# RIXS/REXS Workshop 2023
# 2nd–4th August 2023

**Sakura Hall**
**Tohoku University**

# Book of Abstracts

Institute for Advanced Synchrotron Light Source

QST National Institutes for Quantum Science and Technology

NanoTerasu

Research IMR Institute for Materials Research Tohoku University

IMRT GIMRT Global Institute for Materials Research Tohoku
-International hub for collaborations in materials science-

| Wednesday, August 2nd | |
|---|---|
| -9:05 | Registration |
| Chair: **Masaki Fujita** (IMR, Tohoku University) | |
| 9:05-9:20 | Opening Remarks<br>**Dr. Wataru Utsumi**, Director of ASLS, QST<br>**Prof. Takahiko Sasaki**, Director of IMR, Tohoku University |
| Session Chair: **Mark P. M. Dean** (Brookhaven National Laboratory) | |
| 9:20-10:05 | **Ke-Jin Zhou** (Diamond Light Source)<br>*Spin and charge excitations in quantum materials probed by RIXS* |
| 10:05-10:35 | **Johan Chang** (University of Zurich)<br>*Magnon-magnon Interactions in Soft Mott Insulators* |
| 10:35-11:05 | **Yingying Peng** (Peking University)<br>*Charge order and its dynamics in overdoped Cuprates* |
| 11:05-11:35 | Break |
| Session Chair: **Thorsten Schmitt** (Paul Scherrer Institut) | |
| 11:35-12:05 | **Di-Jing Huang** (National Synchrotron Radiation Research Center)<br>*Excitonic excitations of cuprate superconductors probed with high-resolution RIXS* |
| 12:05-12:25 | **Hiroyuki Yamase** (National Institute for Materials Science)<br>*Plasmon excitations and plasmarons in cuprate superconductors* |
| 12:25-13:30 | Lunch |
| 13:30-14:30 | Poster session |
| Session Chair: **Eugenio Paris** (Paul Scherrer Institute) | |
| 14:30-15:15 | **Urs Staub** (Paul Scherrer Institute)<br>*Resonant Scattering to Study Ultrafast Dynamics of Spins and Electrons and the Relation to the Dynamics of the Lattice* |
| 15:15-15:45 | **Tsuyoshi Kimura** (University of Tokyo)<br>*Circularly polarized resonant x-ray diffraction of multiferroic CuO* |
| 15:45-16:15 | **William Windsor** (Fritz Haber Institute of the Max Planck Society)<br>*Towards control of ultrafast spin dynamics in 4f antiferromagnets* |
| 16:15-16:45 | Break |
| Session Chair: **Atsushi Hariki** (Osaka Metropolitan University) | |
| 16:45-17:15 | **David Hawthorn** (University of Waterloo)<br>*Orbital/Nematic and Charge Density Wave Orders in Cuprate Superconductors Probed by Resonant Elastic X-ray Scattering* |
| 17:15-17:35 | **Hsiao-Yu Huang** (National Synchrotron Radiation Research Center)<br>*Quantum critical scaling of charge order in $La_{2-x}Sr_xCuO_4$ revealed by high-resolution RIXS* |
| 17:35:17:55 | **Mirian Garcia Fernandez** (Diamond Light Source)<br>*Low energy orbital excitations in A-site ordered $SmBaMn_2O_6$* |

| Thursday, August 3rd | |
|---|---|
| Session Chair: **Wei-Sheng Lee** (SLAC National Accelerator Laboratory) | |
| 9:00-9:45 | **Mark P. M. Dean** (Brookhaven National Laboratory)<br>*Electronic structure, magnetic interactions, and charge order in low valence nickelates probed by RIXS* |
| 9:45-10:15 | **Yao Wang** (Clemson University)<br>*Manipulation of Magnetic Excitations and Entanglement Enabled by trRIXS* |
| 10:15-10:45 | **Atsushi Hariki** (Osaka Metropolitan University)<br>*LDA+DMFT approach to resonant inelastic x-ray scattering in correlated materials* |
| 10:45-11:15 | Break |
| Session Chair: **Di-Jing Huang** (National Synchrotron Radiation Research Center) | |
| 11:15-11:45 | **Hakuto Suzuki** (Tohoku University)<br>*Distinct spin and orbital dynamics in $Sr_2RuO_4$* |
| 11:45-12:05 | **Naoya Iwahara** (Chiba University)<br>*Vibronic excitations in resonant inelastic x-ray scattering spectra of spin-orbit Mott insulators* |
| 12:05-12:25 | **Subhayan Roychoudhury** (Lawrence Berkeley National Laboratory)<br>*CleaRIXS: A fast and accurate first-principles method for the simulation and analysis of resonant inelastic x-ray scattering* |
| 12:25-13:30 | Lunch |
| 13:30-14:30 | Poster session |
| Session Chair: **William Windsor** (Fritz Haber Institute of the Max Planck Society) | |
| 14:30-15:15 | **Matteo Mitrano** (Harvard University)<br>*Towards a quantum x-ray spectroscopy of light-driven quantum materials* |
| 15:15-15:45 | **Giacomo Coslovich** (SLAC National Accelerator Laboratory)<br>*Ultrafast Resonant X-ray Scattering Reveals Light-enhanced Charge Density Wave Coherence in a High-$T_C$ Superconductor* |
| 15:45-16:15 | **Nicolas Jaouen** (Synchrotron SOLEIL)<br>*Magnetic chirality in multilayers probed by Soft x-ray (coherent) scattering* |
| 16:15-16:45 | Break |
| Session Chair: **David Hawthorn** (University of Waterloo) | |
| 16:45-17:15 | **Eugenio Paris** (Paul Scherrer Institute)<br>*Ultrafast Dynamics In Quantum Materials Using Time-resolved Resonant Inelastic X-ray Scattering at SwissFEL Furka* |
| 17:15-17:35 | **Kenji Tsutsui** (QST)<br>*Theoretical study of magnetic excitations in a photoexcited Mott insulator on a square lattice* |
| 17:35:17:55 | **Vivek Kumar Bhartiya** (Brookhaven National Laboratory)<br>*Spectroscopic signature of 3D magnetism in a van der Waals ferromagnet $Fe_5GeTe_2$* |
| 19:00-21:00 | Workshop Dinner |

| Friday, August 4th | |
|---|---|
| Session Chair: **Ke-Jin Zhou** (Diamond Light Source) | |
| 9:00-9:45 | **Jinghua Guo** (Lawrence Berkeley National Laboratory)<br>*Opportunities and challenges for the in-situ/operando soft X-ray spectroscopy in energy, catalysis and chemical science* |
| 9:45-10:15 | **Jonathan Pelliciari** (Brookhaven National Laboratory)<br>*Resonant Inelastic X-Ray Scattering to study thin films of quantum materials* |
| 10:15-10:45 | **Wei-Sheng Lee** (SLAC National Accelerator Laboratory)<br>*RIXS Studies on Charge Order Phenomena in Cuprate and Nickelate Superconductors* |
| 10:45-11:15 | Break |
| Session Chair: **Johan Chang** (University of Zurich) | |
| 11:15-11:45 | **Thorsten Schmitt** (Paul Scherrer Institut)<br>*Spin-excitation anisotropy in the nematic state of detwinned FeS* |
| 11:45-12:05 | **Wenliang Zhang** (Paul Scherrer Institut)<br>*Observation of the Flat Magnon Band in the Ferromagnetic Topological Metal $Fe_3Sn_2$ by Resonant Inelastic X-ray Scattering* |
| 12:05-12:25 | **Octave Duros** (Sorbonne Université)<br>*Expression and determination of the crystal electric field in the quantum spin-ice candidate $Yb_2Ti_2O_7$* |
| 12:25-13:50 | Lunch |
| Session Chair: **Hakuto Suzuki** (Tohoku University) | |
| 13:50-14:10 | **Yi Tseng** (Massachusetts Institute of Technology)<br>*Exciton delocalization mediated by charge-transfer dynamics in van der Waals semiconductors* |
| 14:10-14:30 | **Yuan Wei** (Paul Scherrer Institut)<br>*Spin-orbital excitations encoding the magnetic phase transition in the van der Waals antiferromagnet $FePS_3$* |
| 15:00-17:00 | NanoTerasu Tour<br>  |

# Oral Presentation
# Abstracts

# Spin and charge excitations in quantum materials probed by RIXS

Ke-Jin Zhou[a]

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Resonant inelastic X-ray scattering (RIXS) is a photon-in photon-out spectroscopic technique capable of probing collective excitations or ordered states, such as magnons, orbitons, plasmons, phonons, charge-density waves (CDW) in the energy-momentum space. Owing to its high cross-section and focused X-ray beam, RIXS is advantageous in measuring micron-size samples and nanometer-thick films. Over the last decade, RIXS has flourished because of an order of magnitude increase in its energy resolving power.

In the first part of my talk, I will present the results of spin excitations in a prototypical Haldane spin chain material $Y_2BaNiO_5$ [1] and a ferromagnetic Kagome semimetal $Co_3Sn_2S_2$ [2]. The studies establish that RIXS is capable of probing more complex spin excitations beyond simple dipolar spin wave. In the second part of my talk, I will present some of recent studies on unconventional cuprate and infinite-layer nickelate superconductors. In single-layer bismuth-based cuprates, we found anisotropic CDW peaks with elliptical shapes which is interpreted as evidence of unidirectional charge stripes hosted by mutually 90°-rotated anisotropic domains consistent with lanthanum- and yttrium- based cuprates [3]. For the newly discovered infinite-layer nickelates, plenty of spectroscopic results evidence that they resemble cuprates in some respects but also possess its distinct electronic structure. The dispersive spin excitations comparable to that in cuprates validate the existence of strong electron Coulomb interactions in particular the proximity between the strong antiferromagnetic correlations and superconductivity [4]. Besides spin excitations, CDW was discovered in thin films of non-superconducting $NdNiO_2$ [5-7]. This CDW exists in Ni and Nd orbitals demonstrating a multi-orbital character distinct to cuprates [5]. Most recently, abundant H is discovered in $Nd_{0.8}Sr_{0.2}NiO_2$ where superconducting dome appears in a narrow H doping window [8]. The results highlight the critical role of H for superconductivity in epitaxial infinite-layer nickelates.

**References**

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# Magnon-magnon Interactions in Soft Mott Insulators

Johan Chang[1*], Qisi Wang[2], Izabella Bialo[1], Leonardo Martinelli[1]

*[1]University of Zurich, Switzerland*
*[2]City University of Hong Kong, Hong Kong (SAR of China)*

In this talk, recent RIXS results on Mott insulating cuprate compounds will be presented. Analysis of magnon dispersions and magnon-magnon interactions will be the focus. We will discuss higher order magnetic exchange interactions and their renormalisations factors along with their implication for superconductivity in doped compounds.

# Charge order and its dynamics in overdoped Cuprates

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Charge orders (CO) are often considered a significant rival of high-temperature superconductivity in underdoped cuprates [1]. In contrast, overdoped cuprates have traditionally been viewed as conventional Fermi liquids without collective electronic order. However, our discovery of a reentrant CO in heavily overdoped Bi2201 beyond the pseudogap phase challenges this assumption [2]. Despite this, there is no consensus on whether CO is ubiquitous in heavily overdoped cuprates. Determining the extent of CO across different doping and temperature ranges can help disentangle the relationship between superconductivity, pseudogap, and CO phases, which is critical to understanding high-temperature superconductivity. Using Cu $L_3$ edge and O K edge resonant x-ray scattering, we have revealed the presence of CO in overdoped $La_{2-x}Sr_xCuO_4$ ($0.35 \leq x \leq 0.6$) beyond the superconducting dome. We observed similar in-plane momentum and polarization dependence and dispersive excitations as the CO of underdoped cuprates, but its maximum intensity differed along the c-direction and persisted up to 300 K. The Fermi surface instability cannot explain this CO and its origin remains to be understood. Our results suggest that CO is prevalent in the overdoped metallic regime, requiring a reassessment of the traditional understanding of overdoped cuprates as weakly correlated Fermi liquids.

In our recent study on heavily overdoped Bi2201, we also used time-resolved resonant X-ray scattering to examine CO dynamics [4,5]. We found that the charge order dynamics were distinct from those observed in underdoped cuprates when using 800 nm and 400 nm laser pumps, suggesting that the origins of CO in underdoped and overdoped cuprates may differ.

**References**

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# Excitonic excitations of cuprate superconductors probed with high-resolution RIXS

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Recent advancements in synchrotron radiation instrumentation have enabled high-resolution resonant inelastic X-ray scattering (RIXS) to become a powerful technique for probing elementary excitations with momentum resolution and providing direct information about the dynamics arising from fluctuations of spin, charge, and orbital degrees of freedom. This talk will begin with a review of the advances in high-resolution RIXS instrumentation [1], followed by discussions of recent examples of RIXS studies on cuprate superconductors [2, 3].

Electron quasiparticles play a crucial role in simplifying the description of many-body physics in solids with surprising success. However, conventional Landau's Fermi-liquid and quasiparticle theories for high-temperature superconducting cuprates have received skepticism from various angles. Whether the framework of electron fractionalization captures the essential physics of the pseudogap and superconducting phases of cuprates is still an open issue. We will show that excitonic excitation of optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ with energy far above the superconducting-gap energy scale, about 1 eV or higher, is unusually enhanced by the onset of superconductivity. Our findings prove the involvement of such high-energy excitons in superconductivity. Therefore, the observed enhancement in the spectral weight of excitons imposes a crucial constraint on theories for the pseudogap and superconducting mechanisms.

**References**

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# Plasmon excitations and plasmarons in cuprate superconductors

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Charge fluctuations in cuprates may be categorized into two: i) the charge ordering tendency around the momentum $\mathbf{q} \fallingdotseq (0.5\pi, 0)$ and $(0.6\pi, 0)$ in electron- and hole-doped cases, respectively, and ii) the V-shaped dispersion around the Brillouin zone center in both electron- and hole-doped cases. The former feature was interpreted as a *d*-wave bond-charge ordering tendency in electron-doped cuprates, but the origin is still controversial in hole-doped cuprates. The latter feature was discussed controversially at the early stage. The characteristic in-plane and out-of-plane momentum dependences and the observation of a charge gap at the zone center indicated consistently the acoustic-like plasmons specific to layered materials. In particular, a large-*N* theory of the layered *t*-*J* model with the long-range Coulomb interaction can explain the experimental data nearly quantitatively.

In this talk, we first present what kind of charge excitation spectra are predicted in the large-*N* theory of the *t*-*J* model—bond-charge excitations, acoustic-like plasmons, optical plasmons, and particle-hole excitations in momentum-energy space, and then clarify how well the acoustic-like plasmons explain the recent resonant inelastic x-ray scattering (RIXS) data [1]. The theory also explains consistently the optical plasmon observed by electron energy-loss spectroscopy (EELS) [2]. We next study how those charge fluctuations renormalize the one-particle electron property by computing the electron self-energy [3]. In contrast to an electron-phonon interaction and a coupling to magnetic fluctuations, charge fluctuations do not yield a kink in the electron dispersion. Instead, they yield the uniform suppression of the spectral weight on the entire Fermi surface and the quasi-particle weight is reduced down to 10-30 %, depending on parameters. The rest of the spectral weight is transferred to high energy regions, but in a very asymmetric way with respect to zero energy. This asymmetry comes from the strong correlation effect, not from the underlying band structure. In particular, an incoherent band is generated near the energy controlled by the optical plasmon energy. This incoherent band is the so-called replica band dubbed as plasmarons [4]. Recalling that plasmarons were discussed in weakly correlated systems such as graphene, two-dimensional electron systems, and $SrIrO_3$ films, it is intriguing that the same plasmarons are predicted in cuprates, too, where the strong correlation effect is definitely important. The predicted dispersion may be tested by collaborating with a community of angle-resolved photoemission spectroscopy.

**References**

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# Resonant Scattering to Study Ultrafast Dynamics of Spins and Electrons and the Relation to the Dynamics of the Lattice

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In my presentation, I will concentrate on investigations using transient resonant x-ray diffraction to study ultrafast dynamics of electronic and magnetic properties of materials. I will first discuss the case of ultrafast suppression of antiferromagnetic order in insulators, exemplified on different multiferroic materials such as CuO [1], $TbMnO_3$ [2,3] and hexaferrites [4]. These soft x-ray diffraction studies shown how a near infrared laser pulse demagnetize magnetic sublattices of the antiferromagnet. The observed timescales for the suppression of the antiferromagnetic order parameter is strongly material dependent and not yet fully understood. In a second step, I will show how THz pulses, short pulses of electric fields, can interact with the magnetic order leading to coherent (electro) magnon excitations. [5] This type of study is extended with very recent results from the free electron laser SwissFEL. A hard x-ray diffraction study using the BERNINA endstation allowed a direct visualization of the phononic contribution of an electromagnon. A resonant soft x-ray magnetic diffraction study from the brand-new FURKA endsation addressed the corresponding spin motion in real time. [6] The observed phase shift between the coherent resonant excitation of the lattice (phonon) and the spins (magnon) show that the E-field of the excitation drives the lattice first, which in turn changes the magnetic Hamiltonian that creates an effective magnetic field, which in turn drives the spins. In the last example, ultrafast hard x-ray resonant diffraction is used to study the response to a coherent excitation of a quasi-two-level orbital system in $Tb_2Ti_2O_7$. The excitation creates a quantum coherence entanglement of the states, which is observed through an induced spin rotation.

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# Circularly polarized resonant x-ray diffraction of multiferroic CuO

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CuO has attracted considerable attention since its multiferroic nature was discovered in 2008 [1]. This compound exhibits successive antiferromagnetic (AFM) phase transitions at $T_{N1}$ = 213 K and $T_{N2}$ = 230 K. At temperature ($T$) below $T_{N1}$, CuO shows a collinear AFM order with the commensurate propagation vector $\mathbf{q}_{CM}$ (0.5, 0, -0.5). At $T_{N1} \leq T \leq T_{N2}$, the Cu moments show a spiral spin order with the incommensurate propagation vector $\mathbf{q}_{ICM}$ (0.506, 0, -0.483). This spiral spin arrangement breaks inversion symmetry and induces improper ferroelectricity in which spin helicity is a primary order parameter [1]. Apart from the multiferroic nature in the ICM phase, the CM phase of CuO has been proposed to have an orbital current that is characterized by a time-odd polar vector corresponding to an anapole or toroidal moment [2].

In this study, we investigate resonant x-ray diffraction of CuO using circularly polarized x rays [3]. Circular dichroism on magnetic Bragg reflections in two distinct AFM phases (ICM spiral and CM collinear AFM phases) was observed at around the Cu $L_3$ edge. In the ICM phase where spin-spiral-induced ferroelectricity develops, it is reasonably considered that circular dichroism and its sign are ascribed to the spiral magnetic order and the sign of spin helicity (i.e., ferroelectricity), respectively. In the CM phase, weaker but substantial circular dichroic signals are observed. Measurements of spatial distributions of the circular dichroism revealed the presence of magnetic domains with rectilinear domain boundaries in both phases. It is found that the domain structures in the two phases resemble each other, suggesting some correlation between the ICM and the CM phases. Furthermore, a comparison of the magnetic domain structures with dark-field images obtained by electron diffraction measurements revealed that the domain boundaries observed in the two phases were strongly influenced by crystallographic twin structures. This study clarified the correlation between the crystallographic defect structure and the magnetic domain structures in antiferromagnetic CuO.

**References**

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# Towards control of ultrafast spin dynamics in 4f antiferromagnets

Y. W. Windsor[1], S-E. Lee[1], D. Zahn[1], V. Borisov[2], D. Thonig[3], K. Kliemt[4], A. Ernst[5,6], C. Schüßler-Langeheine[7], N. Pontius[7], U. Staub[8], C. Krellner[4], D. V. Vyalikh[9,10], O. Eriksson[2], L. Rettig[1]

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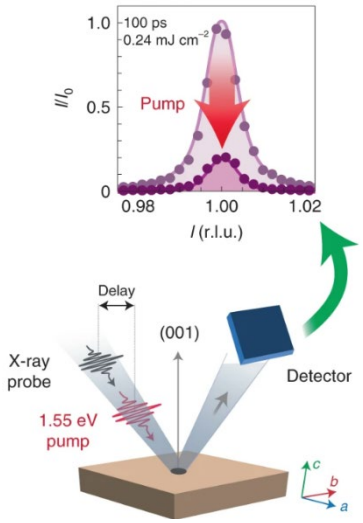
Ultrafast manipulation of magnetism bears great potential for future information technologies. While demagnetization in ferromagnets is governed by the dissipation of angular momentum, materials with multiple spin sublattices, for example antiferromagnets, can allow direct angular momentum transfer between opposing spins, promising faster functionality. Furthermore, a central prospect of antiferromagnetic spintronics is to exploit magnetic properties that are unavailable with ferromagnets. However, this poses the challenge of accessing such properties for readout and control.

Here I will present a series of ultrafast ("pump-probe") resonant soft X-ray diffraction studies that we have conducted, in which we address these prospects in Lanthanide-based antiferromagnets. 4f magnetism is usually mediated indirectly through the conduction electrons, in an effect known as the RKKY interaction. We have studied how tuning RKKY affects ultrafast magnetization dynamics in 4f antiferromagnets. This was done both by systematically varying the 4f occupation [1] and by altering the conduction electrons (thereby altering RKKY's magnitude). We find that the rate of direct angular momentum transfer between opposing moments is directly determined by this coupling. We have further studied light-induced manipulation of the transient ground state in one such material [2]. We trigger a coherent rotation of the entire long-range antiferromagnetic spin arrangement about a crystalline axis, thereby demonstrating deterministic control of this rotation. Our observations allow for a quantitative description of the transient magnetic anisotropy potential of the Lanthanide ions.




**References**

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# Orbital/Nematic and Charge Density Wave Orders in Cuprate Superconductors Probed by Resonant Elastic X-ray Scattering

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Charge density wave (CDW) order has been established as a generic feature of the cuprate phase diagram, competing and co-existing with superconductivity. While most commonly characterized by a Bragg peak at finite $\vec{Q}_{CDW}$ indictive of a translational symmetry breaking order parameter, CDW order can also couple to orbital degrees of freedom, evident by $Q_x = Q_y = 0$ orbital/nematic order. How these orders manifest and couple to each other can depend on the structure of the material and hole doping, and may play an important role in how CDW order varies amongst the different families of cuprates. In this talk, we explore the relationship between structural distortions, ($Q_x = Q_y = 0$) nematic/orbital order and CDW using equilibrium and pump-probe resonant x-ray scattering measurements in the$(La,M)_2CuO_4$ family of cuprate superconductors. These measurements demonstrate orbital/nematic order to be coupled to but distinct from CDW order. We will also show nematic order to be associated with the pseudogap phase of the cuprates, with signatures of nematic order present within the pseudogap phase, but vanishing above the pseudogap critical doping, $p^*$, and temperature, $T^*$ in overdoped cuprates.

**References**

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# Quantum critical scaling of charge order in $La_{2-x}Sr_xCuO_4$ revealed by high-resolution RIXS

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The pseudogap (PG) and strange-metal (SM) phases observed in cuprates above the superconductivity transition temperature are enigmatic. Quantum phase transitions, which are driven by nonthermal fluctuations at absolute zero temperature, play a crucial role in shaping the phase diagram of high-temperature cuprate superconductors. Understanding the PG and the SM phases might hinge on the influence of quantum criticality at finite temperatures. Recently, quantum fluctuations of charge-density waves (CDW) were observed in different families of cuprates [1-4]. In this study, we investigated the high-resolution resonant inelastic x-ray scattering (RIXS) of $La_{2-x}Sr_xCuO_4$ to unravel the quantum fluctuations of CDW in different doping. Our findings provide the spectroscopic signature of quantum critical scaling in cuprates, in which the PG and SM phases correspond to the renormalized classical and quantum critical scaling regimes, respectively. In addition, the characteristic energy $\Delta$ of the CDW fluctuations at zero temperature determines the crossover temperature $T^*$ between the PG and the SM phases, i.e., $T^* \sim \Delta$.

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# Low energy orbital excitations in A-site ordered $SmBaMn_2O_6$.

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In the last few decades plenty of experimental and theoretical research efforts have been devoted to perovskite manganites with chemical formula $R_{1-x}^{3+}A_x^{2+}MnO_3$ ($R^{3+}$= rare earth ions, $A^{2+}$=alkaline-earth ions). These materials show a rich variety of fascinating electromagnetic properties, such as the colossal magnetoresistance (CMR), charge and orbital ordering (COO), and metal-insulator transition. Recently it has been shown that an additional control parameter besides doping content and ionic radii of the A-site ions is the A-site randomness. Hence $LBaMn_2O_6$ compounds with $L$ = Sm, Eu, Gd, Tb, Dy, Ho and Y have attracted a lot of attention.

We present the combination of soft x-ray RIXS and REXS measurements performed in A-site ordered $SmBaMn_2O_6$. The elimination of the A-site randomness in these manganites allows for the orbital order to be long ranged. Evidence of this is that even on polycrystalline samples of this material, orbital order was observed by soft x-ray REXS[1-3]. High quality single crystals of this compound have only recently become available and have allowed for this RIXS study. This compound undergoes a two-step COO transition at temperatures $T_{co1}$ and $T_{co2}$ as the temperature decreases. In the second temperature transition $T_{co2}$, a disappearance of the four-fold periodicity along the c-axis has been suggested to be caused by the change on the stacking manner of the COO phases, however the models have not been uniquely determined due to lack of definitive experimental evidence[1,4-8]. In a single crystal of $SmBaMn_2O_6$ we measured the [1/4 1/4 0] orbital order reflection[1-3]. The energy dependence of this reflection shown in the insert of Figure 1 demonstrates its resonant behaviour. We collected RIXS data shown in Figure 1 at the OO geometry. Low energy excitations could be identified which may be compatible with the Jahn-Teller stretching and the breathing modes[9]. The temperature dependence of the RIXS spectra illustrate the evolution of low energy excitations across the phase diagram of the system. RIXS measurements collected as a function of energy and fixed q further demonstrate the dependence of the low energy excitations with the OO signal. Owing to the strong electron-phonon coupling in strongly correlated electron systems, it is possible to obtain a lot of information about the electronic properties and the phonon behaviour across the orbital ordering phase.

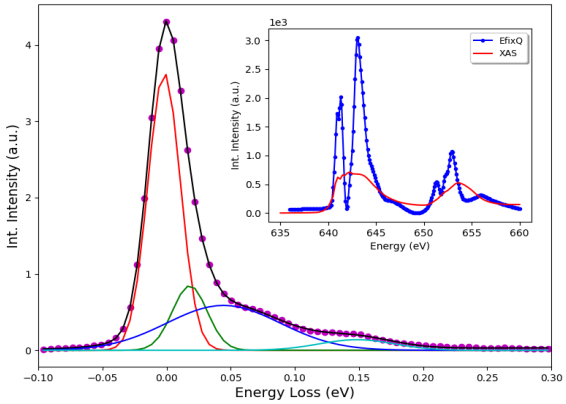


**Figure 1** RIXS spectra measured at the Mn L3 edge. The spectrum has been fitted with 4 gaussians. In the insert is shown in blue the energy dependence of the orbital order [1/4 1/4 0] reflection and in red the XAS, both measured in the vicinity of the Mn.

This is the first combined study of RIXS and REXS in a 3d half doped manganite with orbital ordering.

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# Electronic structure, magnetic interactions, and charge order in low valence nickelates probed by RIXS

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After a 30-year quest, researchers recently succeeded in realizing superconductivity in low valence nickelates [1]. This ignited a vigorous debate regarding the essential electronic properties of these materials and their similarity to cuprates. Some important questions include: Do these materials have appreciable oxygen charge-transfer character and superexchange akin to the cuprates or are they in a distinct Mott-Hubbard regime where oxygen plays a minimal role and superexchange is negligible? Given that cuprates have a propensity to host proximate competing phases such as charge and spin order, one might ask whether the nickelate's phase diagrams also host competing orders and to what extent they are similar to those in cuprates? In this talk, I will give a perspective of where we are with these questions, including our studies of the trilayer low valance nickelate $La_4Ni_3O_8$ [2-5].

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# Manipulation of Magnetic Excitations and Entanglement Enabled by trRIXS

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The rapidly evolving quantum science calls for precise and predictive control of collective electronic properties beyond the classical realm. Among various control knobs, an ultrafast laser pump is a promising approach due to its rich degrees of freedom. Together with the laser's capability of influencing electronic structure comes the necessity to track the instantaneous status of a time-dependent nonequilibrium material. In this talk, I will discuss the application of time-resolved resonant inelastic x-ray scattering (trRIXS) in magnetic materials. By comparing the trRIXS for antiferromagnetic materials, we find that the instantaneous paramagnon excitations can be manipulated by pulsed laser in a predictive manner, following the Floquet theory in the center of the pulse. Such a Floquet engineering works only at doped Mott insulators without long-range order, while the trRIXS study of an undoped Mott insulator violates the Floquet approximation. Leveraging the light-engineered magnetic excitations, we further study the nonequilibrium dynamics of a 1D cuprate chain. Through a self-consistent iteration, trRIXS can probe the transient entanglement of wavefunctions in nonequilibrium materials via the quantum Fisher information. Via this approach, we reveal the possibility of enhancing entanglement in a cuprate chain using an ultrafast laser pulse.

# LDA+DMFT approach to resonant inelastic x-ray scattering in correlated materials

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We present an *ab*-initio method for simulating resonant inelastic x-ray scattering (RIXS) spectra in strongly correlated 3*d* transition-metal and 4*f* rare-earth materials. The method builds on a combination of density functional theory (DFT) and dynamical mean-field theory (DMFT). This method includes both local excitations (e.g. atomic multiplet excitation) and continuum of unbound electron-hole pairs as well as charge-transfer excitations in a unified way and material specific manner [1,2]. The heart of this computational method is a configuration-interaction impurity solver that allows to compute RIXS intensities efficiently from a multi-orbital Anderson impurity model with the DMFT hybridization densities and core orbitals including full core-valence multiplet interaction explicitly in the intermediate states of RIXS.

In this talk, we present recent applications of the LDA+DMFT method for correlated materials: *L*-edge RIXS in NiO (canonical Mott insulator), $LuNiO_3$ (site-selective Mott insulator) [3], infinite-layer nickelate [4], and *M*-edge RIXS for $CePd_3$ (prototype 4*f* Kondo lattice system) [5]. With these examples, we demonstrate the capability of RIXS to advanced understanding of the dynamics and interplay of the spin, orbital and charge degrees of freedom in correlated materials.

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# Distinct spin and orbital dynamics in $Sr_2RuO_4$

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The unconventional superconductor $Sr_2RuO_4$ has long served as a benchmark for theories of correlated electron materials. Recent precision experiments in the superconducting state have cast doubt on the previously advocated spin-triplet pairing scenario, thus revitalizing the order parameter debate and the search for an in-depth understanding of the Fermi-liquid normal state and collective bosonic fluctuations relevant to superconductivity. However, the lack of information on high-energy magnetic excitations from neutron scattering experiments precludes the understanding of the global picture of the spin fluctuations and the role of orbital degrees of freedom. This hinders an experimental test of different theoretical frameworks to the dynamical properties of $Sr_2RuO_4$.

To collect comprehensive information on the spin and orbital fluctuations of $Sr_2RuO_4$, we have performed resonant inelastic x-ray scattering (RIXS) at the Ru $L_3$ edge (2838 eV) [1]. The experiment was performed using the newly-developed IRIXS spectrometer [2] at the P01 Beamline of PETRA III, DESY.

We have observed multiple branches of dispersive spin and orbital excitations from the Fermi liquid state of $Sr_2RuO_4$, including the well-known incommensurate spin fluctuations. Theoretical spin and orbital dynamical response functions calculated within the dynamical mean-field theory excellently capture the key features of the RIXS spectra, whereas perturbative random phase approximation fails to reproduce the experimental observations. By establishing the properties of key collective modes, our results provide a solid baseline for the future identification of the nature and symmetry of superconducting order in this material.

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# Vibronic excitations in resonant inelastic x-ray scattering spectra of spin-orbit Mott insulators

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Spin-orbit Mott insulators with heavy 4d/5d transition metal ions attract significant increasing attention because they could exhibit novel phases such as Kitaev spin liquie phase, excitonic magnetic phase, and multipolar phases [1]. To characterize the nature of the 4d/5d transition metal compounds, precise knowledge of the elementary excitations is desired. To this end, resonant inelastic x-ray scattering (RIXS) has been an indispensable technique to probe complex elementary excitations. The local quantum states of these compounds have been analyzed based on the interplay of ligand-field and spin-orbit coupling. However, the conventional analyses of recent RIXS spectra of cubic double perovskites fail to provide reasonable physical pictures. In the case of the RIXS spectra of $K_2RuCl_6$ (Figure), the spin-orbit based analysis suggested a large reduction of the spin-orbit coupling by 50 % compared with the value extracted from magnetic susceptibility measurements of the same compound [2, 3]. In the case of $K_2IrCl_6$, splitting of a peak in the RIXS spectra was interpreted as ligand-field splitting, which is contradictory to the neutron diffraction data showing no symmetry lowering down to 0.3 K [4]. As an origin of such puzzling situations, presence of dynamic Jahn-Teller (JT) effect in the excited states has been mentioned, whereas the presence and its impact on the RIXS spectra are unknown.

In this presentation, we address two issues: (i) the presence of the dynamic JT effect in the cubic spin-orbit Mott insulators, and (ii) the impact of the dynamic JT effect on the RIXS spectra. Taking $5d^5$ ion ($Ir^{4+}$) as an example, we simulated the RIXS spectra of a single ion with dynamic JT effect in the excited spin-orbit multiplet states [5]. The simulated RIXS spectra show a clear splitting of the RIXS spectra due to vibronic excitations (Figure). Increasing temperature, we could also reproduce the temperature evolution of the RIXS spectra of the Ir compound. Based on the first-principles calculations, we confirmed the validity of the chosen interaction parameters for my simulations. Similarly, we could explain the shape and temperature evolution of the RIXS spectra of $Ru^{4+}$ ($4d^4$) ion by fully considering the dynamic JT effect [6]. Our work established the emergence and the fingerprints of the dynamic JT effect in RIXS spectra of spin-orbit Mott insulators.

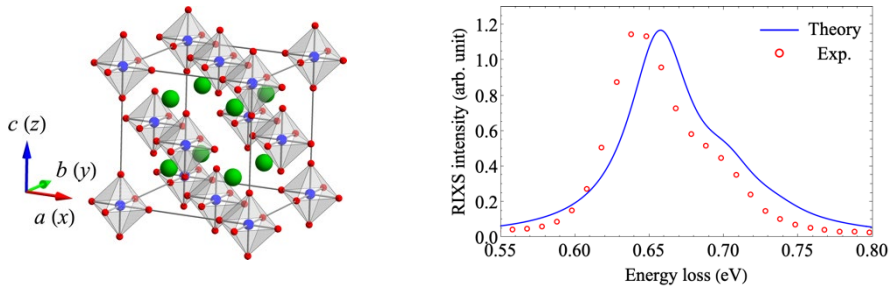


Figure. Left: $K_2MCl_6$ (M = Ru, Ir). Right: RIXS spectra of $5d^5$ ion including the vibronic excitations.

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# CleaRIXS: A fast and accurate first-principles method for the simulation and analysis of resonant inelastic x-ray scattering

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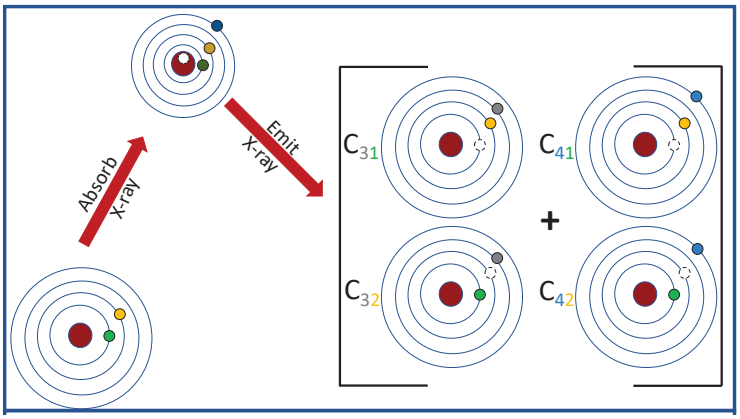
The complexity of the physical phenomena underlying resonant x-ray scattering typically renders the simulation and interpretation of the spectra difficult and expensive. By combining two diverse and established approaches to modeling electronic excited states (namely the response-based approach and the constrained-occupation approach), in this presentation I will introduce an *ab initio*, accurate, and efficient computational framework for the simulation and analysis of RIXS spectra for electronic transitions. The core-hole linear-response RIXS (CleaRIXS) [1] method not only ensures accurate incorporation of the interaction of electrons with core and valence holes, but also automatically maps the salient RIXS features to the relevant electronic transitions. Through comparison with previous methanol C K-edge RIXS measurements, we show the efficacy of the formalism in modeling different regions of the RIXS spectrum and in gaining physical insight about their origins. The importance of including the electron-hole interactions outside the core region is explored, in addition to the connection between CleaRIXS and the determinant-based methods for simulating x-ray absorption and nonresonant x-ray emission. CleaRIXS provides a robust and extendable framework for prediction and interpretation of RIXS processes and for the simulation of complex electronic excited states in general.

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# Towards a quantum x-ray spectroscopy of light-driven quantum materials

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Optically excited quantum materials exhibit striking emergent phenomena ranging from light-controlled magnetism and ferroelectricity to Floquet topological states and light-induced superconductivity. Central to the appearance of these nonequilibrium phenomena is a variety of excitation mechanisms involving intertwined electronic, structural, magnetic, and orbital degrees of freedom and their interaction with ultrafast laser fields. However, understanding how these ingredients determine the nonequilibrium behavior at the macroscale requires microscopic knowledge of many-body interactions and wavefunctions of the excited state.

By using the light-induced renormalization of the effective electronic interactions as an example, I will show how time-resolved x-ray absorption spectroscopy (trXAS) and resonant inelastic x-ray scattering (trRIXS) provide a unique view of the transient electronic structure of light-driven quantum materials [1]. Further, I will discuss how polarization-resolved trXAS and trRIXS are able to map the effective interactions of two cuprate superconductors with distinct effective dimensionality [2,3]. I will then discuss constraints about structure of the driven state wavefunction and implications of our findings for $\eta$-pairing and other driven, long-range-ordered phases.

In addition to these advances, I will introduce a novel approach aimed to witness entanglement in the excited state [4,5]. The key idea is to extract quantum metrological operators, such as the quantum Fisher information, together with operator-specific quantum bounds to determine the presence of multipartite entanglement of the driven state wavefunction from the trRIXS signal. This approach, along with a comprehensive mapping of the excited state using trRIXS and trXAS, represents a significant step towards a quantum x-ray spectroscopy of light-driven quantum materials, and will enable novel routes to harness their unique functionalities.

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# Ultrafast Resonant X-ray Scattering Reveals Light-enhanced Charge Density Wave Coherence in a High-$T_C$ Superconductor

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The use of ultrashort X-ray pulses offers new opportunities to study fundamental interactions in materials exhibiting unconventional quantum states, such as stripes, charge density waves and high-temperature superconductivity. To understand the microscopic interdependence between these order parameters, a probe capable of discerning their interaction on its natural length and time scales is necessary. In this talk, I will focus on recent ultrafast resonant soft x-ray scattering results tracking the transient evolution of charge density wave correlations in the high-$T_C$ superconductor $YBa_2Cu_3O_{6+x}$ [1]. In this study, ultrashort infrared pulses produce a non-thermal quench of the superconducting state while X-ray pulses detect the reaction of charge density waves. The response happens on a picosecond timescale and is characterized by a large enhancement of spatial coherence of charge density waves, nearly doubling their correlation length, accompanied by a smaller increase of their amplitude. Such behavior can be reversed at higher fluences and at higher temperatures (above $T_C$, 65 K) where the photo-induced charge density wave melting process takes place instead.
This ultrafast snapshot directly reveals the coupling between superconductivity and charge density waves on its natural timescale. It demonstrates that their competition manifests inhomogeneously at the nanoscale level, as disruption of spatial coherence, indicating the role of superconductivity in stabilizing topological defects within charge density waves domains. The study highlights a path for enhancing spatial coherence (order) by using light pulses by changing the balance between intertwined orders and it represents a unique opportunity for this growing technique in resonant X-ray scattering. In conclusion, I will also discuss ongoing developments at the LCLS to further expand on these results and unlock novel capabilities to control and observe quantum materials with light pulses.

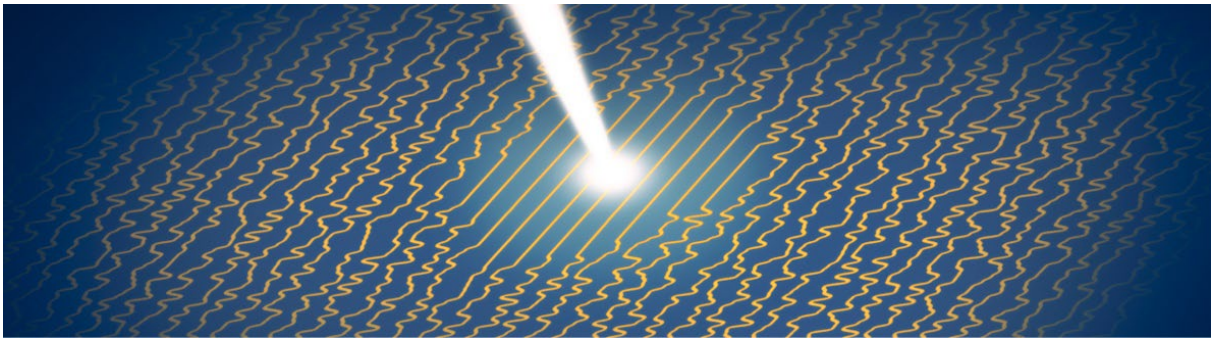

*Image credit: Greg Stewart, SLAC*

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# Magnetic chirality in multilayers probed by Soft x-ray (coherent) scattering

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Many current forthcoming applications of magnetic materials involve heterostructures or alloys containing magnetic and non-magnetic elements. X-ray Resonant (Coherent) Scattering is the technique of choice to probe such phenomena thanks to its element-selectivity and spatial sensitivity.

In this presentation I will introduce the experimental set-up that we developed at SOLEIL and illustrate their capabilities. SEXTANTS [1] is a beamline of the SOLEIL synchrotron, covering the 50-1700eV energy range dedicated to soft x-ray scattering. The resolving power exceeds 10000 and maximum flux on the sample ranges from $1\times10^{14}$ (100 eV) to $2\times10^{13}$ (1000 eV) ph./s/0.1% bw. The beamline main objective is the investigation of the electronic and magnetic properties of solids using three scattering techniques: resonant inelastic x-ray scattering (RIXS), x-ray resonant magnetic scattering (XRMS) and coherent x-ray scattering (CXS), the last one including also imaging via Fourier transform holography (FTH) [2, 3].

In the second part, several recent results obtained at SEXTANTS beamline will be presented, and in particular I will show that XRMS is the tool to study magnetic chirality in prototype Pt/Co multilayers in which Dzyaloshinskii-Morya interaction (DMI) is induced by the inversion symmetry breaking at the interfaces [3] and how it was possible to reveal the hybrid character of the magnetic structure in thick multilayer [4]. I will also present how one can manipulate this magnetic chirality either statically using magnetic field or at the femtosecond timescale using optical light [5].

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 orders of magnitude in the planned upgrade sources, allow nowadays to image the sample with a spatial resolution of few tens of nanometers and a time resolution ranging from ns down to few ps timescale.

If your abstract contains figures and pictures, make sure they are included in the word document.

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# Ultrafast Dynamics In Quantum Materials Using Time-resolved Resonant Inelastic X-ray Scattering at SwissFEL Furka

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In the realm of strongly correlated materials with entangled degrees of freedom, accessing the ultrafast dynamics involving different energy scales in a single experiment, with momentum resolution, promises excellent scientific insight. Time-resolved Resonant Inelastic X-ray Scattering (tr-RIXS) emerges as a unique technique capable of unraveling such information. However, the full realization of its potential has so far been limited by the available time- and/or energy resolution.

The soft-x-ray experimental endstation for condensed matter of SwissFEL - named Furka - aims to provide state-of-the-art time-resolved RIXS/REXS experiments in combination with a widely tunable pump laser radiation. Following the successful completion of the installation phase, Furka will commence its user program in 2024.

In this presentation, I will provide an update on the status of the endstation and present results achieved in the early commissioning phase.

# Theoretical study of magnetic excitations in a photoexcited Mott insulator on a square lattice

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Time-resolved *L*-edge resonant inelastic x-ray scattering (trRIXS) and time-resolved two-magnon Raman scattering (trTMR) have been developed for investigating transient magnetic dynamics on strongly correlated systems [1-3]. The photoexcitations of two-dimensional antiferromagnetic Mott insulators have exhibited interesting phenomena such as weakened intensity of single- and two-magnon excitations as observed in these pump-probe experiments.

In this study, we theoretically investigate magnetic dynamics in a photoexcited half-filled Hubbard model on the square lattice [4]. We adopt a numerically exact-diagonalization technique on the finite-size square lattice to obtain time-resolved spin and two-magnon excitation spectra after a pump pulse. We also examine momentum dependent two-magnon excitations after a pump pulse, which can be observed in trRIXS. We find that new magnetic signals clearly emerge well below magnon energy in both single- and two-magnon excitations when a pump pulse is tuned for an absorption edge.

Analyzing the wavefunction after a pump pulse in terms of the point group symmetry for the square lattice, we find that photoinduced low-energy signals are predominantly created by a photoexcited state with the *E* presentation of the $C_{4v}$ point group. Moreover, we find that an excitonic state of a holon-doublon pair at the absorption edge plays an important role in enhancing the intensity of these excitations. We expect that such characteristic pumping-frequency dependence will be observed in trRIXS and trTMR experiments.

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# Spectroscopic signature of 3D magnetism in a van der Waals ferromagnet $Fe_5GeTe_2$

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The recently discovered family of van der Waals itinerant ferromagnets $Fe_{5-x}GeTe_2$ ($0<x<2$) are strongly correlated electronic systems and realize intriguing ground states i.e. heavy fermions and charge density wave [1-3]. Moreover, the *cleavable* $Fe_5GeTe_2$ displays the highest $T_c \sim 315$ K and has the potential for miniaturized spintronic devices operating at room temperature [4-5]. However, it is not yet known if magnons in $Fe_5GeTe_2$ are confined in 2D or propagate in 3D. To date, investigation of magnons via inelastic neutron scattering is not feasible due to the small size of the available single crystals. In this study, we overcome this issue by employing a high-resolution resonant inelastic X-ray scattering (RIXS) on a mm-size single crystal of $Fe_5GeTe_2$ [5]. A broad magnetic continuum stretching up to 150 meV and displaying a strong intensity modulation along (0 0 *L*) is observed [6]. We show that this intensity modulation is compatible with a dominant inter-slab magnetic interaction enabling 3D propagation of magnons in the room-temperature ferromagnet $Fe_5GeTe_2$ [6].

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# Opportunities and challenges for the in-situ/operando soft X-ray spectroscopy in energy, catalysis and chemical science

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The energy materials and devices have been largely limited in a framework of thermodynamic and kinetic concepts or atomic and nanoscale. Synchrotron based X-ray spectroscopic techniques offers unique characterization in many important energy materials of energy conversion, energy storage and catalysis in regards to the functionality, complexity of material architecture, chemistry and interactions among constituents within. However, it is challenging to reveal the real mechanism of the chemical processes. In the operando soft X-ray spectroscopy characterization of interfacial phenomena in energy materials and devices, it has been found that the microstructure and composition of materials as well as the microstructure evolution process have a great influence on performances in a variety of fields, e.g., energy conversion and energy storage materials, chemical and catalytic processes.

This presentation will show how to best use the in-situ/operando soft X-ray spectroscopy characterization techniques in the last two decades, including soft X-ray absorption spectroscopy (XAS) and resonant inelastic soft X-ray scattering (RIXS) to investigate the real electrochemical mechanism during the operation. The experimental results show how in-situ/operando soft X-ray spectra characterization techniques uncover the phase conversion, chemical and environmental change of elements and other very important information of solid/gas and solid/liquid interfaces in real time, thus further enhance the understanding of real reaction mechanism. Some of the instrumentation development will be presented in regards to the high throughput RIXS spectrometer on BL8.0.1 and newly built RIXS beamline with a combination of in-situ/operando high-resolution and high-throughput endstations on AMBER.

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# Resonant Inelastic X-Ray Scattering to study thin films of 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 start by presenting of the current performance of the SIX beamline at NSLS-II and move forward with our recent RIXS investigations in the field of thin films [1]. I will focus on two different cases, the effect of confinement on the spin excitations of metallic iron [1], and the effect of capping layer on superconducting infinite layer nickelates.

In the case of metallic Fe, the decrease of thickness affects anisotropically the three-dimensional spin excitations of the bulk ferromagnet. In bulk-like films we corroborate the ability of RIXS to detect ferromagnetic spin excitations and I will show a comparison with inelastic neutron scattering. As the thickness is reduced, the out-of-plane spin excitations are gradually softened whereas the in-plane spin excitations persist and retain their dispersion. Using several considerations, we can rule out phenomena such as strain and substrate charge transfer and ascribe this evolution to confinement. Theoretically this trend can be explained by conventional Heisenberg models accounting for the number of broken magnetic bonds in proximity of the interface.

In second part of my talk, I will present the role of the capping layer for the excitations in parent $NdNiO_2$ and optimal-doped $Nd_{1-x}Sr_xNiO_2$. In uncapped $NdNiO_2$ compound we observe damped spin excitations resembling the one observed in cuprates [2]. The intensity of the spin excitations is however strongly decreased compared to the capped compound indicating a renormalization of magnetism. In superconducting materials, independently of the presence of capping, we see an overall softening of the spin excitations compared to the parent compound. Overall, the spin excitations spectrum is suppressed in uncapped samples indicating a strong influence of the capping layer on the magnetism in infinite layer nickelates regardless of doping. Remarkably, the spectral weight of the Ni-Nd charge transfer is also highly suppressed. Finally, I will compare this experimental evidence in relation to previous calculations accounting for the presence or lack of the capping layer.

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# RIXS Studies on Charge Order Phenomena in Cuprate and Nickelate Superconductors

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The role of charge order (CO) in unconventional superconductors is an intriguing and extensively studied research topic. Further insight could be gained by simultaneously tracking the CO order parameter and its excitations in the phase diagram. In this talk, I will highlight our recent high resolution resonant inelastic x-ray scattering (RIXS) studies on the CO phenomena in double-layered cuprates $Bi_2Sr_2CaCu_2O_{8+d}$, where signatures of CO excitations can be identified in the RIXS phonon cross-section [1]. Interestingly, anomalous enhancements of the CO excitations deep inside the superconducting state were observed near optimal doping, despite the suppression of the CO order parameter [2]. By tracking this anomalous temperature dependence throughout the phase diagram, we identify a characteristic doping concentration of the CO phenomena [3] and discuss its implications on putative quantum critical points. If time permits, I will also talk about our recent discovery of CO in infinite-layer nickelate superconductors [4] and discuss the similarities and differences to the CO in cuprates.

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# Spin-excitation anisotropy in the nematic state of detwinned FeS

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The origin of the electronic nematicity, which is characterized by rotational symmetry breaking, in FeSe is one of the most important unresolved puzzles in the study of iron-based superconductors. In both spin- and orbital-nematic models, the intrinsic magnetic excitations at $\mathbf{Q_1}$ = (1, 0) and $\mathbf{Q_2}$ = (0, 1) of twin-free FeSe are expected to provide decisive criteria for clarifying this issue. Although a spin-fluctuation anisotropy below 10 meV between $\mathbf{Q_1}$ and $\mathbf{Q_2}$ has been observed by inelastic neutron scattering around $T_c \sim 9$ K ($<< T_s \sim 90$ K), it remains unclear whether such an anisotropy also persists at higher energies and associates with the nematic transition $T_s$. Here we use resonant inelastic x-ray scattering at the Fe $L_3$-edge to probe the high-energy magnetic excitations of uniaxial-strain detwinned FeSe [1]. A prominent anisotropy between the magnetic excitations along the H and K directions is found to persist to ~ 200 meV. This anisotropy decreases gradually with increasing temperature and finally vanishes at a temperature around $T_s$. Our results reveal an unprecedented strong spin excitation anisotropy with a large energy scale well above the $d_{xz}/d_{yz}$ orbital splitting, suggesting that the nematic phase transition is primarily spin-driven [1].

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# Observation of the Flat Magnon Band in the Ferromagnetic Topological Metal $Fe_3Sn_2$ by Resonant Inelastic X-ray Scattering

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Magnetic metals with kagome structure can host various topologically non-trivial spin or electronic states, providing an extraordinary platform for studying both the fundamental topological physics and potential technological applications. The metallic kagome ferromagnet $Fe_3Sn_2$, which has large spin-orbital coupling, shows considerable interplay between magnetism and non-trivial electronic states[1, 2]. The material has a host of anomalous bulk properties, including a first-order spin reorientation transition[3], a large anomalous Hall effect[4], skyrmionic bubbles[5], and field tunable nematic states[1]. Here, by applying circular polarized X-rays in resonant inelastic X-ray scattering (RIXS), we show that in ferromagnets excitations of different spin/orbital character present distinct magnetic circular dichroism (MCD) and can be selectively probed. Using the MCD RIXS and XAS (X-ray absorption spectroscopy), we report the unambiguous isolation of magnetic signals of a flat spin wave band with large (compared to elemental iron) orbital moments in $Fe_3Sn_2$. The flat mode energy is consistent with the high Curie temperature ($T_C \approx 640$ K) as well as the strong acoustic mode dispersion, implying, together with the substantial spin-orbit coupling indicated by the large orbital moment, that the mode is topological[6]. The measured properties of the spin waves are highly unconventional and include very severe damping as well as a flat mode amplitude, which is maximized in the long wavelength limit where it is ordinarily expected to vanish. Our results illustrate how the magnetic circular dichroism analysis enhances the sensitivity of RIXS to magnetic excitations in ferromagnetic materials and unveil an unconventional flat spin wave band in a kagome ferromagnet.

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# Expression and determination of the crystal electric field in the quantum spin-ice candidate $Yb_2Ti_2O_7$

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Rare-earth titanate pyrochlores are defined by the general chemical formula $R_2Ti_2O_7$ (with $R$, a rare-earth metal) and a distinctive crystalline structure, made of two interpenetrating tetrahedral networks. One of the most interesting aspects of the $R_2Ti_2O_7$ compounds is the presence of fascinating magnetic phases, such as spin-liquid or spin-ice states [1, 2]. The key ingredient for understanding such behaviors is the characterization of the crystal electric field (CEF) acting on the $R$ sites. Here, the CEF is created by the surrounding oxygen ligands and interacts with the rare-earths $4f$ orbitals. Considering the $D_{3d}$ symmetry of the rare earths in $R_2Ti_2O_7$ crystals, the CEF can be expressed as a function of six parameters $B_q^k$ in the Wybourne convention [3]: $\mathrm{H}_{\mathrm{CEF}}^{D_{3d}} = B_0^2\mathbf{C}_0^2 + B_0^4\mathbf{C}_0^4 + B_3^4(\mathbf{C}_{-3}^4 - \mathbf{C}_3^4) + B_0^6\mathbf{C}_0^6 + B_3^6(\mathbf{C}_{-3}^6 - \mathbf{C}_3^6) + B_6^6(\mathbf{C}_{-6}^6 + \mathbf{C}_6^6)$, with $\mathbf{C}_q^k$ the Racah tensor operators. In the more particular case of $Yb_2Ti_2O_7$, a coexistence of ferromagnetic exchange interactions with quantum fluctuations was observed, which prevents any long-range ordering, leading to a so-called quantum spin-ice state [4]. Among the pyrochlore family this compound is one of the most intensively studied overtime, in particular using powerful inelastic neutrons scattering (INS) experiments [5, 6] that reach meV resolution. However, in practice, the determination of CEF parameters is complicated by both the variety of CEF definitions available in the literature and the fact that INS only probes the groundstate atomic manifold. Here, we propose an unified approach based on RIXS to complement INS data and pin down the 6 parameters involved in the CEF expansion. Our strategy is motivated by the remarkable RIXS capabilities: the possibility to probe manifolds situated well above the groundstate and the resonant behavior of *f-f* intensities. We recorded resonant spectra at the Yb $N_{4,5}$ edge by using the AERHA spectrometer [7] at SOLEIL and undertook RIXS calculations in QUANTY within the ligand-field multiplets approach [8] (see Fig. 1) to understand how the CEF affects the *f-f* excitations.

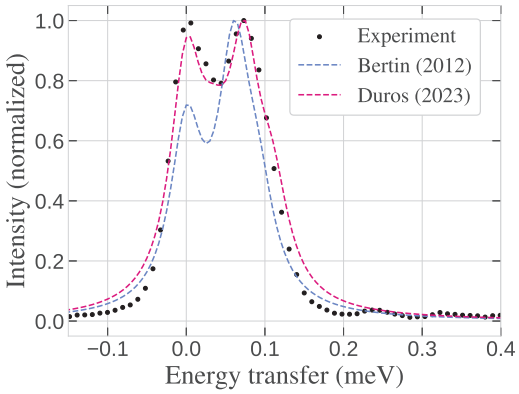


Figure 1: RIXS measured (black points) and calculated (dashed lines) spectra of the *f-f* excitations emphasizing the CEF effect in $Yb_2Ti_2O_7$. Calculations were performed in the experimental conditions with two different sets of $B_q^k$ parameters.

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# Exciton delocalization mediated by charge-transfer dynamics in van der Waals semiconductors

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Excitons provide unique opportunities to understand low-energy dynamics in condensed matter, and are particularly of recent interest in magnetic van der Waals (vdW) semiconductors. At this front, signature of ultrasharp optical excitons have been reported in magnetically-ordered vdW materials hosting $Ni^{2+}$ ions with small charge transfer gap, but a comprehensive understanding of their microscopic nature and connection to magnetic order is still developing. Towards this goal, the momentum dispersion of exciton quasiparticles has been shown to encode crucial insights into their inter-site propagation that governs exciton properties. While this was previously demonstrated in organic semiconductors and alkali-metal halides, such understanding is currently lacking for inorganic vdW semiconductors. Here, we study the ultrasharp spin-flip excitations in the vdW triangular-lattice antiferromagnets $NiX_2$ (X = Cl, Br, I) using Ni $L_3$-edge resonant inelastic X-ray scattering (RIXS). Varying the halogen allows systematically tuning the charge-transfer energy and the magnetic ground state, to examine their roles in the exciton properties. We reveal ultrasharp spin-flip excitations that exhibit nature of multiplets in a $3d^8$ electronic configuration with octahedral ligand coordination. These modes are independent of the disparate magnetic ground states and transition temperatures, as well as their respective spin excitation dispersions. Notably, the observed spin-flip multiplets exhibit momentum-dispersion with a ligand-dependent bandwidth based on a universal crystallographic periodicity, suggesting a charge-transfer exciton character beyond the prototypical on-site inter-orbital dd transitions. From ligand-field and tight-binding modelling, we determine an electronic dispersion driven by ligand-mediated third-nearest-neighbor hopping processes on the 2D triangular lattice. Our momentum-resolved RIXS results broadly establish the nature of spin-flip excitons unlimited to the optical dipole transitions, as well as their flexible chemical tuning and design of delocalization pathways in magnetic vdW materials.

# Spin-orbital excitations encoding the magnetic phase transition in the van der Waals antiferromagnet $FePS_3$

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Magnetic van der Waals (vdW) materials have provided exciting new opportunities in the studies of functional exotic magnetic phases of various symmetry-breaking groups. [1] The interlayer exchange interaction in these materials is very small, which allows for the decoupling of the magnetic layers and makes it an ideal candidate for studying the effects of dimensionality and interlayer coupling on magnetic behavior. [1] Recent studies utilizing primarily optical spectroscopy have demonstrated the sensitivity of the phonon spectral response to the magnetic state down to the few-layer limit. [2] This has opened up an opportunity for understanding and investigating the magnetic properties of these materials at the nanoscale.

$FePS_3$ is one such vdW material that has garnered significant interest due to its unique magnetic and electronic properties. It is an S = 2 zig-zag quasi-two-dimensional antiferromagnetic insulator with a honeycomb lattice. [3] The electronic structure of $FePS_3$ has been resolved by various techniques. X-ray absorption spectroscopy (XAS) has provided insights into how the local crystalline environment shapes the anisotropic electronic structure. [5] Angle-resolved photoemission spectroscopy (ARPES) has been used to study the electronic band structure of $FePS_3$, revealing the presence of spin-split bands due to the strong spin-orbit coupling. [7]

In this talk, resonant inelastic X-ray scattering (RIXS) has been used to study the spin-orbital excitations of $FePS_3$ and their relation to magnetism. By performing RIXS measurements as a function of temperature across the magnetic transition, we reveal that the identified multiplets have a large sensitivity to the spin state. Interestingly, the low-energy spin-orbital excitations in the presence of magnetic order are found to be strongly influenced by the trigonal lattice distortion and negative metal-ligand charge transfer, as revealed by simulation with ligand field theory calculations. This finding not only relates to the fundamental physics of $FePS_3$ but also establishes a generalized approach that is ideal for studying magnetic functional materials and the relation between their low-energy electronic properties and the magnetic state.

Beyond multiplets excitations, the RIXS spectra on vdW materials provide a rich platform for studying a wide range of magnetic phenomena, including spin-orbit coupling, magnetic anisotropy, and spin excitations down to the monolayer level. This will open up exciting new avenues for the design and development of next-generation magnetic devices, such as spintronic and quantum computing technologies.

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# Transition-metal *L*-edge RIXS studies of cathode materials for Li-ion batteries: Relation between the charge-transfer effect and electrode performance

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Resonant inelastic X-ray scattering (RIXS) is a promising tool for the electronic-structure analysis of electrode materials for Li-ion batteries (LIBs), in addition to X-ray absorption spectroscopy (XAS) and X-ray photoemission spectroscopy. Particularly, RIXS at the O *K*-edge is attractive to investigate the oxygen redox in Li-rich layered oxides which are expected as candidates of high-capacity cathode materials. Several groups have reported characteristic mechanisms about the oxygen redox by using O *K*-edge RIXS [1-4].

On the other hand, we have applied transition-metal (TM) *L*-edge XAS and RIXS in combination with multiplet calculations to various cathode materials to reveal not only the valence of TM, but also the hybridization and charge-transfer (CT) effect to the O 2*p* orbitals. The detailed information of the TM 3*d* and O 2*p* orbitals obtained by TM *L*-edge RIXS should be complementary to previous knowledges mainly focused on the valence change of the TMs in prototypical cathode materials. For example, the Mn *L*-edge RIXS and multiplet calculations for $LiMn_2O_4$ revealed very strong CT effects for the $Mn^{4+}$ in the charged state, which is much enhanced compared to the $Mn^{3+}$ in the initial/discharged state. Thus, the redox reaction of $Mn^{3+}/Mn^{4+}$ due to charge/discharge has a strong covalent character with the O 2*p* orbital [5]. In contrast, the redox reaction of $Fe^{2+}/Fe^{3+}$ for $LiFePO_4$ is attributed to an ionic character. The analysis for the Fe *L*-edge RIXS spectra unveiled that the CT effect from the O 2*p* to Fe 3*d* orbitals is very weak for both $Fe^{2+}$ and $Fe^{3+}$ states [6]. The difference between covalent/ionic characters possibly affects the structural stability on the TM-O bonds during charge-discharge, which should be related to the different cycle performances.

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# Cu L-edge RIXS Spectra of Cu-Proteins

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Amicyanine and Azurin are classified as Class I TICu proteins, whose axial ligand is the sulfur atom of methionine. Both protein molecules share the asymmetric structure of four amino acids, Met, Cys, and two His, as the ligand for the copper atom. Azurin contains an additional second axial ligand in addition to methionine in one axial position, as seen in the difference in distance to the oxygen atom. Crystallographic analysis has revealed that this asymmetry differs by a few percent between azurin and amicyanine[1]. Their redox potentials differ by about 30 mV at pH 8.0. In addition, genetic recombination technology has introduced both amino acid mutations, and significant redox potential changes of 500 mV or more have been observed when amino acid substitutions are introduced directly or indirectly at the copper atom. Especially the changes of nearly 100 mV have been reported even at positions more than 10 Å from the copper atom. In order to reveal these phenomena, we examined the resonant inelastic soft X-ray scattering (RIXS) measurement of these solutions.

Soft X-ray emission spectra around Cu-Ledge were observed. These spectra were obtained using a high-resolution soft X-ray emission spectrometer HORNET[2] installed at the end of the BL07LSU of SPring-8. The apparent peak structures of each spectrum were observed. In order to assign these peak structures, we are currently performing density functional theory simulations using a model molecule that imitates the coordination of amino acids around Cu. Using DFT calculation, N.J. Fowler et al. predicted reduction potentials for several Cu proteins containing mutants[1]. In the future, we plan to perform RIXS spectroscopy on mutants with different amino acid residues surrounding Cu or different pH to obtain information on the correlation between the structure and electronic properties around Cu. This information regarding copper proteins has valid for general use in biotechnology, such as enzyme-catalyzed fuel cells or valorization of lignocellulosic biomass.

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# Probing spin-polarized electronic structures of halfmetallic Heusler alloys using RIXS-MCD

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Toward the next-generation device development, the spintronics fully utilizing the spin-degrees of freedom gathers wide attention. To design the spintronic devices, it is essential to optimize the spin-polarized electronic structures of the magnetic materials under the magnetic fields. Especially, the spin polarization in the vicinity of the Fermi level ($E_F$) for the conducting electrons and the size of spin-dependent band gap are key parameters in the magnetic layers, which are usually covered by capping layers in the device. Resonant inelastic soft X-ray scattering (SX-RIXS), which is a photon-in/ photon-out spectroscopy, is a suitable technique to investigate buried electronic structures with element specific way by tuning the incoming photon energy to the core-level absorption edges [1,2]. Moreover, any external perturbations, such as magnetic field, can be applied to study the electronic structures under the *operando* conditions [3,4].

Recently, we established a versatile experimental technique to study the spin-polarized electronic structures of the magnetic materials by using SX-RIXS at the transition metal (TM) $2p_{3/2}$ absorption-edge in an external magnetic field [5,6]. The effective magnetic field of the TM $3d$ states yields the spin-polarized $2p$ core states due to the effective Zeeman splitting, and thus the magnetic circular dichroism (MCD) in the SX-RIXS spectra can reveal the spin-polarized electronic structures thanks to the dominating selection-rule of the dipole-allowed transition between the core-level $2p$ states and the $3d$ states for the circularly polarized light.

We applied this technique to probe the spin-polarized electronic structures of halfmetallic Heusler alloys [5,6], in which one of the spin-subbands is metallic and the other is semiconducting with a gap at the $E_F$. The half-metallic electronic structures of the ferromagnetic $Co_2MnSi$ and ferrimagnetic $Mn_2VAl$ are proved from the difference of the on-set energies between the positive and negative MCD signals (Figs. 1(b), (d)), reflecting the up and down spin components in the valence band electronic structures, respectively. Moreover, the RIXS-MCD allows us to confirm which spin-subband is opened as the halfmetallic gap, namely down and up spin gap in these materials, respectively, giving the useful information to design the spintronic devices.

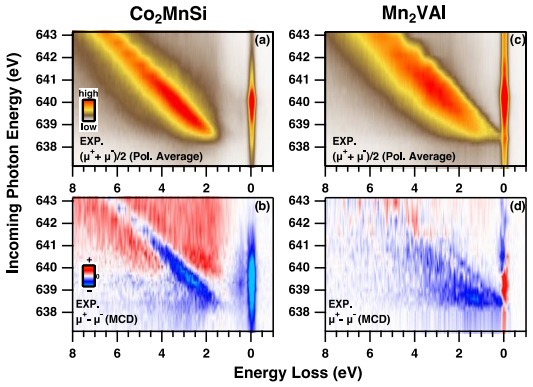


Fig. 1 Intensity plot of the polarization averaged RIXS and the RIXS-MCD data at the Mn $2p_{3/2}$ edge for $Co_2MnSi$ in (a, b), and $Mn_2VAl$ in (c, d).

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# Control of magnetic properties in iridate thin films by variation of strain and thin film thickness

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The complex 3*d* transition-metal oxides are one of the most fascinating and technologically relevant material systems, with a plethora of physical properties. The 5*d* transition-metal compounds offer the potential to further enrich the physics of this class of materials, as one can generate new physical properties by making use of strong spin-orbit coupling. To this end, the Ruddlesden-Popper series $Sr_{n+1}Ir_nO_{3n+1}$ has attracted considerable scientific attention due to the possibility of generating spin-orbit driven Mott insulating states [1]. In the first member of this series, the layered $Sr_2IrO_4$ (SIO-214) compound, the large spin-orbit coupling combined with a moderate Coulomb repulsion result in a Mott insulating $J_{\mathrm{eff}} = 1/2$-ground state, which is similar to the $S = 1/2$-ground state of high-temperature superconducting Cu-oxides. Additionally, the second member of the series, the bilayer $Sr_3Ir_2O_7$ (SIO-327), exhibits an antiferromagnetic excitonic insulating state [2]. To investigate the impact of strain and finite size effects on the magnetic properties of these compounds, we present a comprehensive study of epitaxially-strained SIO-214 and SIO-327 thin films. By using X-ray resonant scattering (REXS), we show that the out-of-plane magnetic correlation length of SIO-214 thin films is strongly dependent on the thin film thickness, but independent of the strain state of the thin films (see Fig. 1) [3]. Resonant inelastic X-ray scattering (RIXS) further demonstrates that strained SIO-214 thin films exhibit similar exchange constants than bulk SIO-214, while strained SIO-327 thin films clearly show a modified inter-layer coupling and enhanced anisotropic interactions mainly caused by finite size effects.

These results provide a clearer picture for the systematic control of the magnetic degrees of freedom in epitaxial thin films of SIO-214 and SIO-327 and highlight the potential for a rich playground, allowing to explore the physics of 5*d* transition-metal compounds.

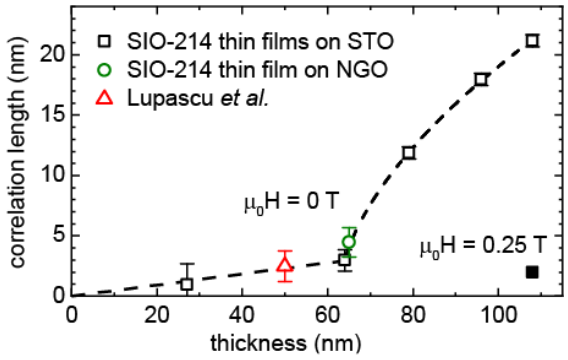


Figure 1: Out-of-plane correlation length as a function of SIO-214 film thickness deduced from REXS measurements [3].

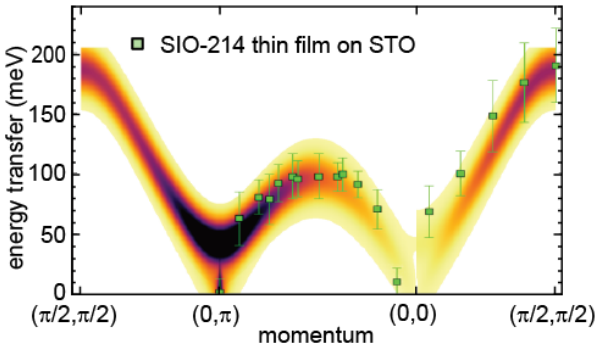


Figure 2: Single magnon dispersion of a SIO-214 thin film derived from RIXS measurements and Spin-W simulations.

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# Comparative RIXS investigation of five- and six-fold oxygen-coordinated 214-type cuprates

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Identifying the governing principle of transition temperature ($T_c$) is a central issue of high-temperature superconductivity in cuprates. The electronic structure of the so-called 214-type cuprate $La_{2-x}Sr_xCuO_4$ (LSCO) has been intensively studied to understand the mechanism of its low $T_c$. A short distance between $CuO_2$ planes and apical oxygen atoms is suggested detrimental for superconductivity in LSCO from several perspectives. First, small crystal-field splitting between the $d_{x2-y2}$ and $d_{z2}$ orbitals cause orbital hybridization and hence dilution of orbital characters around the Fermi level [1,2]. The short apical-oxygen distance also results in the increase of the charge-transfer gap and thus effective on-site effective Coulomb interaction $U$. It was suggested using the Hubbard model that larger $U$ for LSCO facilitates tendency toward charge ordering, which competes with superconductivity and could in turn lower $T_c$ [3].

While $La_{2-x}Sr_xCuO_4$ normally has six-fold octahedral oxygen coordination around Cu (T-type structure, Fig. 1), 214-type cuprates can also crystalize with five-fold pyramidal coordination (T*-type structure, Fig. 1) [4]. The lack of one apical-oxygen atom is expected to enhance crystal-field splitting and reduce the charge-transfer gap [5]. These changes would alleviate degradation of superconductivity, and, therefore, straightforward consideration predicts higher $T_c$ for the T*-type cuprates. However, maximum $T_c$ of the T*-type cuprates is even lower than that of the T-type counterpart [6]. Closer investigations of the electronic structure of the T*-type cuprates are thus desired.

We have performed comparative Cu $L$-edge RIXS studies for the T- and T*-type cuprates at Diamond Light Source beamline I21. The $d_{z2}$ component of the $dd$ excitation was observed at significantly higher energy for the T*-type cuprate, indicating larger crystal-field splitting between the two $e_g$ orbitals. As previously reported for multi-layer cuprates with five-fold oxygen coordination [7], larger magnon-dispersion anisotropy was found for the T*-type cuprate along the (π, 0) and (π, π) directions. Once hole-doped, a signature of charge ordering was observed for the T*-type cuprate but with much less pronounced manner than the case of the T-type cuprate. While the above aspects should be in favor of higher-temperature superconductivity, the magnon band width governed by the nearest-neighbor exchange was smaller for the T*-type cuprate, which may act negatively on $T_c$ [8]. The results suggest the delicate valance of multiple interactions governing the $T_c$ of the 214-type cuprates.

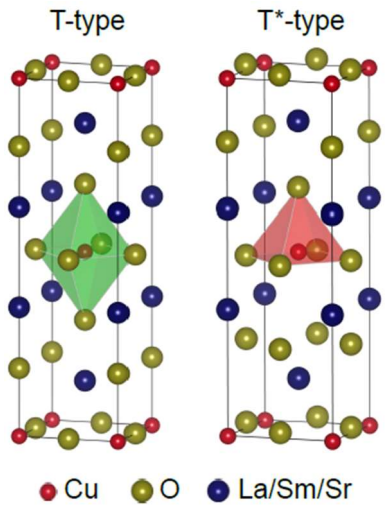


Fig.1: Crystal structure of the T- (left) and T*-type (right) cuprates.

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# Orbital excitations on the cusp of Mott-band insulator crossover in 1T-$TaS_2$

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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 com-pounds due to the more extended nature of the 5d orbital wavefunction, where direct experimental observations are expected urgently. 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/5p orbitals and the S 3p ligand state at the Ta L3 edge. 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. 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[1].

Our discovery provides new insight into the Mott vs. band insulator debate in 1T-$TaS_2$ 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.

**Acknowledgments**

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# Magnetic properties of binary ferrofluids investigated by RIXS-MCD spectroscopy

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Ferrofluids are suspensions of magnetic nanoparticles in a liquid, which have found numerous applications in biomedicine (magnetic hyperthermia, improved MRI agents…), technology (magnetic recording, permanent magnets…) or fine arts. Intriguing macroscopic properties of ferrofluids arise from their nanoscale structuring: when the magnetic dipolar interparticle interaction are large enough, particles assemble in chains and rings even in the absence of a magnetic field. These assemblies turn into microsized chains when an external magnetic field is applied. Variation of the nanoparticle size, shape and composition (e.g., hard or soft magnetic material) changes the magnetic anisotropy, which plays a crucial role in magnetic dipole interactions. This results in a modification of the structuring of ferrofluids as well as their collective magnetic properties like coercivity, magnetic saturation or remanence.

Understanding and controlling interparticle interactions could lead to improving the efficiency of known ferrofluids for applications and the discovery of novel magnetic responsive materials. Magnetic binary ferrofluids, i.e. those composed of two types of nanoparticles with different magnetic anisotropy, are particularly appealing because they offer an unprecedented interplay of magnetic dipole interactions. However, bulk magnetometry only allows to probe the average magnetic properties of the ferrofluid, which preventing from selectively measuring the contribution of each component. There is therefore a need for chemically selective magnetic measurements, which can be realized using Resonant Inelastic X-ray Scattering spectroscopy combined with X-ray Magnetic Circular Dichroism with hard x-rays (RIXS-MCD, [1]).

Element selective magnetization curves detected by RIXS-MCD have been measured at the GALAXIES beamline of the SOLEIL synchrotron on binary ferrofluids with different particle size, shape and composition. Zero Field Cooled and Field Cooled magnetic measurements have been performed to investigate how the structuring induced by an external magnetic field modifies the measured collective properties. Ferrofluid samples have been measured in the frozen state using a dedicated cryo liquid cell [2] which is inserted between the poles of an electromagnet. RIXS-MCD experiments are complementary to bulk magnetometry and First Order Reversal Curve diagram measurements of collective magnetic properties. Combined with the information obtained from Cryogenic Transmission Electron Microscopy and Electron Holography regarding the structuring, our results pave the way for a better understanding of the relationship between magnetic properties and structuring of binary ferrofluids.

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# The role of oxygen species on water uptake of Cyclic Olefin Polymers

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Currently, super hydrophobic material with relatively low water absorption property has gained considerable interest due to their wide range of applications to blood contacting devices. Within this context, a prominent candidate is a series of Cyclic Olefin Polymers (COPs) which are formed of the alternating repeating units of ethylene and five membered ring with various side groups. (Figure 1). COPs have a low water absorption property (<0.01%), four times less than polycarbonate and 10 times than poly-methyl methacrylate (PMMA) even in high humidity environment.

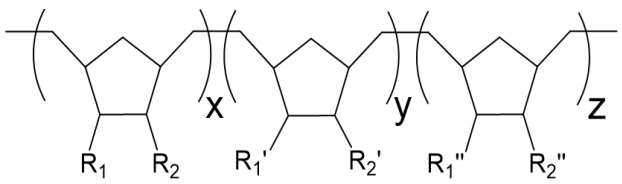


Figure 1. Typical molecular structure of COPs. Depending on the combination of $R_1$, $R_2$, X, Y and Z, COPs exhibit different functionalities.

The high biocompatibility of COPs results from its high hydrophobic nature following the interactions with living cells, which reduce side effect and facilitates the normal flow of blood and plasma. In addition, Anti-fouling property of super hydrophobic surface could prevent the attachment of organic substances and microorganism. These attributions are critical to medical applications such as filter membranes, prosthetic heart valves, drug eluting stents (DES), and extracorporeal membrane oxygenation (ECMO), which is used for oxygen therapy for critical patient suffering from COVID-19. Furthermore, due to their verified hemocompatibility and low cytotoxicity [1], COPs also have good potential as biomaterial for drug delivery and bio-interface. However, the mechanism of biocompatibility in the superhydrophobic COPs has not been fully understood under in detail and needs extensive studies.

The purpose of this study is to understand hydrogen-bonded structure of water and ratios of adsorbed water phases during humidification of the strongly hydrophobic COPs, which should be responsible for biocompatibility. A combination of O K-edge X-ray Absorption (XAS), Oxygen 1s X-ray emission (XES), and X-ray Photoelectron Spectroscopy (XPS) was used to unravel the nature of the oxygen species generated during the humidification treatment and clarify the mechanism of biocompatibility. The absorption of water on the hydrophobic surface is energetically not favorable and proceeded through a stepwise process. At the low relative humidity regime (<90% RH), it was found that water absorbed rather in an isolated domains on the hydrophobic COP surface. For RH>95% the saturation level is surpassed and the majority of absorbed water on the COP surface has a disordered bulk like water structure. Based on the pre-edge feature in the XAS spectrum, the existence of carbonyl group, peroxide and hydroxide species on the redried COP film was observed. This implies that in addition to the interaction between the bare hydrophobic COP surface and water, the slight hydrophilicity introduced by the oxygen functional groups created on the COP surface may contribute to the COP's biocompatibility.

**Acknowledgement**
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# Construction of Ultrahigh Energy Resolution 2D-RIXS at NanoTerasu: Advancements and Current Progress

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Resonant inelastic X-ray scattering (RIXS) is a powerful technique for studying the energies of electronic states and elementary excitations in materials by analyzing the energy of X-rays scattered off matter. Recent advances in energy resolution have enabled around 30 meV energy resolution at the Cu *L* edge [1,2], making RIXS an increasingly important tool in materials science. However, to observe lower-energy excitations, even higher energy resolution is desired. Thus, efforts are underway to enhance the resolution of RIXS measurements through different approaches.

At NanoTerasu in Tohoku, Japan, we are currently constructing an ultrahigh energy resolution RIXS facility. This advanced facility consists of a dedicated beamline [3] and a 2D-RIXS spectrometer. Our goal is to achieve a total energy resolution (combined resolution of the beamline and RIXS spectrometer) of sub-10 meV below $h\nu$=1000 eV. To achieve such ultrahigh energy resolution, we employed the $h\nu^2$-concept [4,5] for our RIXS spectrometer, to compensates for the reduced measurement efficiency inherent in enhancing energy resolution.

Figure 1 shows the optical layout of the 2D-RIXS facility. The beamline and the 2D-RIXS spectrometer span ~76 m and ~12 m, respectively, and the RIXS spectrometer is planned to have a 2θ rotation range of 30–150°. The beamline has no exit slit, and vertically energy-dispersed X-ray from the beamline monochromator is directly irradiated onto a sample. The RIXS spectrometer features an imaging mirror that collects scattered X-rays from the sample and images them onto a detector while maintaining the relationship between the vertical position and the incident energy. Simultaneously, the grating in the RIXS spectrometer disperses and focuses the scattered X-rays horizontally onto the detector. Thus, the energies of the incident and scattered X-rays are "two-dimensionally" resolved on the detector, which is why we call "2D-RIXS". By integrating the intensity of the spectra along the incident energy, we can improve measurement efficiency.

In the poster, we will provide an overview of the optical design of the 2D-RIXS facility and discuss our efficiency improvements.

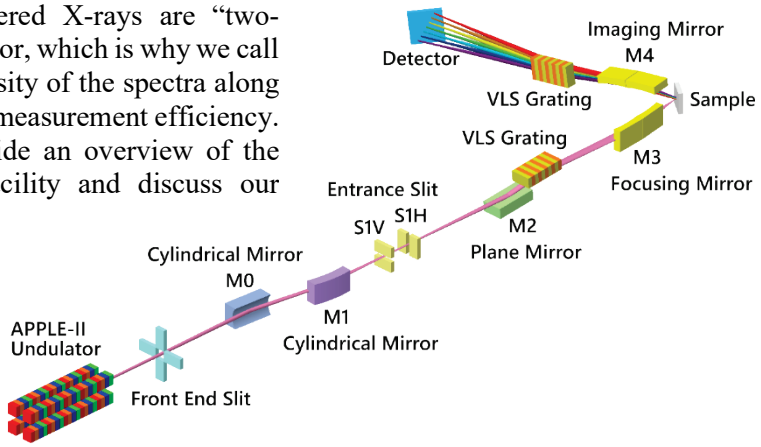


Figure 1: Schematic representation of the optical layout of ultrahigh resolution 2D-RIXS facility at NanoTerasu.

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# Charge order in cuprates revealed by resonant inelastic x-ray scattering

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I will present recent results on charge ordering, which strongly competes with high-temperature superconductivity, in underdoped cuprates investigated by Cu *L* edge (931.5 eV) resonant inelastic x-ray scattering (RIXS) experiments carried out at the beamline ID32 of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. In particular, we focused on one particular member of the cuprate family, i.e., the stoichiometric, nearly-disorder free high-temperature superconductor $YBa_2Cu_4O_8$ (Y124). We find charge ordering qualitatively akin to that observed in non-stoichiometric $YBa_2Cu_3O_{6+x}$ (Y123), which has been extensively studied as a model system for charge ordering in cuprates. However, the temperature evolution of the charge order of Y124 is distinct from the 'gradual freezing' of Y123, albeit the finite correlation length persisting even at the lowest temperature indicates a short-range nature of the charge order in Y124 as well. Interestingly, the onset temperature of the charge order was found to match the temperature of the charge-transport anomaly. In this framework of connecting transport and charge order, I will also address the uniaxial stress response of the transport coefficients in Y123, which show a remarkable correspondence with the onset of charge order [1].

Our findings establish the presence of charge ordering in Y124 and provide new insights into the relationship between disorder, charge-ordering formation, and other prominent phenomena in underdoped cuprates.

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# Spin quadrupole excitations in frustrated ferromagnetic chain

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Frustrated quantum magnets frequently exhibit non-trivial ground states that do not have a classical analogue, since the combined effects of frustration and quantum fluctuations destroy conventional magnetic order. The spin nematics, characterized by spin quadrupoles, is a prominent example. Here, we focus on a spin nematic liquid state in a frustrated ferromagnetic chain with nearest-neighbor ferromagnetic and next-nearest-neighbor antiferromagnetic exchange interactions in a magnetic field. To clarify the property of the spin nematic liquid state from the viewpoint of excitation dynamics, we have studied excitation spectra by dynamical density-matrix renormalization group methods [1,2,3,4]. In the spin nematic regime at high magnetic fields, we find gapless longitudinal and gapped transverse spin excitation spectra, in accordance with quasi-long-range longitudinal and short-range transverse spin correlations, respectively. Moreover, in the quadrupole channel, we observe gapless excitations at $q=\pi$, signaling quasi-long-range antiferro-quadrupole correlations. These characteristics of excitation spectra are useful and essential to identify the spin nematic state by experiments in real compounds. However, conventional magnetic probes are usually insensitive to spin quadrupoles. In this context, based on symmetry arguments, it has been suggested that resonant inelastic x-ray scattering (RIXS) would provide a route to detect quadrupole excitations [5,6]. We will present detailed numerical results of quadrupole excitation spectra and discuss possible relevance to RIXS.

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# Entangled spin-orbital-lattice order in $Ba_2MgReO_6$

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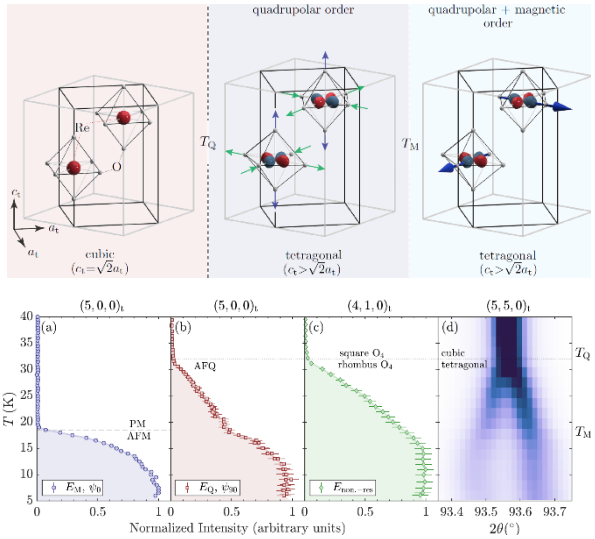
Quantum materials which exhibit an intimate coupling between the local structural distortions, magnetic dipoles and charge quadrupoles have generated sustained interest because they have the potential to give rise to new exotic phases of matter such as quantum spin liquids, axion insulator etc. In cases where the coupling between the lattice, spin, and orbital degrees of freedom is particularly strong, these entities become deeply intertwined, and cannot be considered as separate components.

The long-range order of such local entangled states can be stabilized in $5d^1$ double perovskite (DP) systems, owing to the comparable lattice strain, spin-orbit coupling, electron correlations and magnetic exchange energies. One such example of a realization is in the $5d^1$ DP $Ba_2MgReO_6$, which displays two successive anomalies at $T_Q$ and $T_M$, which demarcate three distinct phases [1].

Using a combination of REXS at the Re $L_3$ edge (DESY, Germany), RIXS at the Re $L_3$ edge (Spring-8, Japan), RIXS at the O $K$ edge (SLS, Switzerland), density functional theory (DFT) calculations, quantum chemistry (QC) calculation and heat capacity measurements at high field (30 T), we find mounting evidence for a highly coupled spin-orbital-lattice eigenstate in $Ba_2MgReO_6$.

Our REXS results show that below $T_Q$, $Ba_2MgReO_6$ develops anti-ferroic order of eigenstates composed of an entanglement between the rhombic in-plane movement of oxygen ligands (green arrows in Figure) and an ordering of Re $5d^1$ $Q_{xy}$ charge quadrupoles. Simultaneously, we also find that there is ferroic order of eigenstates composed of an entanglement between the apical movement of oxygen ligands (blue arrows in Figure) and an ordering of Re $5d^1$ $Q_{z2}$ charge quadrupoles.




Our RIXS measurements indicate that this entangled eigenstate arises from an inherent coupling between the structural and charge degrees of freedom. Moreover, our QC and DFT calculations show that the entanglement between these degrees of freedom are the lowest energy ground state of the system, which accounts for why they work cooperatively.

Hence, our results verify the mean-field models [2-4], dynamical mean-field [5] and density functional calculations [6-8] which all predict charge quadrupolar order of $5d^1$ ions in DP systems. Furthermore, we make a case for a single order parameter composed from an intimate coupling between the spin-orbital and lattice degrees of freedom.

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# LDA+DMFT approach to resonant inelastic x-ray scattering in strongly correlated magnetic systems

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Resonant inelastic x-ray scattering (RIXS) has become a valued tool for studying materials with strongly correlated electrons, such as transition metal compounds, thanks to a remarkable improvement of the energy resolution in the last decade. Though RIXS has a broad sensitivity to excitations of spin, orbital, and charge degrees of freedom, its drawback is often complicated experimental spectrum, which calls for its theoretical modelling. For strongly correlated electron systems, impurity models, such as cluster model including an excited metal site and its surrounding ligands, have been widely used for the spectral analysis. However, since band excitations are lacked in the cluster model, a disentanglement of the localized and delocalized RIXS features is prohibited. Band-structure approaches for RIXS often suffer from difficulties in incorporating the strong correlation effects. In this context, recently, we introduced an *ab-initio* method based on local density approximation (LDA) + dynamical mean-field theory (DMFT) for modelling RIXS spectra of strongly correlated electron systems [1,2]. In this method, DMFT supplements electron correlation effect to the material specific bands provided by LDA. Thus, the LDA+DMFT method allows us to describe continuum of band excitations (with unbound electron-hole pairs) as well as bound excitations (such as intraatomic multiplet excitations) simultaneously.

In this work, we apply the LDA+DMFT method to study magnetic and orbital excitations in two correlated magnetic systems $Fe_2O_3$ and $RuO_2$ of much recent attention. For $Fe_2O_3$, an antiferromagnetic Mott-insulator with a large spin degrees of freedom $S$=5/2, we present a detailed comparison with the high-resolution RIXS at the Fe $L_3$-edge and reveal the energy dispersion as well as the excitation mechanism of higher-order magnons [3]. For $RuO_2$, a representative compound of altermagnetic system [4], we will discuss possible magnetic circular dichroism in $M$-edge x-ray absorption spectroscopy and RIXS, and its relation to the unique spin properties in this material.

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# High resolution spin texture imaging in spin caloritronics device structures: New opportunities for REXS at 4th generation light sources.

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The rare earth iron garnets (RIG), since conception have required novel probes to understand their magnetic properties, e.g. neutron diffraction. The recent renaissance in RIG research is being driven by their potential in novel spin (calori)tronic [1] devices. The development of emerging nanotechnologies exploiting spincaloritronic physics, requires new microscopic tools to reveal how atomic structure and magnetism are related at the nanoscale. The advent of new 4th generation synchrotron sources, such as the ESRF EBS, promise to deliver higher coherence and higher flux focused X-ray beams, to meet the knowledge demands for 21st century technologies. Recent advancements in REXS with x-ray nanobeams at the ESRF have revealed spin textures [2], responsible for degrading the thermoelectric efficiency of the SSE, with high spatial resolution. Here, we present new nanobeam diffraction results from the new ESRF EBS, where we demonstrate an increase in spatial resolution from 400nm to 80nm. Furthermore, the expansion of these imaging techniques to combine REXS and Bragg ptychography methods, with a potential spatial resolution of 16nm, will also be presented.

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# Role of magneto-crystalline anisotropies in complex rare-earth garnets

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Rare-earth crystals offer a rich variety of fascinating phenomena, such as emergent gauge theories, topological phases, magnetoelasticity, multiferroicity, and unconventional ordering. Complex garnet crystals combining transition-metal (TM; e.g. Fe) and rare-earth (RE; e.g. Tb) ions are considered among the most intriguing ones because of the intertwined physics arising from the open *d*- and *f*-shell electrons of TM and RE ions, respectively. The microscopic mechanisms underpinning their magnetic and electric response are considered pivotal for harnessing energy-conversion in garnet-based spintronic devices, especially to move beyond the yttrium iron garnet paradigm [1].

Motivated by the knowledge that crystal-fields anisotropies allowed for the description of the magnetostatics of individual ions and for the spin-dynamics of their effective lattices [2, 3], we complement experiments on the structure and dynamics of rare-earth iron garnets – $RE_3Fe_5O_{12}$ – with theoretical studies whose building blocks exploit the wave-functions of the RE *f*-electrons. Focussing on the challenges posed by terbium iron garnet – $Tb_3Fe_5O_{12}$ – we discuss the role of the electric and magnetic contributions to the multipoles of the RE ions, and we overview the predictions permitted by the knowledge of the $D_2$ point-group symmetry of a rare-earth ion in a garnet [4]. In relation to experiments, we tackle the azimuthal dependence from the theory of resonant elastic x-ray scattering (REXS), detailing on the competing contributions from the crystalline axes and the magnetic dipolar moments [5].

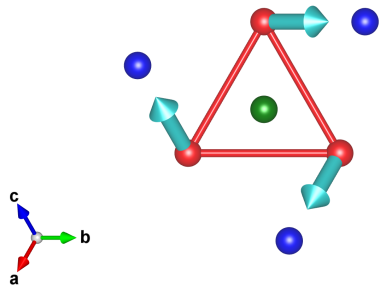


Figure 1: Local crystalline anisotropies (light blue arrow) of the simplest magnetic unit of Tb-ions (red spheres, *c*-sites) in a Tb-garnet – the triangle is the basic 'simplex' of the hyperkagome lattice which host the RE-ions in garnet crystals. The blue and the green spheres, respectively *d*- and *a*-sites, show the TM ions nearest neighbours to the central RE-triangle from the unit-cell of a garnet.

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# The Study on Water Repellent Behavior of Hydrophobic Coatings by Soft X-ray Spectroscopy

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Self-assembled monolayers (SAMs) are nanostructures known for their ability to modify interfacial properties of substrates such as metals and metal oxides, and provide useful functionality such as biocompatibility [1]. Among them, SAMs containing fluorine are known to provide unique properties such as high water repellency, slipperiness, and lubricity due to their low surface tension, and have been extensively studied and developed in various industrial fields [2, 3]. Understanding the hydrophobic mechanism of materials at the molecular level is useful for the development of functional materials, but little is known about the influence of the chemical structure of SAMs on water adsorption behavior.

In this study, we employed soft X-ray emission spectroscopy (XES), a powerful tool for examining the occupied electronic structure of light elements including water, to acquire a deeper understanding of water adsorption behavior on SAMs with different water repellency and chain structures.

We conducted XES measurements on hydrocarbon and fluorocarbon chain structures of SAMs with different water repellency by using a setup developed by our research group that enables XES measurements under precise humidity control (**Fig. 1**). The results in **Fig. 2** demonstrated that water molecules were progressively trapped on the surface and within the chain, as indicated by the enhancement of the $1b_1'$ peak corresponding to trapped water with an increasing supply of water molecules for the hydrocarbon chains, while no such behavior was observed in the fluorocarbon chains (Medium humidity). On the other hand, there was no significant change for both chains in the initial adsorption phase of water (Low humidity). Based on the results of XES and molecular dynamics simulations, we discuss in detail the behavior of water adsorption on molecular chains with different water repellency.

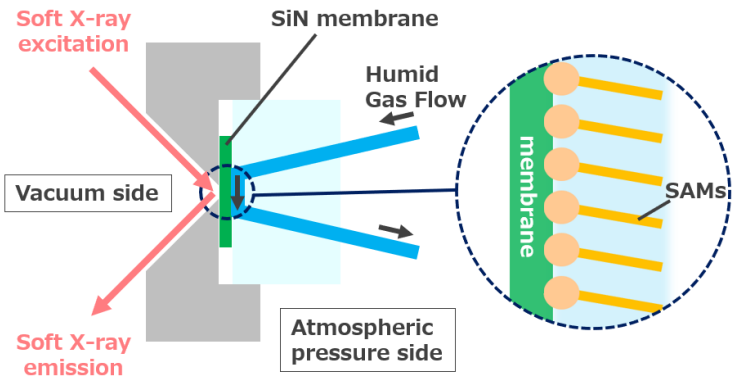


**Fig. 1.** Schematic of experimental setup for XES measurements and magnified image of substrate surface.

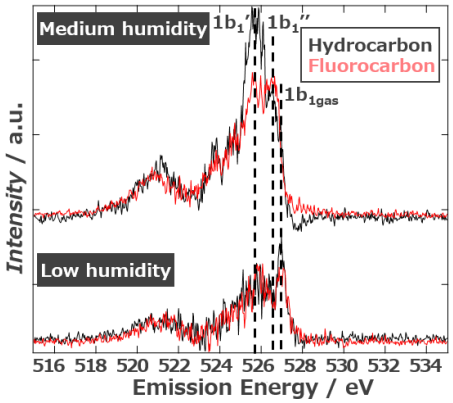


**Fig. 2.** O1s XES spectra obtained during humid gas flowing over SAMs.

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# Cu $L_3$-edge resonant inelastic x-ray scattering on isostructural copper oxides $CaCu_3Fe_4O_{12}$ and $EuCu_3Fe_4O_{12}$

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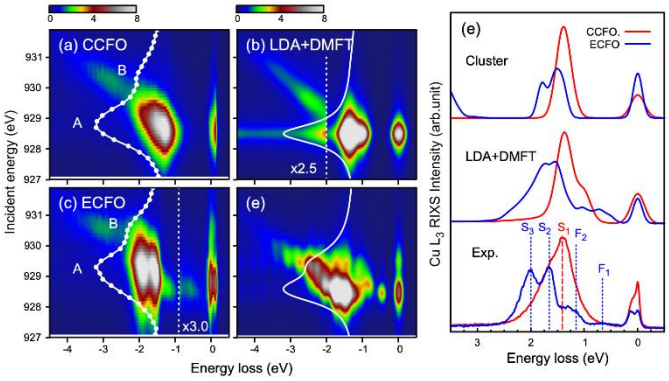
We report a Cu $L_3$-edge resonant inelastic x-ray scattering (RIXS) study for the electronic structure of isostructural copper perovskite oxides $CaCu_3Fe_4O_{12}$ (CCFO) and $EuCu_3Fe_4O_{12}$ (ECFO) with different insulating and magnetic ground states: CCFO is a ferromagnetic insulator and ECFO is an antiferromagnetic insulator [1,2]. As shown in Figs.(a,c), the Cu $L_3$-edge RIXS map exhibits a strong material dependence. The RIXS spectra are analyzed using local density approximation (LDA) + dynamical mean-field theory (DMFT) [3] and a conventional $CuO_4$ cluster model.




The experimental Cu $L_3$-edge RIXS spectra for (a) CCFO and (c) ECFO. The LDA+DMFT Anderson impurity model spectra for (b) CCFO and (d) ECFO. White lines in panels are Cu $L_3$-edge XAS spectra for corresponding compounds. (e) The comparison of the experimental and calculated spectra by the LDA+DMFT method and $CuO_4$ cluster model.

A low-energy excitation is observed at $E_{Loss}$~0.7eV in ECFO, that is absent in CCFO, see Figs.(a,c,e). We identified this feature as a transition from the Zhang-Rice singlet ground state to a Zhang-Rice triplet excited state. Besides, clear double peaks at $E_{Loss}$~2eV, see Fig.(e), are observed in the *dd* excitations of ECFO, while CCFO exhibits only a broad single-peak *dd* feature. The double peaks of ECFO originate from an interplay of crystal-field and Hund's excitations. Based on the experimental findings as well as a comparison of the LDA + DMFT and cluster-model simulations, we discuss the microscopic origin of different electronic properties in the two compounds.

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# Anisotropic Magnetic Dipole Detectable by Resonant X-ray Scattering

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Magnetic and electric dipoles are a source of magnetism and ferroelectricity, respectively. Electric quadrupoles, described by electron multipoles, cause orbital ordering. Such fundamental degrees of freedoms play important roles in phase transitions and various responses with broken symmetry. Magnetic and electric multipoles are mainly discussed within single ion site. Recently, an augmented magnetic multipole has been generalized as a cluster inter-atomic multipole. For example, a 120-degree antiferromagnetic structure in $Mn_3Sn$ is regarded as a magnetic cluster composed of the same symmetry as a magnetic octupole. The augmented magnetic octupole is related to an anisotropic magnetic dipole, which can be detected by x-ray magnetic circular dichroism [2,3]. I will discuss a possibility of anisotropic magnetic dipoles detectable by resonant x-ray scattering with some specific examples.

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# Hard X-ray High Energy Resolution Spectroscopy Beamline at High Energy Photon Source

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Since 2019, the 6-GeV green-field 4th generation (with emittance <60 pm·rad) synchrotron facility, High Energy Photon Source (HEPS), is being built in Beijing, China. [1-2] The Hard X-ray High Energy Resolution Spectroscopy Beamline in HEPS Phase-I provides a suite of inelastic x-ray scattering spectroscopies using hard X-rays of 7~26 keV, such as nuclear resonant scattering, x-ray Raman scattering (XRS) and (non-)resonant inelastic x-ray scattering (IXS) spectroscopies. [3-4] For IXS, a 4 m-wide and 3 m-high "Qiankun (乾坤)" spectrometer is designed (Figure 1). A RIXS module with switchable 1 m/2 m Rowland circles as well as the rest XRS modules can be installed on the spectrometer, sharing the same granite base, KB mirror and sample stages. Specifically, the RIXS module and high energy resolution monochromators enable RIXS experiments with tunable energy resolution in the energy range of 7~15 keV. Once completed, the new spectrometer of the new beamline at the new synchrotron source will serve users for a variety of applications spanning over condensed matter physics, chemical/material sciences, etc. In this presentation, we will show the progresses of the beamline as well as related scientific perspectives.

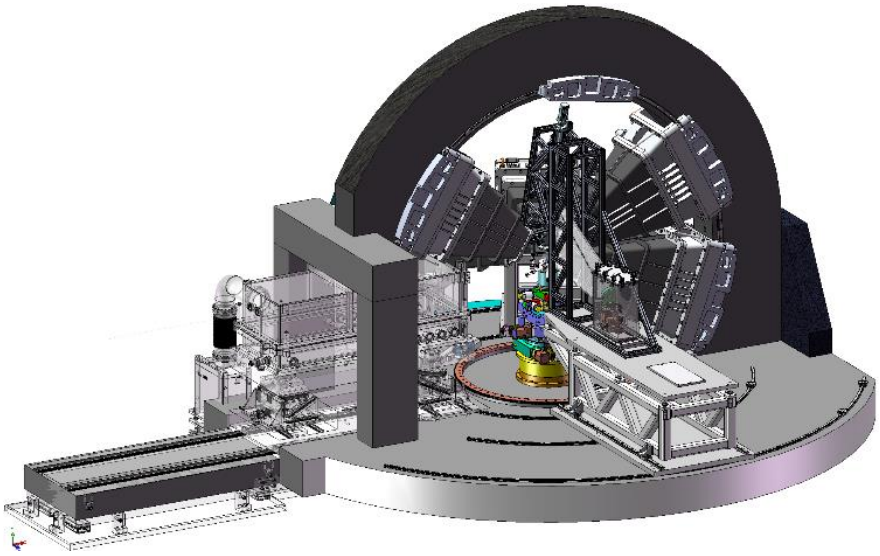

Figure 1. "Qiankun (乾坤)" spectrometer with RIXS module installed.

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# Momentum-resolved RIXS studies on high-temperature superconductor Bi2212

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We used momentum-resolved measurements of Cu $L_3$-edge resonant inelastic x-ray scattering (RIXS) to investigate the charge excitations of optimally-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212). Our momentum-resolved RIXS data are in agreement with the calculated charge susceptibility. The observed charge excitation in the 2D momentum plane indicates the presence of long-ranged Coulomb interaction between valence electrons. However, the scattering pattern notably differs from the ring-like feature reported in prior RIXS measurements. In addition, the RIXS spectral weight of charge excitation changes across $T_c$, providing experimental evidence of the superconducting gap. Our results confirm that RIXS can probe the evolution of charge gap across the superconducting phase transition.