proposals indicate that they are likely the long-sought excitations underlying either the anomalous strange metal behavior [4] or the superconducting pairing mechanism [5] of HTS.

To get a better insight about the nature of CDF and their implication in the physics of cuprates, we have recently performed a thorough RIXS investigation on several YBCO and Bi2212 samples in a very broad doping range, including doping levels at the extreme sides of the superconducting dome, where CDF can be studied alone, i.e. without any contribution of CDW. This occurrence gave us to possibility to explore with unprecedented accuracy the doping and temperature evolution of the CDF correlation length, intensity and energy. Our results pave the way for a better understanding of the role charge order has in shaping the ground state of HTS.

References
Charge-density-waves (CDWs) in cuprate high-temperature superconductors have a long history, and a remarkably varied phenomenology across different materials’ families and as a function of carrier doping and magnetic field. The current understanding of CDWs hinges on a series of commonly accepted facts: (i) CDWs develop at a well-defined, doping- and family-dependent ordering vector $Q_{\text{CDW}}$; (ii) density modulations arise from Cu-3$d$ + O-2$p$ orbitals in the CuO$_2$ planes; and (iii) CDWs cohabit with the superconducting and the pseudogap state but have never been found in the presence of static magnetic order.

In this talk, I will present recent results that challenge this established phenomenology. We studied CDWs in the family of REBa$_2$Cu$_3$O$_{7-\delta}$ superconductors where hole doping can be achieved via control of oxygen stoichiometry or by rare earth substitution, as investigated here in the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ series. In this system, we used resonant soft X-ray scattering at both the Cu-L$_3$ ($2p \rightarrow 3d$) and Pr-M$_5$ ($3d \rightarrow 4f$) resonances to observe CDWs exhibiting multiple ordering channels at once and develop out of a magnetic insulating ground state which should not in principle be compatible with density modulations from mobile carriers.

In the metallic regime with low Pr content ($x = 0.275$ and 0.5), we found a conventional CDW with single ordering vector (CDW-1Q) and resonating exclusively at the Cu-L$_3$ edge. In the magnetic insulator end compound PrBa$_2$Cu$_3$O$_{7-\delta}$, however, we discover a very different and unprecedented form of CDW with two independent spatial modulations and ordering vectors $Q_1 = 0.36$ and $Q_2 = 0.28$ r.l.u. (CDW-2Q). Unlike previous reports in all other cuprates, this unique twofold or double-Q charge order resonates at both the Cu the Pr edges, indicating an active and unprecedented role of 4$f$ states in the formation of CDWs.

These findings, and in particular the emergence of CDWs out of a magnetic insulator, reveal new important details about the nature of the CDW state and its interplay with neighbor electronic phases. They challenge several known facts about the physics of the CuO$_2$ planes and offer new keys to understand the origin of CDWs in cuprates.
Magnetic reconstructions of Sr$_2$IrO$_4$ thin films via interfacial interactions

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Interfacial interactions are of major importance in manipulating materials properties and obtaining better understandings of microscopic mechanisms. Resonant inelastic x-ray scattering (RIXS) has been proven to be a powerful technique to probe magnetic excitations in Ir oxides.$^1$ In particular, magnetism of heterostructure Ir oxide thin films have been successfully explored.$^{2-4}$ In this talk, I present RIXS measurements on Sr$_2$IrO$_4$ thin films under two types of interfacial interactions: uniaxial strain interface giving rise to 0.8% to 2.2% orthorhombicity and heterogeneous electronic interface between Mott insulator and metal. Large uniaxial strain interface is realized by a-axis oriented Sr$_2$IrO$_4$ thin film on LaSrGaO$_4$ and Ca$_3$Ru$_2$O$_7$ and insulator-metal interface is realized by using Sr$_2$RuO$_4$ and Ca$_3$Ru$_2$O$_7$ substrates. (π,0) magnon is mostly insensitive to two types of interfacial interactions. Interestingly, (π/2, π/2) magnon shows a lager response to the insulator-metal interfacial interactions. I discuss the implications of such observations.

References
Nearly itinerant electronic ground state in the intercalated honeycomb iridate \( \text{Ag}_3\text{LiIr}_2\text{O}_6 \)

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The introduction of atomic species between honeycomb planes in insulating Iridium oxide Kitaev magnets is emerging as a powerful approach to engineer magnetic interactions with the end goal of suppressing long-range order. Here, we use x-ray spectroscopy at Ir L\(_3\)/L\(_2\) absorption edges to study powder samples of the intercalated honeycomb magnet \( \text{Ag}_3\text{LiIr}_2\text{O}_6 \) [1]. We argue that the topochemical exchange of interlayer Li atoms with Ag in \( \text{Ag}_3\text{LiIr}_2\text{O}_6 \) [2,3] atoms result in a nearly itinerant electronic structure with enhanced Ir-O hybridization that fundamentally alters the magnetism. X-ray absorption reveals that the magnetism in \( \text{Ag}_3\text{LiIr}_2\text{O}_6 \) is characterized by an asymmetric spin density with strong spin-orbit coupling and a larger orbital component than the parent compound \( \alpha\)-Li\(_2\)IrO\(_3\). Resonant inelastic x-ray scattering spectra probing the Ir electronic structure of \( \text{Ag}_3\text{LiIr}_2\text{O}_6 \) is captured by incorporating Ir-Ir hopping integrals, demonstrating that the local \( j_{\text{eff}} = 1/2 \) picture is not a valid basis (See Figure). \( \text{Ag}_3\text{LiIr}_2\text{O}_6 \) must be understood as a new type of nearly itinerant model quantum magnet. We posit that similar effects may be at play in other intercalated honeycomb iridates. Our results provide an empirical foundation to develop suitable effective Hamiltonians in these next-generation frustrated and confirm the importance of metal - ligand hybridization in the magnetism of transition metal oxides.

References
Ferromagnetic Kitaev interaction and the origin of large magnetic anisotropy in \( \alpha \)-RuCl\(_3\)

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\( \alpha \)-RuCl\(_3\) is a two-dimensional, honeycomb lattice material that has emerged as a candidate system in which to find Kitaev spin liquid physics. However, despite recent intensive study, the magnetic Hamiltonian of this material is not fully understood. Experimental datasets have been fit using a variety of different magnetic Hamiltonians, which vary not only in the magnitude but even in the signs of the interaction terms. We present resonant elastic x-ray scattering measurements that determine the magnetic moment direction in the ordered phase, information that constrains the two leading anisotropic terms in the magnetic Hamiltonian. Our findings indicate that in \( \alpha \)-RuCl\(_3\) the Kitaev (K) magnetic interaction is negative, while the off-diagonal interaction known as the Gamma term is positive. Further, the observed out-of-plane tilting angle of the magnetic moments indicates the presence of a substantial Gamma term, on the order of the size of the K interaction. We note that this parameter regime naturally explains the anisotropy in magnetization that has long been known to exist in this material. Our findings therefore place unambiguous experimental constraints on the magnetic interactions in \( \alpha \)-RuCl\(_3\), and can lend insight into the behavior of this remarkable magnetic material.
Spin order formation and dynamics without a global inversion symmetry

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Logic and memory devices created from novel spin-orbit materials are scalable and energy-efficient alternate to the current CMOS devices. For example, Skyrmions, vortex-like topological spin textures, found in cubic FeGe is a well-known candidate for a racetrack memory device. In this talk, I will show that such topological spin textures can also form in pseudo-amorphous Fe\(_x\)Ge\(_{1-x}\) films (a-FeGe) due to a short-range Dzyaloshinsky–Moriya interaction (DMI). Our resonant soft x-ray scattering studies revealed two distinct magnetic transitions at \(T_{M1} \sim 180\) K and \(T_{M2} \sim 100\) K present in this system whose orderings are strongly connected to the interplay of spin mobility and the local DMI interaction. We found a helical spin order in the temperatures between \(T_{M1}\) and \(T_{M2}\), and isolated Skyrmions close to \(T_{M2}\). That is, amorphous thin films without a global inversion symmetry can harvest spin textures similar to which form in crystals. We further used coherent x-ray speckle analysis and showed the existence of spontaneous spin texture fluctuation in a wide range of temperatures. The understanding of isotropic and anisotropic spin textures formation and dynamics in a-FeGe can lead to flexible and low-cost synthesis novel devices.

References
Dynamic electron correlations with charge order wavelength along all directions in the copper oxide plane

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In strongly correlated systems the strength of Coulomb interactions between electrons, relative to their kinetic energy, plays a central role in determining their emergent quantum mechanical phases. We perform resonant x-ray scattering on Bi\textsubscript{2}Sr\textsubscript{2}CaCu\textsubscript{2}O\textsubscript{8+δ}, a proto-typical cuprate superconductor, to probe electronic correlations within the CuO\textsubscript{2} plane. We discover a dynamic quasi-circular pattern in the x-y scattering plane with a radius that matches the wave vector magnitude of the well-known static charge order. Along with doping- and temperature-dependent measurements, our experiments reveal a picture of charge order competing with superconductivity where short-range domains along x and y can dynamically rotate into any other in-plane direction. This quasi-circular spectrum, a hallmark of Brazovskii-type fluctuations, has immediate consequences to our understanding of rotational and translational symmetry breaking in the cuprates. We discuss how the combination of short- and long-range Coulomb interactions results in an effective non-monotonic potential that may determine the quasi-circular pattern. [1]

References
High energy spin excitations in the quantum spin liquid candidate Zn-barlowite probed by inelastic x-ray scattering

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One of the most pressing questions in condensed matter physics is about the nature of the quantum spin liquid (QSL) ground state and its fundamental excitations. This exotic ground state is characterized by long-range quantum entanglement of the spins coinciding with the absence of long-range magnetic order.[1] The most promising lattice that can host a QSL is the kagome lattice of corner-sharing triangles with magnetically frustrated antiferromagnetic $S=1/2$ spins; i.e. Cu$^{2+}$.[2] A novel Cu$^{2+}$ kagome material, Zn-substituted barlowite (Cu$_3$Zn$_x$Cu$_{1-x}$(OH)$_6$FBr), was recently identified as a promising QSL candidate due to its relatively low amount of disorder.[3] However, detailed studies of the low-energy excitations of Zn-barlowite have been impeded by the lack of a successful method of growing single crystals large enough for inelastic neutron scattering (INS). Resonant inelastic X-ray scattering (RIXS) therefore offers a unique opportunity to probe the spin excitations of this kagome QSL candidate in a higher-energy regime than INS. We present the first RIXS measurements on small single crystals of Zn-substituted barlowite. Varying the scattering angle, polarization, and temperature allows us to probe the contributions of phonon and magnetic scattering to the observed RIXS spectra, revealing a broad continuum indicative of scattering from multiple pairs of spinons.

References


Resonant Elastic X-Ray Scattering Study of Skyrmion Lattices

– Microscopic Properties, 3D Structure, and Dynamics –

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Magnetic skyrmions in noncentrosymmetric chiral magnets form ordered lattices with a periodicity ranging from 3-100 nm. This lengthscale lends itself to soft x-ray scattering experiments owing to the large resonant scattering cross-section for 3\textsuperscript{d} elements, the excellent reciprocal space resolution, as well as the tunable surface sensitivity.

We will present an overview of the capabilities of resonant elastic x-ray scattering (REXS) for the study of magnetic skyrmions \cite{1}, highlighting the following effects:

1) Microscopic skyrmion properties \cite{2}: By exploiting the polarization dependence of REXS, the exact surface helicity angles of twisted skyrmions for both left- and right-handed chiral bulk Cu\textsubscript{2}OSeO\textsubscript{3} can be determined.

2) 3D spin structure of skyrmions \cite{3,4}: Using polarization-dependent REXS we found a continuous transformation of the vertical skyrmion tubes in out-of-plane fields from pure Néel-twisting at the surface to pure Bloch-twisting in the bulk over a distance of several hundred nanometers. On the other hand, in in-plane fields, perpendicularly ordered skyrmions form. Their surface state is stable over a wider temperature range compared to the bulk state in out-of-plane fields and it results from the presence of magnetic interactions unique to the surface.

3) Confinement effects in thin membranes \cite{5}: By small-angle REXS in the transmission geometry, a wide range of skyrmion materials can be studied with high reciprocal space resolution. As the membrane thickness has to be thin enough, often coinciding with the characteristic lengthscale of the skyrmion order, confinement and surface effects can be observed in FeGe and Cu\textsubscript{2}OSeO\textsubscript{3}.

4) Skyrmion lattice dynamics \cite{6,7}: The dynamics of skyrmions is determined by their topology. Among the many unusual properties of skyrmions is the tendency of their direction of motion to deviate from that of a driving force; the angle by which they diverge is a materials constant, known as the skyrmion Hall angle. In circular field gradients, skyrmions can be driven to undergo rotation with well-defined dynamics. In a linear field gradient, the shearing of the skyrmion lattice can be used to measure the skyrmion Hall angle.

This research was supported by EPSRC (UK) under grant EP/N032128/1.

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Spin-correlation in correlated quantum materials

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In correlated quantum materials, strong correlation and competing interactions are responsible for a great variety of emergent phenomena, such as, colossal magneto-resistance, high $T_c$ superconductivity, and quantum spin liquid. Among them, spin-correlation is an important jigsaw piece of the fascinating puzzles. Spin-correlations used to be a territory of the inelastic neutron scattering. Over the last decade, resonant inelastic x-ray scattering (RIXS) has become well-known in examining spin-correlation in a large group of quantum materials [1-5]. RIXS holds advantages of polarization control, being element specific, and has no limit of excitation energy. However, it is also accepted that the core-hole effect and the many-body physics can be hurdles of extracting spin-correlations. In this talk, I will briefly present two classical examples, NiO and La$_2$CuO$_4$, through which we learned the strength of RIXS in detecting multiple magnetic excitations, and how one can extract the pure spin-correlation by examining the detailed RIXS form factor [6-7].

References
Ultrafast renormalization of the onsite Coulomb repulsion in a cuprate superconductor


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Intense ultrashort electromagnetic fields are an increasingly important tool to realize and control novel emergent phases in quantum materials. Among a variety of nonthermal excitation pathways, a particularly intriguing route is represented by the direct light-engineering of effective many-body interactions, such as electron hopping amplitudes and electron-electron repulsion. Achieving a light-induced dynamical renormalization of the screened onsite Coulomb repulsion (“Hubbard $U$”) would have far-reaching implications for high-harmonic generation [1], attosecond spectroscopy [2] and ultrafast magnetism [3] in the solid state. However, experimental evidence for a dynamically controlled Hubbard $U$ remains scarce [4,5].

Here, we employ time-resolved x-ray absorption spectroscopy (trXAS) to demonstrate the ultrafast renormalization of the Hubbard $U$ parameter in the prototypical cuprate superconductor La$_{1.905}$Ba$_{0.095}$CuO$_4$ (LBCO, $x$=9.5%) [6]. Our element-specific measurements reveal that intense femtosecond optical pulses (1.55 eV, 50 fs) induce a dramatic shift of the x-ray absorption maxima associated with transitions to the upper Hubbard bands (UHB), while the transition energy into Zhang-Rice singlet states near the Fermi level remains unaffected. Based on exact-diagonalization calculations of the time-dependent spectrum within single- and three-band Hubbard models, we determine a pump-induced suppression of the Hubbard $U$ up to 10% of its equilibrium value on the Cu sites.

Our results represent a first precision measurement of dynamically renormalized Hubbard $U$ in strongly correlated oxides and define a novel strategy for the control of superconductivity, magnetism, as well as to the realization of other long-range-ordered phases in light-driven quantum materials.

References

Coherent X-ray scattering give rise to speckle pattern due to interference of scattered wave fronts that are randomly phase shifted by the morphology of the sample. Speckle pattern is representative of heterogeneity and can be used to measure fluctuations. We will describe use of coherent X-ray scattering to study thermally induced spontaneous fluctuation in a square magnetic nanostructure that exhibits an antiferromagnetic ground state. The magnetic nanostructure consists of elongated thin film segments whose shape anisotropy forces their magnetizations to behave as Ising spins. The arrangement of mutually perpendicular nanomagnets in a square array result in an asymmetry in the inter-island interactions that favors an antiferromagnetic ground state. We performed soft X-ray Photon Correlation Spectroscopy (XPCS) to study thermally induced spontaneous superdomain wall fluctuations as the sample approaches phase transition temperature [1]. We demonstrated the existence of different types of fluctuations and relaxation; a low temperature ballistic and a high temperature diffusive type, as the sample goes through the AF phase transition. We will also discuss generation and control of soft X-ray orbital angular momentum (OAM) beams [2]. We used a square nanomagnetic array with a patterned topological defect such that the defect has a charge of two for the structural lattice and charge of one for the magnetic lattice. Coherent X-ray diffraction from the structure produces photons with even and odd OAM quantum numbers at the structural and AF Bragg peaks, respectively [3]. We show that the OAM content could be controlled by magnetic field. Soft X-ray OAM beams have the potential to unravel new information about quantum properties in materials.

This work is supported by US DOE (Award# DE-SC0016519, DOE BES DE-AC02-05-CH112 (MSMAG)).

References:

Chirality in multilayers probed by Soft x-ray (coherent) scattering

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Nowadays, magnetic chirality has become a topic of utmost importance considering the ever-growing interest in static and dynamic properties of topological magnetic structures such as magnetic skyrmions and domain walls and their possible implications in future high-density data storage devices [1]. One effective way of inducing chiral magnetic structures is to consider magnetic systems showing a dominant Dzyaloshinskii-Moriya interaction (DMI) [2]. To master the stabilization of such magnetic objects, it is crucial to access the magnetic chirality, which still remains a challenge. Recently analyzing the amplitude of magnetic satellites of a crystal Bragg peak, Zhang et al. [3] showed that x-ray resonant magnetic scattering (XRMS) is a powerful tool to access to the relevant topological parameters (chirality, winding number, ...).

First, we will report that small angle XRMS is a straightforward tool to reveal directly the properties of chiral magnetic systems. In our approach using dichroism in reflectivity, the existence of a Bragg peak is not required and thus it can be used for any type of materials (ultra thin films, amorphous,…). We show that it can straightforwardly and unambiguously determine the main characteristics of chiral magnetic distributions in perpendicularly magnetized multilayers [4]: its chiral nature, the quantitative winding sense (clockwise or counterclockwise), and its type, i.e., Néel (cycloidal) or Bloch (helical). We will also show that this approach combined with micromagnetic simulations can be used to tailor hybrid chiral spin texture in multilayers [5]. I will also present how one can manipulate this magnetic chirality either statically using magnetic field [6] or at the femtosecond timescale using optical light [7].

In the last part I will present the latest development of resonant scattering and in particular how the use of the x-ray coherence already available at modern light source, and expected to increase by few order of magnitude in the planned upgrade sources, allow nowadays to image magnetic skyrmions [8,9] and chiral domains [9] wall with a spatial resolution of few tens of nanometers and a time resolution ranging from ns down to few ps timescale.

References
Structural involvement in the charge density wave in 1T-TiSe₂

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Static and dynamic resonant and non-resonant x-ray diffraction results are presented to study the interplay between the crystal structure and the electronic order of the Se 4p orbitals associated with the charge density wave (CDW) formation in 1T-TiSe₂. Different superlattice reflections are studied, with some appear far below the CDW phase transition. The static data give clear evidence for the occurrence of a second structural distortion within the CDW phase, [1] that is related to the changes in the electronic structure as recently found by circular photogalvanic current measurements that predicted a gyrotropic (chiral) electronic structure. [2] Our structural data can be fully described by the onset of Se out-of-plane displacements in a nonchiral crystal structure. We can relate the electronic modifications directly to this structural distortion which is also related to the change in transport properties. No indication for electronic or structural chirality is found by resonant x-ray diffraction in the ground state.

It is commonly assumed that the ultrafast laser pulse disturbs primarily the electronic order, which in turn destabilizes the atomic structure. Contrary to this conception, we show here that structural destabilization of few atoms causes melting of the macroscopic ordered charge-density wave in 1T-TiSe₂. Using ultrafast pump-probe non-resonant and resonant X-ray diffraction, we observe full suppression of the Se 4p orbital order and the atomic structure at excitation energies more than one order of magnitude below the suggested excitonic binding energy. [3] Complete melting of the charge-density wave occurs 4-5 times faster than expected from a purely electronic charge-screening process, strongly suggesting a structurally assisted breakup of excitonic correlations. Our experimental data clarify several questions on the intricate coupling between structural and electronic order in stabilizing the charge-density-wave in 1T-TiSe₂. The results further show that electron-phonon-coupling can lead to different energy-dependent phase-transition pathways in condensed matter systems, opening new possibilities in the conception of non-equilibrium phenomena at the ultrafast scale.

References
The trapping effect of an ANNNI Devil’s staircase revealed by coherent soft REXS.


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Frustration, the impossibility of satisfying at the same time all the two-point interactions present in a system, can generate exotic states, peculiar excitations and non-trivial dynamics. In magnetism, surprisingly simple perturbations of ordered states realized on specific geometries can result in deep effect on their global stability, so that a resulting high level of complexity can be indeed taken into account by fairly simple models. This is the case predicted by the Axial Next Nearest Neighbor Ising (ANNNI) model [1-4], where an AntiFerroMagnetic (AFM, next nearest) perturbation of a dominant FerroMagnetic (FM, nearest) interaction can result in a plethora of states, linked by an infinite series of transitions across a dense region of adjacent propagation vectors (the Devil’s staircase).

We investigated Lu$_2$CoMnO$_6$ by means of coherent soft X-ray Resonant Elastic Scattering (REXS) [5]. The system shows a specific magnetic structure, and commensurate to incommensurate transitions as a function of temperature. X-ray Photon Correlation Spectroscopy (XPCS) analysis of the REXS signal produced by the two families of available magnetic reflections revealed that this system can be well explained in terms of a modified ANNNI model. The characteristic Devil’s staircase traps the evolution of its magnetic state across a specific region of its phase diagram. The slow and continuous relaxation of the incommensurate magnetic propagation vector, and the inverted temperature dependence of its dynamics compared to the commensurate order, represent the two distinctive signatures predicted by the frustrated model in the specific case. The REXS investigation accesses the intimacies of the transitions and how the system relaxes to equilibrium, thus allowing to tune the microscopic interaction behind the frustration.

References

RIXS study of Infinite-layer Nickelate Superconductors

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After decades of efforts, the first nickelate superconductor has been recently discovered in Sr-doped infinite-layer nickel oxides Nd$_{1-x}$Sr$_x$NiO$_2$, soon followed by the doped Pr and La-doped infinite-layer nickelates. They are isostructural to the infinite-layer cuprates and nominally possess nine electrons in the 3d orbitals, reminiscent of cuprates. Yet, characterizing the underlying elementary excitations to identify the differences and similarities to the cuprates are critical to gain further insight into the microscopic mechanism of the nickelates. In this talk, I will present our recent XAS and RIXS study on the electronic structures and magnetic excitations in the infinite-layer nickelates Nd$_{1-x}$Sr$_x$NiO$_2$. We find that the parent compound of the infinite-layer nickelates is not a charge-transfer compound [1], unlike other nickelates and cuprates. Furthermore, magnetic excitations, which are found to be consistent with the spin wave of spin-$\frac{1}{2}$ antiferromagnetic interaction in a square lattice, possess high energy scales at zone boundaries [2], consistent with a Mott-Hubbard system in the strong onsite Coulomb interaction regime. Upon doping, the doped holes are found to possess significant weight of the Ni 3d$_{x^2-y^2}$ character in a spin-singlet state, while the Nd 5d states appears to be somewhat insensitive to the doped holes [3]. These results indicates that the strong correlation effects in the Ni 3d$_{x^2-y^2}$ orbital, in essence of the Mott physics, play an important role in sculpting the electronic properties of infinite-layer nickelates.

References
Time-resolved RIXS for quantum materials

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While time-resolved studies have been widely applied in optical ranges, there are a tremendous opportunities to study quantum materials using x-rays in the time-domain. These include the mapping of charge, spin, orbital and lattice collective modes, allowing for the assessment of couplings between them. Moreover, certain long-standing problems in the areas of materials science can now be addressed using time-resolved RIXS. I will present an overview of these opportunities and give a current synopsis of progress in this area.
Ultrafast manipulation of magnetic states holds great promise for progress in our understanding of new quantum states and technical applications, but our current knowledge of transient magnetism is very limited. Here we overcome persistent limitations of the field by using time-resolved resonant x-ray scattering, enabling studies of transient magnetic correlations throughout the entire Brillouin zone at ultrafast timescales. Using Sr$_3$Ir$_2$O$_7$ as a model material we demonstrate that femtosecond laser pulses can excite transient magnons at large wavevectors in gapped antiferromagnets and that they persist for several picoseconds [1]. This incoherent transient behavior is fundamentally different to what is observed in nearly gapless magnets such as Sr$_2$IrO$_4$ [2] - an isotropic Heisenberg-like material with comparable Néel temperature and similar energy scales. Here transient correlations are apparent only at the center of the magnetic Brillouin zone but not at its boundary. We interpret our results in the context of a spin-bottleneck scenario, suggesting that materials with isotropic magnetic interactions are preferred to achieve rapid manipulation of magnetism.

References
Roles of electron-phonon interactions in the phase transitions of rare-earth nickelates


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In recent years, correlated transition metal oxides have attracted significant attention due to their remarkable structural, electronic, magnetic, and optical properties that provide many ways to finely tune their functionalities. An example is the rare-earth nickelates, $R$NiO$_3$, which are unusual due to their negative charge-transfer behaviour with an electronic configuration of Ni $3d^8$ ($L = $ oxygen ligand hole) in their paramagnetic metallic state [1]. Most $R$NiO$_3$ (except for $R =$ La) undergo metal-insulating (MIT) and antiferromagnetic (AFM) transitions at low temperatures accompanied by a breathing distortion in their crystal structure [2,3]. This leads to a bond disproportionation where expanded NiO$_6$ octahedra (Ni $3d^8$ configuration, no holes on oxygen, and high spin) alternate with collapsed NiO$_6$ octahedra (Ni $3d^8L^2$, average of two holes on oxygen, and low spin), resulting in an AFM ordering vector of $q_{AFM} = (\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ pseudo-cubic [1–4]. This close relationship between breathing distortion and electronic configurations suggests that electron-phonon couplings (EPC) lie at the heart of the MIT in $R$NiO$_3$. Recent progresses in both theory [5,6] and instrumentation [7] have allowed the use of high-resolution resonant inelastic x-ray scattering (RIXS) to probe this EPC. Here, we use high-resolution RIXS at the SIX beamline [7] of National Synchrotron Light Source II to track the momentum- and temperature-dependent evolution of the EPC across the phase transitions of $R$NiO$_3$ thin films for $R =$ La, Nd, and Sm. We find that the EPC of the phonon mode related to the breathing distortion of NdNiO$_3$ reduces significantly by ~25% at the onset of its MIT just below 180 K. This drastic change of EPC is consistent with the polaronic condensation scenario [8], which proposes that the MIT of $R$NiO$_3$ is caused by the condensation (melting) of polaronic carriers at low (high) temperatures. Furthermore, we also find that the momentum-dependent EPC of AFM insulating NdNiO$_3$ becomes maximum at 0.5 $q_{AFM}$, signifying a strong magneto-elastic coupling consistent with a recent observation of possible multiferroicity in this material [9]. These results reveal the intimate connection between electronic, lattice, and spin degrees of freedom in $R$NiO$_3$ that can be exploited for future advanced functional devices, and demonstrate the capability of high-resolution RIXS in probing and even quantifying the electron-phonon interactions in advanced materials.

References
Resonant Quasielastic X-ray Scattering

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In this talk I will discuss using quasielastic x-ray scattering at resonance. This will be focused on quantum materials studies at low energy, and we will also explore the connection with both inelastic and elastic scattering. We will start with optical radiation, followed by a discussion of experiments associated with x-ray scattering studies in the time domain [1]. We will describe how these types of measurements are currently carried out with new advancements at X-ray Free Electron Laser sources [2]. After some method mechanics [3], we will describe measurements of relaxation of magnetic correlations [4]. We also examine the latest developments at LCLS in detector technology and instrumentation in this area and conclude with a perspective for future studies in this area.

References


Oxygen vacancies play a crucial role in the control of the electronic, magnetic, ionic, and transport properties of functional oxide perovskites. Rare earth nickelates (RENiO$_3$) have emerged over the years as a rich platform to study the interplay between the lattice, the electronic structure, and long-range magnetic order. However, the electronic phase diagram of oxygen-deficient nickelates has remained long unreported. In this study, we investigate the evolution of the electronic and magnetic structure in thin films of RENiO$_{3-x}$, using a combination of X-ray absorption spectroscopy and imaging, resonant X-ray scattering, and extended multiplet ligand field theory modeling. We find that oxygen vacancies modulate the electronic and magnetic ground state of RENiO$_{3-x}$ in unprecedented ways. First, oxygen vacancies directly alter the electronic configuration within the Ni-O orbital manifolds, leading to a dramatic evolution of long-range electronic transport pathways despite the absence of nanoscale phase separation. Second, the removal of oxygen sites bridging between neighboring NiO$_6$ octahedra affects the magnetic superexchange pathways that couple local moments and support long-range magnetic order. The resulting suppression of the antiferromagnetic order parameter is, surprisingly, not followed by a corresponding decrease of the magnetic transition temperature, until long-range magnetism is abruptly erased without an accompanying structural transition. We propose the progressive disruption of the 3D magnetic superexchange interaction network upon introduction of oxygen vacancies as the mechanism behind the sudden collapse of magnetic order in oxygen-deficient nickelates. Our work underscores the potential of oxygen vacancies in tuning correlated phases and triggering novel phase transitions in the transition metal perovskites.
Resonant Inelastic X-Ray Scattering to study ultrathin quantum materials

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The understanding of the interactions leading to the intriguing properties of quantum materials requires the investigation of their elementary excitations in energy and momentum space. In this context, Resonant Inelastic X-ray Scattering (RIXS) has emerged as a powerful probe with prime sensitivity to the electronic (spin, orbital, and charge) and lattice degrees of freedom. Thanks to recent developments RIXS has been employed in the investigation of many different systems, including cuprates, Fe-based superconductors, and low dimensional magnets. One of the latest interests has been the investigation of ultrathin films and materials where the properties are markedly different than the bulk.

In my talk I will present our recent RIXS investigations in the field of ultrathin films [1,2]. I will focus on two different cases, the effect of confinement on the spin excitations of metallic iron [1], and the evolution of the spin fluctuations in FeSe from the bulk down to the monolayer [2]. I will show how RIXS can identify the elementary excitations in samples as thin as a single unit cell and by comparison with the respective bulk materials, I will discuss how the limited thickness affects the spin excitations and the consequences in the description of the interactions of those systems.

In the case of ultrathin FeSe grown on SrTiO$_3$ the spin excitations are strongly hardened and flattened by the thickness. This can be explained thanks to Fermiology arguments accounting for the topological transition of the Fermi surface from the bulk to the monolayer. In the second case of metallic Fe the decrease of thickness affects anisotropically the three-dimensional spin excitations of the bulk ferromagnet. In the out-of-plane direction the spin excitations are gradually softened whereas the in plane spin excitations persist and retain their dispersion. This evolution can be explained by conventional Heisenberg models accounting for the number of broken magnetic bonds in proximity of the interface.

References
Possible application of time-resolved resonant-inelastic x-ray scattering to photoexcited Mott insulators

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The recent development of time-resolved resonant inelastic x-ray scattering (TRRIXS) opens a new avenue for probing collective two-particle excitation, from which one can investigate novel photoinduced nonequilibrium phenomena in the wide range of momentum and energy spaces. RIXS can probe not only charge excitation but also magnetic excitation if one uses incident x rays tuned for L edge in transition metals. If the lifetime of an intermediate state in the L-edge RIXS process is short enough, the dominant contribution to the RIXS spectrum comes from the dynamical charge and spin structure factors. It is also numerically shown that even for a realistic lifetime scale of an intermediate state in cuprate materials, the magnetic excitation in RIXS gives information on the dynamical spin structure factor [1]. Therefore, TRRIXS is an ideal tool for characterizing transient spin dynamics.

We theoretically investigate momentum dependent spin excitation that evolves after pumping within a femtosecond timescale in the antiferromagnetic Mott insulator on a square lattice [2]. Using a numerically exact-diagonalization technique based on the time-dependent Lanczos method for a half-filled Hubbard model with a 4x4-site lattice, we find novel momentum-dependent transient spin dynamics. In particular, we demonstrate characteristic temporal oscillations for the intensity of the dynamical spin structure factor, showing an antiphase behavior for two orthogonal directions in the momentum space, which are parallel and perpendicular to the electric field of a pump pulse. The same behavior is also seen in the static spin structure factor as demonstrated in Fig.1. Their oscillation period in time is determined by two-magnon excitation in the Mott insulator. This theoretical prediction will be confirmed for Mott insulating cuprates and iridates once TRRIXS is ready for a femtosecond timescale.

Fig. 1. Time t dependence of the static spin structure factor for the half-filled 4x4 Hubbard lattice with nearest-neighbor hopping $t_0=1$ and on-site Coulomb interaction $U=10$. The profile of the pump pulse is shown at the bottom left-hand side.

References

RIXS study of electron-phonon coupling in Cuprates and orbital excitations in (Li,Fe)OHFeSe


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The origin of unconventional superconductivity (SC) in Cu-based and Fe-based high-$T_c$ superconductors remains to be solved, which is one of the most important challenges in condensed matter physics. In conventional superconductors, SC is mediated by electron-phonon coupling (EPC). However, the role of EPC in unconventional superconductors is strongly debated. Spin fluctuations are believed to play an important role in Cu- and Fe-based superconductors, because once antiferromagnetism is suppressed, superconductivity will appear. On the other hand, the interplay between lattice vibrations and superconductivity cannot be ignored. For example, the superconductivity of the monolayer FeSe grown on SrTiO$_3$ can be raised to more than 65K due to the electron-phonon interaction [1]. Quantitative determination of EPC is very important for understanding unconventional superconductivity.

With an improved energy resolution, resonant inelastic x-ray scattering (RIXS) has recently become an effective tool for directly determining the momentum dependence of the EPC strength [2,3]. In this talk, by using RIXS at Cu $L_3$-edge and O $K$-edge, I will show the results of our recent studies on EPC in several cuprate families. This includes the doping, momentum and temperature evolution of EPC. The underlying interplay between EPC and charge order will also be discussed [4]. Finally, I will also show our recent RIXS study of (Li,Fe)OHFeSe with a $T_c$ over 40K, which has a Fermi surface and band structure similar to the monolayer FeSe. We clearly reveal the orbital excitations in (Li,Fe)OHFeSe and provide insights for understanding the high-$T_c$ in iron selenides.

References
2p3d RIXS angular dependence at spin-flip excitations: A method to identify the square peg in a round hole

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Conventional wisdom suggests that one photon that carries one unit of angular momentum (1\(\hbar\)) can change the spin angular momentum of a single magnetic site with one unit (\(\Delta M_s = \pm 1\hbar\)) at most following the selection rules in the case of strong 2\(p\) spin-orbit coupling. This implies that a two-photon process such as 2\(p3d\) resonant inelastic X-ray scattering (RIXS) can change the spin angular momentum of the system with a maximum of two units (\(\Delta M_s = \pm 2\hbar\)). Recent 2\(p3d\) RIXS measurements on NiO single crystal confirmed this where single and double spin-flip excitations were observed at the Ni\(^{2+}\) magnetic site [1].

While it is obvious for high spin Ni\(^{2+}\) ions in NiO possessing two unpaired 3\(d\) electrons that only two spins can change their angular momenta, the situation is more complicated for a high spin Fe\(^{3+}\) ion in \(\alpha\)-Fe\(_2\)O\(_3\). The ground state here is a \(^6A_{1g}\) with a projected spin momentum \(M_s = 5/2\). In this case there are possibly five spins that could be reversed. In this talk, I will first address the fundamental question: Can we manipulate more than two spins in magnetic systems using a two-photon process as RIXS? Guided by theoretical calculations, I will discuss the mechanism behind higher-order spin-flip processes in magnetic systems. This model is furthermore confirmed by measuring higher order (single, double, triple and quadrupole) spin-flip excitations and their angular dependence in a \(\alpha\)-Fe\(_2\)O\(_3\) single crystal at the ultra-high resolution I\(21\) RIXS setup (\(\Delta E = 32\ meV\)) at Diamond Light Source [2].

Next, I will show that by employing the polarization dependence of RIXS, one can directly probe the spin ordering with chemical and site selectivity. Applied on the prototypical ferrimagnetic mixed-valence system, magnetite ([Fe\(^{3+}\)]\(_A\)[Fe\(^{3+}\)\(_B\),Fe\(^{2+}\)]\(_B\)O\(_4\)), one can distinguish spin-flip excitations at the A and B antiferromagnetically coupled Fe\(^{3+}\) sublattices and quantify the exchange field. Furthermore, it is possible to determine the orbital contribution to the magnetic moment from detailed angular dependence measurements [3]. RIXS dichroism measurements performed at spin-flip excitations with nanometer spatial resolution will offer a powerful mapping contrast suitable for the characterization of magnetic ordering at interfaces and engineered spin textures.

2- H. Elnaggar, et. al., publication in preparation.
The term 'proximate Kitaev spin liquid' was coined for materials such as Na$_2$IrO$_3$ and $\alpha$-RuCl$_3$. They feature dominant bond-directional Kitaev exchange between spin-orbit entangled j=1/2 moments but nevertheless host magnetic order at low temperature instead of the wanted Kitaev quantum spin liquid with fractional Majorana excitations. The 'proximate spin liquid' reflects the expectation to find a regime above the magnetic ordering temperature in which aspects of the Kitaev spin liquid such as the fractional nature of excitations may be realized. Using resonant inelastic x-ray scattering (RIXS), we derive a comprehensive picture of the unusual magnetic excitations of Na$_2$IrO$_3$.

References
Resonant tender x-ray scattering from Ru-4d conduction electrons in the centrosymmetric skyrmion host Gd$_3$Ru$_4$Al$_{12}$

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The spin and orbital texture of conducting electronic states in magnetic solids is a focus of intense scrutiny, being relevant to a multitude of emergent phenomena. These include novel optical responses, nonlinear transport properties, as well as spin current generation and control. Complex magnets with sizable local moments and intermediate spin-orbit coupling, e.g. in Ruthenium-based oxides and intermetallics, are believed to be suitable targets in this regard.

Here, we report on the spiral ordering of Ruthenium spins, as driven by their interaction with local rare earth moments in centrosymmetric Gd$_3$Ru$_4$Al$_{12}$. Resonant elastic scattering of tender x-rays was carried out at beamline P09 of the PETRA-III synchrotron at DESY in Hamburg, Germany. A specialized all-in vacuum beam path suppresses absorption of x-rays in air ($E = 2.8$–3 keV).

We exploit the resonant x-ray scattering process (Fig. 1) as an element-specific probe, targeting the spin texture of the Ru-4d electrons directly. Through comparison of scattering signals at the Ru-$L_2$ and $L_3$ absorption edges, we disentangle the spin and orbital textures in real space. Our analysis converges on a picture of a distorted spiral texture for Ru-4d, which results from a competition between Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions and spin-orbit coupling.

The experimental result is discussed in the framework of the Kondo Hamiltonian. We highlight the relevance of our findings for the topological Hall effect in the field-induced skyrmion lattice phase of Gd$_3$Ru$_4$Al$_{12}$ [1], and for the mechanism underlying the novel emergent (or quantum) inductance response in the spiral phase [2].

References
Width of core level resonances in XAS and the importance of the induced intermediate state interferences for RIXS

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Low energy excitations (spin, orbital, charge) RIXS and REXS at transition metal $L_{23}$ edges or lanthanide $M_{45}$ edges are governed by the excitation of a core electron into a locally bound exciton and its subsequent decay into the ground-state or into a low energy excited state. Experimentally one can address the intermediate state of RIXS and REXS with the use of x-ray absorption.

Modeled on a finite Hilbert space the imaginary part of the intermediate state propagator of RIXS and REXS is given by a finite set of delta functions. The coupling to a continuum transforms these delta functions into peaks with a finite width. In an isolated atom the important decay channels are the Auger and Fluorescence decay. For RIXS / REXS these decay channels smoothen the resonance energy dependence and result in interference between the possible intermediate state resonances. They also shorten the lifetime of the core hole and thereby determining the extend of the low energy excitations made.

Additional decay channels of the local x-ray excited exciton emerge in solids. Besides the decay of the core hole by filling it with electrons from other shallow core states one can make the exciton decay by charge excitations on neighboring sites. Electrons from the valence bands can scatter of the core excited exciton into conduction band states.

In this talk we will introduce an efficient method to include these additional decay channels present in solids, but not in atoms. By example of calculations on some transition metal and rare earth compounds we will show that they can dominate the line width broadening in XAS. We will also elaborate what this means for the RIXS spectra and the intensity ratio of the possible excitations visible in RIXS.

X-ray absorption spectrum of SrTiO$_3$ calculated in orange based on a ligand field theory calculation based on density functional theory parameters. In blue the same ligand-field theory model but now coupled to the valence and conduction bands of SrTiO$_3$ using a bath discretization with 1000 points. No additional broadening was added. The XAS final state enters as the RIXS intermediate state and the coupling to a continuum allows one to describe possible charge excitations made with RIXS.

References
Orbital engineering in YVO$_3$-LaAlO$_3$ superlattices

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Heterostructures offer new opportunities to manipulate quantum materials and create novel and technologically relevant phases, such as superconductivity and magnetism. One possibility for the targeted manipulation of quantum states is the epitaxial stabilization of certain orbital occupations, pursued by the so-called orbital engineering. These local modifications of orbital occupations and orbital overlap are decisive for the strength and sign of magnetic superexchange interactions, and therefore stabilize new artificial spin orders. In addition, they are of great fundamental interest and could find important practical applications in future (antiferro)-spintronic devices [1] and all-oxide solar cells [2]. The success of these endeavors will crucially depend on a thorough understanding of the underlying mechanisms and their hierarchy. This is the central motivation of our study, in which we verify a detailed picture of the reconstructions in a prototypical correlated-electron system. In rare-earth vanadates ($R$VO$_3$), the interactions between spin, orbital and lattice degrees of freedom lead to a complex phase diagram, consisting of various spin and orbital orders that change as a function of lattice distortions [3].

To further explore this lattice coupling, here we use resonant x-ray reflectometry to probe the electronic structure of thin slabs of YVO$_3$ embedded in a superlattice with the band insulator LaAlO$_3$. We extend the previously established methods of reflectometry analysis to a general form applicable to $t_{2g}$ electron systems and extract quantitative depth-dependent x-ray linear dichroism profiles. Our data reveal an artificial, layered orbital polarization, where the average occupation of $xz$ and $yz$ orbitals in the interface planes next to LaAlO$_3$ is inverted compared to the central part of the YVO$_3$ slab (see Figure). This novel phase is stable down to 30 K, while bulk-like orbital ordering is absent. We identify the key mechanism for the electronic reconstruction to be a combination of epitaxial strain and spatial confinement by the LaAlO$_3$ layers, in good agreement with predictions from ab initio theory.

References
Coherent x-ray scattering studies of complex oxides

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Mesoscale phenomena play an important role in the dynamics of phase transitions in electronic materials. In order to fully understand and tailor nanoscale functionalities of electronic materials, detailed access to the nanoscale regime, correlation length scales and their temporal evolution is required. X-ray photon correlation spectroscopy (XPCS) provides a unique way to characterize nanoscale heterogeneities and their correlations across the phase transition. It allows us to study domain dynamics and fluctuations by capturing high resolution coherent speckle patterns in reciprocal space which can be considered as a fingerprint of the sample in real space. In this talk, I will present our recent XPCS studies across the phase transitions in complex oxides. The first part of my talk will focus on studying orbital domain dynamics in magnetite near instulator-to-metal transition. By tuning to Fe and O resonant studies, we studied the fluctuations in orbital system and found two distinct regimes below Verwey transition. The second part of my talk will focus on the evolution of domain fluctuations in low-strain BaTiO$_3$ thin films, and compare the behavior near the domain transformation (a/b to a/c type) temperature and the Curie temperature. The observed time evolution and reconfiguration of domain patterns highlight the role played by phase coexistence and elastic boundary conditions in altering fluctuation timescales in ferroelectric thin films.
New theoretical insights into how resonant inelastic x-ray scattering probes electron-phonon interactions in solids

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Resonant inelastic X-ray scattering (RIXS) is increasingly used to characterize low-energy collective excitations in materials and their coupling to charge carriers. However, RIXS experiments often require theoretical descriptions to interpret the data due to the complicated cross-section associated with the resonant inelastic scattering process. This aspect has created a need for accurate theories describing the influence of the electron-phonon interaction on RIXS spectra. To date, the majority of experiments have used a single-site framework \cite{1}, which neglects carrier mobility at all stages of the scattering process, to interpret the observed phonon excitations. I will discuss our recent work extending this formalism to account for electron itinerancy and other effects on lattice excitations using complementary density matrix renormalization group \cite{2} and variational approximation \cite{3} methods. Here, I will focus on the role of electron mobility and discuss the regimes where the single-site framework fails and how this might alter the interpretation of RIXS experiments.

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\cite{2} A. Nocera \textit{et al.}, Scientific Reports \textbf{8}, 11080 (2018).
Probing electron-phonon coupling away from the Fermi level with resonant inelastic x-ray scattering

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Electron-phonon coupling (EPC) is responsible for a wide range of condensed matter phenomena, from the basic transport properties of metals to collective phases such as superconductivity. Based on an ever-improving energy resolution, and seminal theoretical work by Ament et al. [1], there is growing interest in RIXS as a direct probe of EPC. Much of this interest is based on the momentum resolution of RIXS, which allows the EPC to be mapped throughout the Brillouin zone. The ability to tune the incident photon energy, however, has thus far been underexploited.

In this talk, I will demonstrate the ability of RIXS to probe EPC away from the Fermi level, setting it apart from established techniques such as angle-resolved photoemission and Raman spectroscopy. I will present carbon K-edge RIXS measurements of graphite, taken with the unprecedented energy resolution offered by beamline I21 at the Diamond Light Source. By tuning the incident x-ray energy, we are able to separately probe the couplings of the low-energy $\pi^*$ and high-energy $\sigma^*$ electronic states, finding qualitatively different spectra for each. To successfully model our data, we have to go beyond the commonly used model by Ament et al. [1] and instead employ a Green’s function method [2] to efficiently account for the full phonon dispersion.

Our results show how RIXS can be applied to a new range of technologically-important out-of-equilibrium situations. Further, they highlight the crucial role of the phonon dispersion in RIXS, even for relatively flat optical modes.

References
Resonant inelastic x-ray scattering study of vector chiral ordered kagome antiferromagnet

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We study the resonant inelastic x-ray scattering (RIXS) features of vector chiral ordered kagome antiferromagnets [1]. Utilizing a group theoretical formalism that respects lattice site symmetry, we calculated the $L$-edge magnon contribution for the vesignieite compound BaCu$_3$V$_2$O$_8$(OH)$_2$. We show that polarization dependence of the $L$-edge RIXS spectrum can be used to track magnon branches. We predict a non-zero $L$-edge signal in the non-cross $\pi-\pi$ polarization channel. At the $K$-edge, we derived the two-site effective RIXS and Raman scattering operator for two-magnon excitation in vesignieite using the Shastry–Shraiman formalism. Our derivation considers spin-orbit coupling effects in virtual hopping processes. We find vector chiral correlation (four-spin) contribution that is proportional to the RIXS spectrum. Our scattering operator formalism can be applied to a host of non-collinear non-coplanar magnetic materials at both the $L$ and $K$-edge. We demonstrate that vector chiral correlations can be accessed by RIXS experiments.

References

Posters - Abstracts
Resonant Elastic X-ray Scattering (REXS) is a unique element, site, and valence specific probe to study the charge, spin and orbital degrees of freedom and multipole orders in solids and thin films [1,2]. This technique, which combines features of diffraction and spectroscopy, has been successful in unraveling different order parameters and solving magnetic structures.

Concerning magnetic structure determination, REXS is complementary to neutron techniques, and some situations make it essential: when the involved magnetic species present a too large neutron absorption cross-section, like Eu, Dy, Gd… [3], when the magnetic moments cannot be determined unambiguously with neutron experiments [4], or when more than one magnetic species is involved.

Different types of data can be collected during a REXS experiment: intensities of a set of magnetic reflections, full linear polarization analysis data, or azimuthal dependence data. The analysis of these data is highly complex and no crystallographic software has been developed yet to enable users to solve magnetic structures from a REXS experiment.

MagStREXS is a crystallographic software dedicated to the determination of Magnetic Structures through Resonant Elastic X-ray Scattering and the preparation of magnetic diffraction experiments. It is under development since mid-2017 at beamline P09 [5] at PETRA III (DESY), and is based on CrysFML, a library developed to facilitate the creation of crystallographic software that includes some functionalities especially oriented to deal with magnetic structures and their symmetry.

Hereby, we will present an overview of MagStREXS, its current status and some of the magnetic structures which have already been solved with it in the field of highly correlated systems.

References
Using resonant coherent x-ray diffraction (RCXD), we study the formation of antiferromagnetic domains in the correlated antiferromagnet PrNiO$_3$. We demonstrate that it is possible to quantitatively extract the arrangements and sizes of the first-formed domains from single resonant coherent x-ray diffraction patterns. At the onset of the antiferromagnetic transition, the ordered domains are dilute in the beam spot, thus resulting in relatively simple coherent diffraction patterns, which can be inverted manually through a combination of visual inspection, system knowledge and trial and error. The success of our analysis suggests that a resonant Bragg coherent diffractive imaging approach with iterative phase retrieval algorithms may be effective in studying both these and even more complex antiferromagnetic spin textures. As an outlook, we argue that the same approach could be extended to a time-structured light source in order to study the motion of dilute dynamically driven domains or to track the motion of topological defects in an antiferromagnetic spin texture.

Figure: Progression of the antiferromagnetic RCXD pattern as temperature is cycled through the coupled antiferromagnetic / metal-insulator transition of PrNiO$_3$. 
Integrating machine learning into the setup of X-ray scattering setups

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The role of machine learning and computer vision in materials science has grown in recent years, and opportunities have arisen for the application of these approaches in X-ray scattering. In this work, we discuss how supervised machine learning techniques can be integrated in the experimental setup of the setup of X-ray photons for scattering studies. The goal is to obtain state-of-the-art automated results, and we focus on models like random forests and artificial neural networks.
A-type antiferromagnetism in a room van der Waals magnetic metal

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The emerging cleavable two-dimensional van der Waals (vdW) magnetic metals present attractive research platforms for studying the low-dimensional magnetism and also offer opportunities for spintronic applications \cite{1-6}. One of the recently uncovered vdW magnets, Fe\textsubscript{5-x}GeTe\textsubscript{2} (Fe5GT), has a bulk ferromagnetic ordering temperature at \(\sim 315\)K \cite{7-8}. Its magnetic order persists at similarly high temperature in exfoliated flakes with thicknesses down to 10 nm. More intriguingly, with Co substitution into this compound, the magnetic ordering temperature further increases with increasing Co doping concentration \cite{9-10}. Meanwhile the magnetic ground state evolves from ferromagnetic to antiferromagnetic, in concomitant to the alternation of stacking order with increasing Co replacement. However, details of the antiferromagnetic order and how it evolves with doping are still unknown. Here, by utilizing the resonant soft x-ray scattering technique, we investigated the magnetic properties of a 45\% Co doped Fe5GT sample. The temperature and field dependent magnetic ground state was studied at both Fe and Co L edges. Our study highlights the important role of both Fe and Co in governing the magnetic properties of this compound.

References
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Site-specific spectroscopic measurement of spin and charge in (LuFeO$_3$)$_m$/ (LuFe$_2$O$_4$)$_1$ multiferroic superlattices

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Interface materials offer a means to achieve electrical control of ferrimagnetism at room temperature as was recently demonstrated in (LuFeO$_3$)$_m$/ (LuFe$_2$O$_4$)$_1$ superlattices$^1$. A challenge to understanding the inner workings of these complex magnetoelectric multiferroics is the multitude of distinct Fe centres and their associated environments. This is because macroscopic techniques characterize average responses rather than the role of individual iron centres. Here, we combine optical absorption, magnetic circular dichroism and first-principles calculations to uncover the origin of high-temperature magnetism in these superlattices and the charge-ordering pattern in the m = 3 member. In particular, interface spectra establish how Lu-layer distortion selectively enhances the Fe$^{2+}$ to Fe$^{3+}$ charge-transfer contribution in the spin-up channel, strengthens the exchange interactions and increases the Curie temperature. Comparison of predicted and measured spectra also identifies a non-polar charge ordering arrangement in the LuFe$_2$O$_4$ layer. This site-specific spectroscopic approach opens the door to understanding engineered materials with multiple metal centres and strong entanglement.

References
Strain-induced orbital energy shift in antiferromagnetic RuO$_2$ revealed by resonant x-ray scattering

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In the ruthenium perovskite family, epitaxial strain has been shown to enhance magnetization and tune $T_c$ in superconductors. Recently strain engineering has been shown to induce superconductivity in the previously non-superconducting metal RuO$_2$ [1]. Long thought to be an ordinary, paramagnetic metal, neutron diffraction has revealed a surprising antiferromagnetic ground state in RuO$_2$ [2,3]. Here we present a magnetic resonant x-ray scattering (RXS) study of the same RuO$_2$ films used to demonstrate strain-induced superconductivity. At the Ru-$L_2$ edge we can access the forbidden magnetic reflection (100) and probe the magnetic ground state, domain sizes and electronic properties. We notably observe a large, strain-dependent shift of Ru $e_g$ orbitals to higher energy in the sample that hosts novel superconductivity.

References

Microscopic study of the spin Seebeck effect in YIG with resonant inelastic x-ray scattering

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The SSE in insulating Y3Fe5O12 (YIG) was discovered in 2010, defining it as a spin-wave-based transport effect generated by applying a temperature gradient [1]. Since then a multitude of transport studies have been reported indicating the temperature and thickness dependence, and the effect of magnetic field on SSE [2-3]. From a theoretical perspective, many models have been built to explain the experimental results [4-6]. However, no consensus has been reached on the mechanism behind the SSE in YIG; this is due to the lack of a suitable microscopic probe compatible with temperature gradient conditions and sensitive to the elementary excitations leading the transport.

Here, we present state-of-the-art resonant inelastic x-ray scattering (RIXS) results on a YIG device operating in presence of spin Seebeck effect (SSE). The study uniquely reveals a momentum (q) and energy (\(\omega\)) resolved picture of the excitations involved in the SSE, providing key information on the microscopic mechanism behind this transport phenomenon. By controlling the applied temperature gradient across the YIG device, our RIXS data display a clear change of the spectral weight: for increasing temperature gradient, the spectral weight is enhanced when q is parallel to the magnon current direction, while suppressed when q is antiparallel to the magnon current direction. This opposite behaviour at opposite q directions proves that our measurement is sensitive to the flowing magnon current. More specifically, we detect the shift of the magnon distribution induced by the temperature gradient with (q,\(\omega\)) resolution. This result on one hand uncovers for the first time the direct observation of the magnon current, thanks to the sensitivity of RIXS. On the other hand, the fine details of the spectral weight variation versus energy and momentum point to a crucial role of low energy acoustic spin wave mode in the SSE.

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Spectroscopic characterization of electronic structures of ultra-thin single crystal \( \text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3 \)

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We have successfully fabricated high quality single crystalline \( \text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3 \) (LSMO) film in the freestanding form that can be transferred onto silicon wafer and copper mesh support. Using soft x-ray absorption (XAS) and resonant inelastic x-ray scattering (RIXS) spectroscopy in transmission and reflection geometries, we demonstrate that the x-ray emission from Mn 3\(s\)-2\(p\) core-to-core transition (3\(s\)PFY) seen in the RIXS maps can represent the bulk-like absorption signal with minimal self-absorption effect around the Mn L\(_3\)-edge. Similar measurements were also performed on a reference LSMO film grown on the SrTiO\(_3\) substrate and the agreement between measurements substantiates the claim that the bulk electronic structures can be preserved even after the freestanding treatment process. The 3\(s\)PFY spectrum obtained from analyzing the RIXS maps offers a powerful way to probe the bulk electronic structures in thin films and heterostructures when recording the XAS spectra in the transmission mode is not available.
The nature of electron itinerancy is a fundamental descriptor of a solid-state material. In the case of insulators, band and Mott insulators represent two drastically different regimes: the former can be described using spatially extended electron wavefunctions and well-defined quasiparticles, while the latter features localized electron wavefunctions with strong local correlations. Recently the nature of the insulator behavior has been under intense investigation in a number of 5d transition metal compounds due to the more extended nature of the 5d orbital wavefunction. Addressing this problem and pinpointing the role of electron correlation calls for experimental observations that can uniquely distinguish the Mott vs. band insulator limit. Here we perform resonant inelastic X-ray scattering studies of 1T-TaS$_2$ which has till recently been long considered a prototypical Mott insulator. We observed five electronic excitations arising from the interband transitions of the Ta 5d orbitals and the S 3p ligand state at the Ta L$_3$ edge (figure (a) and (b)). These excitations cannot be explained within the framework of standard molecular orbital multiplet calculations that are based on a localized picture with strong electronic correlations (fig. (b)). Instead, calculations from the band dispersions can reasonably capture the number and energies of the orbital excitations with small deviations. We argue that the insulating phase of 1T-TaS$_2$ is on the cusp of band-Mott insulator crossover and leans heavily towards the band insulator limit. Our discovery provides new insight into the Mott vs. band insulator debate in TaS$_2$ and other 5d transition metal materials and puts RIXS as a new tool capable of detecting the orbital excitations in materials with weaker electron correlation and localizations.

Figure (a): RIXS spectra as a function of the incident energy across the Ta L$_3$ resonance at 10K. The excitations’ energy positions are marked in red roughly; (b) the RIXS spectrum and the corresponding fitting peaks at the incident energy of 9882 eV. Multiplet calculation results with/without spin orbital coupling (SOC) are listed in below, respectively.
Interface carriers and enhanced electron-phonon coupling effect in Al2O3/TiO2 heterostructure

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Recent works demonstrated the existence of two-dimensional electron system (2DES) in an Al2O3/TiO2 thin-film heterostructure with a surprisingly high sheet carrier density (~10^14 cm^-2) [1,2]. Al2O3/TiO2 2DES does not necessitate the two confronting lattices’ epitaxial conjunction for its structural and chemical stabilization [1]. The loose requirement of achieving a 2DES is an advantage because it allows compatibility to an industrial-mass-production fabrication. Therefore, it is crucial to verify and characterize the 2DES (i.e., Ti^{3+} species at the interface) to develop the binary-oxide-based 2DES further. In this work [3], we study the electronic structure and electron-phonon couplings in an Al2O3/TiO2 thin film with 2DES using resonant inelastic X-ray scattering (RIXS). The RIXS results (FIG. 1a) unequivocally show that the Ti^{3+} state indeed exists at the deep interface to serve as an n-type dopant for the 2DES. The electronic structure of Ti^{3+} species is scrutinized as entirely separated from that of the Ti^{4+} host lattice. Furthermore, features of sub-eV energy loss phonon modes are clearly observed, indicating substantial electron-phonon coupling effects. Such low energy loss features are enhanced in thinner TiO2 samples, implying that polaronic local lattice deformation is enhanced due to the presence of Ti^{3+} (FIG. 1b). These findings suggest that the 2DES properties can be controlled via well-established TiO2 engineering, thereby enthroning the binary oxide heterostructure as a promising candidate for 2DES device applications.

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DOI:10.1002/adfm.202104430

FIG. 1 (a) Ti L3 edge RIXS spectra. (b) Illustration of the electron-phonon coupling effect.
Cobalt-based oxides are complex and interesting because of the multiple electronic configurations of cobalt cations. Co in LaCoO$_3$ (LCO) is in 3+ and has multiple spin states depending on temperature: low-spin ($S=0$, $t_{2g}^6 e_g^0$), intermediate-spin ($S=1$, $t_{2g}^5 e_g^1$), and high-spin states ($S=2$, $t_{2g}^4 e_g^2$), which have nearly degenerate ground state energies. Recently, we have synthesized LaCoO$_3$/LaTiO$_3$ superlattices displaying strong orbital polarization [1]. Density functional theory (DFT)+U calculations show that Co in these superlattices is 2+ due to the charge transfer from Ti to Co, with two relevant low-energy spin states (low-spin $S=1/2$, $t_{2g}^6 e_g^1$ and high-spin, $S=3/2$, $t_{2g}^5 e_g^2$) [2,3].

Here, we combine a DFT+U based theoretical approach and Resonant Inelastic X-ray Scattering (RIXS) to compute RIXS spectra. Since the DFT+U calculations can capture the physics of both the dispersive and localized bands, this approach is capable of describing the broad, fluorescent-like features in RIXS (e.g., which are strong features for RIXS of LCO and LaNiO$_3$) as well as localized excitations (e.g., characteristic RIXS features of LCO+LTO superlattice). We find that this theoretical approach allows us to interpret the RIXS spectra for Co- and Ni-based bulk materials and superlattices.

References
Multi-spin excitations in an antiferromagnetic S=5/2 system

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Hematite (α-Fe\textsubscript{2}O\textsubscript{3}), an S=5/2 antiferromagnetic system, is a promising candidate in spintronic and magnonics due to its functional spin-related properties \cite{1,2}, such as a weak in-plane magnetic anisotropy and a low spin dissipation.

A key element for the microscopic understanding of spin-wave-based transport phenomena in insulators is the knowledge of the intrinsic spin dynamics of the system, involved in both the transport mechanism as well as in the dissipation processes. The spin dynamics of bulk hematite has been studied in the early days by inelastic neutron scattering (INS) \cite{3}, revealing the presence of an acoustic and an optical single-magnon branch up to 100 meV. The evolution of the spin dynamics in thin films - as the ones used for devices - remains still unknown due to the lack of suitable probes.

Recently, resonant inelastic x-ray scattering (RIXS) has emerged as a highly performing technique for the study of the spin dynamics in magnetic thin films \cite{4}. Additionally, it has shown great sensitivity in unraveling the higher ranked spin excitations such as Δs=0 and 2 \cite{5,6}, elusive to conventional techniques like INS. Here, we extend this approach to hematite thin films by using Fe L\textsubscript{3}-edge RIXS. Below 100 meV, we resolve two modes fully consistent with the Δs=1 magnon branches detected by INS in the bulk whereas at higher energy (up to 200 meV), multi-spin excitation modes are identified. With the support of LDA+DMFT calculations built around the Anderson impurity model \cite{7}, we interpret these to be Δs=2 and Δs=3 spin excitations at single magnetic site, respectively. These observations shed new light on the high-energy eigenstates of the hematite magnetic Hamiltonian, providing novel information on the spin dynamics and suggesting new pathways for the transport dissipation processes.

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Evidence of fractional magnetic excitations in a large ring-exchange cuprate observed by high resolution RIXS


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The spin ½ square-lattice antiferromagnet is one of the most studied systems in condensed matter theory, both because it is purely quantum mechanical in nature and because of its connection to the problem of High-Critical Temperatures superconductivity. After the seminal paper by Anderson, much effort has been put in the quest for exotic ground states and excitations. In particular, theoretical studies have predicted the existence of different forms of Resonating Valence Bond (RVB) states in many Heisenberg Hamiltonians with either next-nearest neighbour, frustrating couplings (e.g. $J_1$-$J_2$ model [1]) or multi-spin couplings (e.g. $J$-$Q$ models [2]). These states are interesting because they cannot be described by the canonical Linear Spin Wave Theory (LSWT), and exhibit fractionalized collective excitations, each carrying spin ½, that are the 2-dimensional version of spinons in 1D spin chains. Despite many efforts, these states have so far eluded conclusive observation in the AF square lattice. Moreover, a recent exact diagonalization study [3] has shown that the ring-exchange interaction, which couples 4 spin across a square and is sizable in most layered cuprates, might drive as well quantum phase transition towards a RVB ground state. In this context, we have used Cu L₃ Resonant Inelastic X-ray Scattering (RIXS) to study the spin excitations of CaCuO₂, a compound where CuO₂ antiferromagnetic planes are indefinitely stacked and separated by Ca ions. This compound is special for the very large nearest neighbour AF coupling $J$ (∼180 meV), and an exceptionally large $J_c \sim J$. At low momentum transfer in reciprocal space, we find that the magnetic spectrum is well described by a single magnon as predicted by LSWT. However, close to the magnetic zone boundary, we observe a strong decrease of the magnon spectral weight, which decays into a broad asymmetric continuum at high energies, accounting for more than 80% of the total spectral weight. Polarization analysis on the scattered beam reveals that this continuum entirely belongs to $\Delta S=1$ excitations. All these properties are in line to what is expected for a two-spinon continuum. Therefore, we propose that this compound lies close to the quantum phase transition driven by the ring exchange predicted by the theory, and we interpret these high energy excitations as a continuum of spinons.

Ultrahigh Resolution $h\nu^2$ RIXS Spectrometer at New 3 GeV Synchrotron Facility in Tohoku

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A new 3 GeV ring is under construction in Tohoku, Japan and is scheduled to be operational in 2024. In this synchrotron radiation facility, we are planning to build an ultrahigh resolution RIXS system by integrating an $h\nu^2$ RIXS spectrometer and a dedicated beamline. An APPLE-II type undulator is employed to provide variable polarization (linear horizontal and vertical, and left and right circular polarization) and covers the energy range of 250–2000 eV. The beamline was designed specifically for the $h\nu^2$ RIXS spectrometer [1–3], and the dispersed light from the monochromator will be irradiated directly onto the sample. The target total energy resolution (combined resolution of the beamline and RIXS spectrometer) is <10meV below $h\nu=1000$eV. In such an ultrahigh resolution, measurement efficiency is one of the factors that determine the energy resolution [4]. Therefore, to solve the bottleneck caused by the very low count rate in the ultrahigh resolution RIXS, we decided to employ the $h\nu^2$ RIXS spectrometer to improve the measurement efficiency and realize the RIXS facility with single meV resolution.

We will construct a ~76 m long beamline with ~12 m long RIXS spectrometer which can rotate continuously by 120° in the longest area in the synchrotron radiation facility. For the $h\nu^2$ RIXS spectrometer, the vertically energy-dispersed X-ray from the beamline monochromator is horizontally focused on the sample. An imaging mirror in the RIXS spectrometer vertically images scattered X-ray from the sample on a position-sensitive detector with keeping its relationship between the vertical position and the incident energy. Simultaneously, the grating in the RIXS spectrometer horizontally disperses and focuses the scattered X-ray on the detector. Thus, the energies of incident and scattered X-ray are resolved on a two-dimensional imaging detector. Therefore, the horizontal focusing size on the sample and the imaging characteristics to the detector are important to achieve the ultrahigh resolution by the $h\nu^2$ RIXS spectrometer. For the imaging mirror, a monolithic Wolter type I mirror will be employed. In the presentation, we will introduce the optical design of the RIXS spectrometer, especially the optimization of the imaging mirror and outline the improvement of the efficiency and the feasibility of the ultrahigh resolution by the $h\nu^2$ RIXS spectrometer.

References
Electrical tuning of Metamagnetic Transition of Quasi-2D $J_{\text{eff}} = 1/2$ Antiferromagnet by In Situ Anisotropic Strain

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With the emergence of antiferromagnetic (AF) spintronics in recent years, Iridates have attracted great interests because of their exotic ground state with magneto transport properties, driven by the interaction between strong spin-orbit coupling (SOC) and electron correlations. As a prominent example, Sr$_2$IrO$_4$ is a quasi-two-dimensional Jeff =1/2 canted AF Mott insulator. Similar to S=1/2 moments, $J_{\text{eff}}$=1/2 moments have no single-ion anisotropy. They are however also very different in that the $J_{\text{eff}}$=1/2 moments can form significant inter-site quadrupoles that are highly sensitive to lattice distortion via the so-called ‘pseudo-JT’ effect. The strength and symmetry of the pseudospin-lattice coupling can be externally controlled by the application of in situ strain. In this work, we investigate the tuning of the metamagnetic transition of the AF structure in Sr$_2$IrO$_4$ by applying anisotropic strain of the B$_2$g symmetry. By measuring the magnetoresistance (MR) and angular dependence of MR at different magnitude of in situ anisotropic strain, we observed clear shifts of the critical field of the metamagnetic transition. X-ray resonance magnetic scattering (XRMS) is performed to reveal the underlying modulation of the AF structure.
Modification of Cu-charge density wave at a YBa$_2$Cu$_3$O$_7$/manganite interface

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We present a resonant inelastic and elastic X-ray scattering (RIXS/REXS) study of epitaxial YBa$_2$Cu$_3$O$_7$/Nd$_{1-x}$(Ca$_{1-y}$Sr$_y$)$_3$MnO$_5$ heterostructures (NYN). We show that the Copper charge density wave (Cu-CDW) order of the near optimally doped YBa$_2$Cu$_3$O$_7$ layers can be strongly modified via the hole doping and tolerance factor of Nd$_{1-x}$(Ca$_{1-y}$Sr$_y$)$_3$MnO$_5$, i.e. by changing x and y.

At $x=0.35$ we observe a quasi-2D Cu-CDW order with $d_{z^2-r^2}$ orbital character that resembles the one that is commonly found in strongly underdoped bulk YBCO [1]. The strength of the corresponding Bragg peak at $Q_z=0.3$ r.l.u. gets strongly enhanced as the tolerance factor of the manganite layers $^5$ is decreased and its CE-type antiferromagnetic and charge/orbital ordered (COO) is reinforced.

Upon increasing the hole doping of the manganite layers to $x=0.5$, we observe a new kind of Cu-CDW order which has a much smaller wave vector of $Q_z=0.1$ r.l.u., a larger correlation length of about 40nm and a different orbital character, i.e. $d_z$ rather than $d_{z^2}$, than the one commonly found in the bulk cuprates[2].

The origin of this new Cu-$d_z$ charge order is presently not understood but seems to be rooted in the particular properties of the cuprate/manganite interface. The RIXS and additional x-ray absorption spectroscopy (XAS) data provide evidence for an important role of the orbital reconstruction of the Cu-ions at the interface with the manganite and a related transfer of electrons from the manganite to the cuprate. In particular, they show that the Cu-$d_z$ orbital of the interfacial Cu ions is strongly shifted up in energy and lies close to the Fermi-level such that it contains a significant part of the hole carriers, which usually mainly reside in the Cu-$d_{x^2-y^2}$ orbital [1,2]. This orbital reconstruction may well exhibit a lateral modulation along the interface that is linked with the anomalous $d_z$-type Cu-CDW order. While further studies are required to fully understand the interfacial coupling mechanism(s), the possibility of tuning the Cu-CDW holds great prospects for studying its relationship with high temperature superconductors and hopefully, for future quantum devices.

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Charge Condensation and Lattice Coupling Drives Stripe Formation in $\text{La}_{2-x}\text{Sr}_x\text{NiO}_{4+\delta}$

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Revealing the predominant driving force behind symmetry breaking in correlated materials is sometimes a formidable task due to the intertwined nature of different degrees of freedom. This is the case for $\text{La}_{2-x}\text{Sr}_x\text{NiO}_{4+\delta}$, in which coupled incommensurate charge and spin stripes form at low temperatures. Here, we use resonant x-ray photon correlation spectroscopy at Ni $L_3$-edge to study the temporal stability and domain memory of the charge and spin stripes in $\text{La}_{2-x}\text{Sr}_x\text{NiO}_{4+\delta}$. Although spin stripes are more spatially correlated, charge stripes maintain a better temporal stability against temperature change. More intriguingly, charge order shows robust domain memory with thermal cycling up to 250 K, far above the ordering temperature, while the spin order loses its domain memory once the sample is warmed across the magnetic transition temperature. These results demonstrate the pinning of charge stripes to the lattice and that charge condensation is the predominant factor in the formation of stripe orders in nickelates.

References
Electron-phonon excitations in the 1D Hubbard-Holstein model probed by Resonant Inelastic X-Ray Scattering

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The prospect of accessing electron-phonon (e-p) coupling strengths using resonant inelastic x-ray scattering (RIXS) has attracted significant attention in recent years. This is largely due to the theoretical framework developed in Ament \textit{et al.} \cite{Ament2011} that provides an analytic expression to derive e-p coupling from RIXS measurements. Essentially the knowledge of the one and two-phonon intensity suffices, as it maps directly to the ratio between the e-p coupling strength and the core-hole lifetime. This method assumes that the electrons in the system are completely localized during all stages of the RIXS scattering process. A recent study by Bieniasz \textit{et al.} \cite{Bieniasz2020} that incorporates electron mobility into the problem, demonstrated that the local approximation underestimates e-p coupling obtained from multi-phonon peak analysis. Furthermore, the electron mobility may also induce a momentum dependence on the phonon excitations that is not present in the underlying e-p coupling constant. These predictions however are mostly applicable to band insulators due to the limits of the variational method adopted by Bieniasz \textit{et al.}. Here, we study this problem in the strongly-correlated many-particle limit using DMRG \cite{Nocera2018}. By explicitly computing the RIXS intensity, we study the phonon excitations in long finite-sized Hubbard-Holstein chains, and compare our results against both the single-site \cite{Ament2011} and single-particle \cite{Bieniasz2020} limits.

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Crossover of the high-energy spin fluctuations from collective triplons to localized magnetic excitations in doped Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$ cuprate ladders

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We studied the low-dimensional doped ladders in Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$ by Cu L$_3$-edge RIXS. We observed an evolution from collective two-triplons ($x = 0$) to localized modes $\sim$280 meV ($x = 12.2$). Density matrix renormalization group calculations suggest that the flat magnetic dispersion at $x = 12.2$ comes from hole localization. This is supported by polarization-dependent RIXS results with decoupled non-spin-flip $\Delta S = 0$ and spin-flip $\Delta S = 1$ channels. The depleted $\Delta S = 0$ weight at low energy for $x = 12.2$ agrees with carrier-immobility. Our results show that interstitial rock salt impurities affect both the magnetic and superconducting correlations in CuO$_4$ plaquettes. We characterize the spin and charge fluctuations in the phases where pairing in SCCO emerges with high Ca content at elevated pressure.
Ultrafast dynamics and coupling of magnetic sublattices in room-temperature multiferroic hexaferrite

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Understanding ultrafast magnetism, such as demagnetization, all-optical magnetization switching, and even cross-correlation with other degrees of freedom, e.g., phonons, has created a recent significant interest in the field of condensed matter physics due to the potential for applications in data storage technologies. In addition, it is fundamentally interesting how such processes work and what drives them at the atomic (spin) level. Although most investigated systems are metallic, magnetoelectric multiferroics of type II bear great potentials because they have an intrinsic correlation among ferromagnetic (FM) and antiferromagnetic (AFM) sublattices and electric polarization ($P$). So far, however, ultrafast dynamics in multiferroics have been much less investigated \cite{1}.

In this presentation, we report time-resolved resonant x-ray diffraction (tr-RXD) to study ultrafast magnetism in a room-temperature hexaferrite possessing FM and AFM sublattices \cite{2}. An advantage of tr-RXD over conventional optical techniques is that we can separately investigate the sublattices having different propagation vectors. Namely, we followed two reflections after 400 nm fs optical laser excitation; one that is sensitive to the AFM component and one that is sensitive to the FM component. We observed an oscillation in tr-RXD intensities of both reflections with the same frequency but with opposite phase (see Fig. 1). This observation indicates a coherent excitation of a magnon mode triggered by the above-bandgap excitation. The two magnetic sublattices entangle in the magnon mode and should give rise to transient modulation of $P$. Such a coupled mode is known as an electromagnon \cite{3}, involving two characters, phononic and magnonic. It enables simultaneous ultrafast control of the two characters. A microscopic mechanism of the coherent magnon excitation is proposed based on the ultrafast back-and-forth interactions between the spins and the lattice: direct modulation of magnetic frustration via transient lattice distortion launched by the ultrafast demagnetization.

![Fig 1. Tr-RXD intensities of (a) the AFM-sensitive (003) reflection and (b) FM-sensitive (004) reflection measured at 84 K (black) or room temperature (the others).](image)

References

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Damping of intermediate-spin excitonic dispersion in LaCoO$_3$ by thermal fluctuations

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The charge, spin, and orbital degree of freedoms play an essential role in the physics of phase transitions in solids. LaCoO$_3$, a compound exhibits multiple spin-state crossovers which coupling with complex magnetic and electric phase transition. At low temperature, LaCoO$_3$ is a nonmagnetic insulator with Co ions in the low-spin (LS, $S = 0$, $t_{2g}^6 e_g^0$) ground state. Upon heating, it undergoes a crossover to a paramagnetic Curie-Weiss insulator ($T \sim 100$ K) and, eventually, a Curie-Weiss metal ($T \sim 500$ K). Traditionally, the spin-state crossover has been described as a thermal population of excited atomic multiplets. Despite its long history, the opinion on the nature of the first-excited Co$^{3+}$ multiplet remains split between the high-spin (HS, $S = 2$, $t_{2g}^4 e_g^2$) and intermediate-spin (IS, $S = 1$, $t_{2g}^5 e_g^1$) states.

We present Co L$_3$-edge resonant inelastic x-ray scattering (RIXS) of bulk LaCoO$_3$ across thermally induced spin-state crossover around 100 K. Tomiyasu et al. has shown that the Co L$_3$-edge RIXS is a unique method to study further details of the electronic structure on LaCoO$_3$[1]. Owing to a high energy resolution of 25 meV, we observe unambiguously the dispersion of the IS excitations in the low-temperature regime [2,3]. Approaching the intermediate temperature regime, the IS excitations are damped and the bandwidth is reduced. The observed behavior can be well described by a model of mobile IS excitons with strong attractive interaction, which we solve by using dynamical mean-field theory for hard-core bosons [3]. Our results provide a detailed mechanism of how HS and IS excitations interact to establish the physical properties of cobaltite perovskites.

References
Van der Waals 2D cuprate superconductors – potential device building blocks for quantum applications

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A multitude of exotic electronic structures facilitating intricate quantum states has been revealed in recent experiments performed on 2-dimensional van der Waals (vdW) heterostructures and superconductors [1-4]. In contrast to bulk crystals, the dimensionality of vdW heterostructures confine electrons in low dimension modifying carrier densities, their symmetry, and properties. The enhanced transport characteristics caused by the restraints of the vdW heterostructure can be harnessed in the design of novel ultra-thin devices, which would be potential candidates for advancements in quantum computing [5], such as already developed gate tuneable vdW 2D BSCCO (Bi\textsubscript{2}Sr\textsubscript{2}CaCu\textsubscript{2}O\textsubscript{8+x}) heterostructures (Fig. 1\textsuperscript{a}) [6].

![Image](image-url)

**Fig. 1:** (a) gate tuneable vdW 2D BSCCO heterostructure [6], (b) STEM image of the BSCCO crystal structure showing ILMs [7], reciprocal space map showing LRO and SRO reflections around the [002] Bragg peak [7], coherent soft X-ray nanodiffraction maps of a thin BSCCO flake ($T_c \approx 60$ K) at (d) 30 K and (e) 150 K, and (f) optical microscopy image of the flake.

Bulk BSCCO crystals are characterised by two incommensurate lattice modulations (ILMs), a much stronger long- (LRO) and a weaker short-range (SRO) order (Fig. 1\textsuperscript{b-c}). Because the bulk properties of BSCCO seem to be preserved down to a few atomic layers while the origin of ILMs is not entirely established (purely structural or mixed with the Cu planes electronics), thin BSCCO crystals are interesting candidates to corroborate if and how the relation between structural and electronic contributions evolves at low dimensionality. In this work, we have investigated a thin BSCCO flake partially protected by a layer of hexagonal boron nitride (hBN) using resonant coherent soft X-ray zone plate based nanodiffraction and report for the first time ILMs and structural [004] peak maps taken across the superconducting transition temperature ($T_c \approx 60$ K) (Fig. 1\textsuperscript{d-f}). We believe this sheds light onto the structure-electronics interplay as we find the intensity of ILMs depends on the superconducting state of the flake while [004] seems not.

**References**

Resonant Soft X-ray Study of Spin Stripe Domain Fluctuations in a Complex Oxide

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The emergence of novel quantum states in transition metal oxides, leaded by strong correlations between spin, charge, lattice, and orbital degrees of freedom, have fascinated researchers for over decades [1]. In these states, the electrons exhibit collective behaviors due to the symmetry breaking and are often represented in the energy domain. Recent X-ray photon correlation spectroscopy has revealed the instability of spin-density wave order (SDW) in La\textsubscript{1.775}Sr\textsubscript{0.225}NiO\textsubscript{4} [2,3]. Here, we present the spatial distribution of the spin order inhomogeneity by applying resonant x-ray ptychography to, mapping the SDW at the 70 K onset temperature. With the real-space image of SDW domains, it is found these domains are anisotropic in size and space. However, for SDW domain, its coherent length of SDW along different direction are strongly coupled in space. This indicates that thought the coupling of spins within each metal-oxygen plane is much stronger that between spin in neighboring planes, both the two coupling of spins has the effect on the SO dynamics. We suggested that the observed slow dynamics for SDW in LSNO is attributed to the transition between the “order” and “disorder” region near the boundary of SDW, which originate from thermal flocculation.

References

Resonant inelastic x-ray scattering study on (Li,Fe)OHFeSe

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The origin of superconductivity in Fe-based superconductors remains unclear. Although spin fluctuation plays an important role in Fe-based superconductors, other factors, such as orbital fluctuation, are also in play [1,2]. The intercalated iron selenide (Li,Fe)OHFeSe (FeSe11111) is a single-phase bulk superconductor, which can reach a high $T_c$ of 42K [3,4]. Intriguingly, it shows remarkably similar electronic behaviors as those in monolayer FeSe with the $T_c$ up to 65K [5], providing a bulk counterpart to explore the origin of high $T_c$ in iron selenides. Here we performed an extensive study on FeSe11111 by using resonant inelastic x-ray scattering (RIXS) at Fe L₃-edge. We have observed four Raman-like features at ∼ 0.1 eV, ∼ 0.3 eV, ∼ 0.7 eV, ∼ 2.5 eV, which are dispersionless versus momentum transfer. Moreover, these excitations show different temperature behaviors. Using atomic multiplet calculations we identify the excitations at ∼ 0.3 and 0.7 eV as d-d excitations involving spin and/or orbital degrees of freedom. Our results uncover the excitations in FeSe11111 and provide an important insight to understand the superconductivity in iron selenides.

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Plasmons and bond-charge excitations in layered $t$-$J$ model

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We employ the layered $t$-$J$ model with the long-range Coulomb interaction and study the charge excitation spectrum at leading order in a large-$N$ formalism. We find that the spectrum is characterized by a dual structure in energy space [1]. In the low-energy region typically less than the superexchange coupling $J$, various kinds of bond-charge excitations are dominant [1]. In particular, $d$-wave bond-charge excitations exhibit softening along the direction (0,0)-(π,0) [2], which explains the charge ordering tendency observed in Nd$_{2-x}$Ce$_x$CuO$_4$ [3]. The doping dependence of the $d$-wave bond-charge excitations [4] can also capture the experimental observations [5,6]. In the high-energy region typically larger than $J$, on the other hand, the usual on-site charge excitations become dominant and yield plasmons [1]. The plasmons exhibit a strong dependence on out-of-plane momentum $q_z$ as is well known in a layered system [7,8,9]. In particular, acoustic-like plasmons are realized for finite $q_z$ and show a V-shaped dispersion around in-plane momentum (0,0) with a gap proportional to interlayer hopping $t_z$ [9]. The acoustic-like plasmons well explain [10,11,12] the charge excitation spectra observed around (0,0) in various electron-doped systems Nd$_{2-x}$Ce$_x$CuO$_4$ [13,14,15,16], La$_{2-x}$Ce$_x$CuO$_4$ [16,17], Sr$_{1-x}$La$_x$CuO$_2$ [18], and hole-doped systems La$_{2-x}$(Sr,Ba)$_x$CuO$_4$ [12,19] and Bi$_2$Sr$_{2-x}$La$_x$CuO$_{6+δ}$ [12]. Furthermore, plasmons have a big impact on the electron self-energy and reduce the quasiparticle residue substantially [20].

References

Electronic structure of the frustrated diamond lattice magnet NiRh$_2$O$_4$

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NiRh$_2$O$_4$ is the first known realization of a spin-1 magnet on a diamond lattice and is predicted to host a variety of exotic phenomena such as topological paramagnetism [1], spiral spin liquid [2], quantum criticality [3], and excitonic magnetism [4], caused by frustrated nearest and next-nearest neighbor exchange, as well as orbital degeneracy. Thermodynamic measurements found no sign of magnetic ordering and inelastic neutron scattering found excitations suggestive of a valence bond solid ground state [5]. Theoretical works using both ab-initio and effective single-site models have explained these results by proposing a spin-orbital singlet ground state for NiRh$_2$O$_4$ [6]. Our recent RIXS measurements are mostly consistent with these predictions, but contain a feature that cannot be explained by a single-ion model with Coulomb interaction, crystal field, and spin-orbit coupling. Based on ab-initio calculations [6], we find that this feature is consistent with a nonlocal orbital excitation between Ni and Rh ions. Another feature suggests significant magnetoelastic coupling, common to orbitally degenerate A-site spinels [7]. These results provide insight into frustrated magnetism beyond fully localized moments in systems with coupled spin, orbital, and lattice degrees of freedom.

References
Disentangling spin and charge excitations and their evolution in the phase diagram of Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ superconducting cuprate

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The knowledge of the elementary excitations is vital for understanding the physics of superconducting cuprates. The spin excitations, conventionally studied by neutron scattering, has been long debated on whether they are governed by localized spins or itinerate electrons in doped cuprates where coherent quasiparticles emerge[1, 2]. Nowadays, with the development of the Resonant Inelastic X-ray Scattering (RIXS) techniques, RIXS has played an increasingly important role in completing our knowledge in the spin excitations of cuprates, especially in the high-energy range and momentum space away from $(\pi, \pi)$ where neutron scattering usually has limited cross sections. Intense spin excitations has been unveiled to persist to over-doped region in different systems [3, 4]. However, the controversy in localized or itinerate interpretations of the measured excitations continues. One obstacle is that the RIXS measured excitations in doped cuprates are usually of mixed charge and spin character, making the correct assignment of the spectral profile to individual excitations difficult [5, 6]. By utilizing the different geometry factors of spin-flip and non-spin-flip scattering process in RIXS, it has been shown that the spin and charge excitations in cuprates can be resolved by azimuthal dependent RIXS measurement [6]. Using this method, here we present the disentangled spectral profiles of the spin and charge excitations in superconducting Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ with momentum and doping developments. The resolved spectral profiles of spin excitations can be well fitted by a single damped harmonic oscillator, with larger damping factors along $(\pi, \pi)$ direction than $(\pi, 0)$. While the spin excitations show more intensities along $(\pi, \pi)$ direction than $(\pi, 0)$, the charge excitations behave in a reverse way, which highlights their distinct nature. Moreover, we find that spin spectral weights increase with increasing doping at intermediate $q$ along $(\pi, \pi)$ but decrease along $(\pi, 0)$ direction. We explain the experimental results by comparing them against the density matrix renormalization group (DMRG) calculations for a $t$-$J$—like model on a square lattice geometry. Ultimately we conclude that the observed persistence of the collective spin excitations is closely connected to the, intuitively expected, persistence of the short-range magnetic excitations in the doped cuprates.

References