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Oral Presentation Abstracts

RIXS beyond cuprates: an overview

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RIXS is a photon-in photon-out spectroscopy technique that provides element-sensitive and bulk-sensitive information about excitations in condensed matter systems. In the last two decades, energy momentum dispersion relations of collective excitations such as magnons, spinons, and phonons have been measured. In addition, RIXS has become an extremely powerful tool for studying orbital physics, providing detailed information on energy levels of orbital states as well as collective excitations of orbitals called orbitons. Although cuprates have been the favorite of RIXS community, RIXS is playing increasingly important roles in the study of many other quantum materials, such as transition metal oxides and rare-earth compounds. In this talk, I will focus on recent progress made in the study of magnetic materials with 5d and 4d transition metal elements. In particular, spectacular advances in the hard x-ray RIXS instrumentation has made it possible to investigate magnetic excitations in iridates with unprecedented energy resolution. High-resolution RIXS is an ideal probe of magnetism in iridates, which exhibit exotic quantum phases due to the strong spin-orbit coupling. In addition, the similarity between Sr_2IrO_4 and cuprate parent compounds make RIXS investigations of various iridates of particular interest to the RIXS community. Recent studies also revealed that orbital physics in osmates, rhenates, and ruthenates can be studied using RIXS. Finally, scientific opportunities for RIXS investigations of quantum materials beyond cuprates will be discussed.

X-ray core level spectroscopy, what do we learn and which level of theory do we need for each type of experiment.

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During this introductory talk, I will describe several forms of core level (x-ray) spectroscopies (absorption, photo-emission, resonant and non-resonant elastic and inelastic x-ray scattering). I will discuss what one can learn from these experiments, either by sum-rules, fingerprinting, (i.e. comparing them to spectra from known compounds) or by comparing experiment to theory. I will discuss which level of theory is needed to describe these experiments and what currently can and cannot be done (DFT, MLFT, DMFT). I will show how these different levels of theory are implemented in QUANTY, a flexible script language.

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Studies of Phonons with Inelastic X-Ray Scattering and First-Principles Simulations: Anharmonicity, Electron-Phonon and Spin-Phonon Couplings.

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A detailed understanding of atomic vibrations in solids is needed to refine microscopic theories of thermal transport and thermodynamics, and is of practical interest for the design of new functional materials. In particular, anharmonic phonon-phonon couplings are important to rationalize and design both ferroelectrics and thermoelectrics, in which strong anharmonicity is found near lattice instabilities. In superionic compounds, the anharmonic dynamics leading to large ionic mobility remain insufficiently understood and strongly debated. The interplay between phonons and other degrees-of-freedom of charge and spin is also critical to rationalize multiferroics, or metal insulator transitions, as well as high-pressure superconductivity. As illustrated in our recent studies [1-10], inelastic x-ray scattering (IXS) and inelastic neutron scattering (INS) measurements can be used to map phonon dispersions and linewidths throughout the Brillouin zone. In particular, IXS can conveniently probe phonons in small crystals, as function of temperature and pressure. In this presentation, I will highlight some recent achievements and opportunities for IXS studies of phonons in topical materials. In addition, our group performs density functional theory (DFT) simulations of the phonon dispersions, linewidths, and experimental cross-sections, in order to rationalize measurements. Such combined studies provide critical insights into the dominant phonon scattering mechanisms, including anharmonic phonon-phonon scattering, electron-phonon coupling, spin-fluctuation scattering, or scattering by crystal defects. I will illustrate the power of IXS studies in studies of the microscopic underpinnings of phase stability and thermal conductivity in a number of materials, such as VO_2 [1-3], $CuCrSe_2$ [4], Mo_3Sb_7 [5], and $CuCrO_2$ [6]. In complementary INS studies, we identified the dominant anharmonic effects and origins of low thermal conductivity in PbTe, SnTe and SnSe [7,8] and traced the origin of anharmonicity to the underlying electronic bonding with DFT. Improper multiferroics, such as $YMnO_3$, will also be discussed [9]. This deeper understanding of anharmonic phonon quasiparticles opens new avenues to tailor microscopic thermal transport and design new materials.

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Nonequilibrium Lattice Dynamics in photoexcited bismuth

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We describe recent results using the Fourier-transform inelastic scattering (FT-IXS) approach for measuring non-equilibrium lattice dynamics. FT-IXS measures the temporal evolution of phonon-means-square displacements following and external pump-pulse through broad-band femtosecond x-ray diffuse scattering without the use of a crystal analyzer, see e.g. refs [1-3]. Using photo-excited bismuth as an example, we show how FT-IXS yields microscopic information on excited state dynamics, including the first direct, channel resolved measurement of third-order anharmonic coupling constants between the coherent zone-center A1g phonon to pairs of high wavevector acoustic phonons throughout the Brillouin zone. The results are within an order of magnitude of first principles-based calculations [4]. We also measurements of the excited-state phonon dispersion and extract the transient interatomic forces, associated with a partial reversal of the Peierls distortion. We find that along with the A1g mode softening, we find that both the optical and acoustic branches are renormalized, including considerably more softening of the transverse acoustic modes than predicted by theory [5]. The changes in the phonon dispersion are driven primarily by a softening of the nearest neighbor forces along the bonding directions concomitant with a change in quasi-equilibrium position induced by photoexcitation [6,7].

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Charge fluctuations and electron phonon coupling studied by very high resolution RIXS

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In the last 10 years, soft x-ray RIXS has decisively contributed to the assessment of the magnetic properties [1,2,3,4] and to the discovery of charge density waves of high Tc superconducting cuprates [5,6]. The advent of last generation instruments, such as ERIXS at ID32 of the ESRF [7], is opening new opportunities thanks to the superior energy resolution and sample manipulation.

I will review some of the most recent results of our group. Especially significant is the discovery of dynamic charge density fluctuations in underdoped and optimally doped Nd123 and Y123, that are directly related to the marginal Fermi liquid behavior of cuprates in the normal state [8]. And I will present a series of experiments aimed at extracting the momentum dependent electron-phonon coupling in cuprates and in STO directly from the RIXS spectra [9,10].

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A spectroscopic journey into the critical electron states in batteries

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Energy storage through electrochemical devices (batteries) is under pressure to be greatly improved for today's sustainable energy applications, especially the electric vehicles and power grid using renewable energy sources. Often the bottleneck of the energy density is the positive electrode, with *3d* transition-metal oxides as the active material. Fundamentally, the electrochemical cycling of a battery directly triggers changes on the electron occupation numbers in TM-3d and O-2p states, as well as the evolution of the electronic configuration. Directly detecting and quantifying these chemical changes associated with the reduction-oxidation (redox) reactions in batteries is critical to clarify the cycling mechanism and to reveal the battery failure modes.

In this presentation, we first explain the advantages of utilizing soft X-ray spectroscopy, including X-ray absorption (XAS), emission, and inelastic scattering spectroscopy, as a direct probe of the critical valence states in transition-metal oxide based battery materials. We will discuss recent soft X-ray findings of both the transition metals and oxygen states in batteries. For transition metals, we show that soft XAS could explain an intriguing interstitial water effect on electrochemical performance through its sensitivity to the spin states[1], and full energy-range fast mapping of resonant inelastic X-ray scattering (mRIXS) could clarify a century-long speculation of novel Mn states in high-performance electrodes[2]. For oxygen, we explain the limitations of conventional XAS spectroscopy, and show that mRIXS provides the ultimate spectroscopic probe of the intrinsic oxygen redox reactions in batteries[3]. Further, the independently quantified spectroscopic results of transition-metals and oxygen successfully decipher the electrochemical capacity in battery electrodes, providing an ultimate probe to understand the evolution of the reactions mechanism upon electrochemical potentials and cyclings.

Brief introductions on basic electrochemical properties will be explained for a broad audience. The discussions on spectroscopic results will focus on revealing the critical electron states and its meanings for both fundamental understanding and practical improvements.

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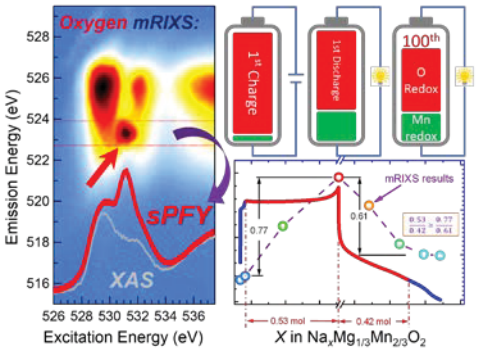

Figure: (left) Soft X-ray mRIXS detects the lattice-oxygen redox reactions in a $NaMn_{1/3}Mn_{2/3}O_2$ electrode, while XAS (gray spectrum) misses the key signature (red arrow). The independently quantified Mn (through mRIXS, not shown) and oxygen (through sPFY shown as the red spectrum) redox contributions deciphers the total electrochemical capacity at different potentials upon cycling. Evolutions on the Mn and O reactions are clearly revealed for hundreds of cycles. Figure from Ref.[4] with permission.

New developments in Inelastic X-ray Scattering at the Advanced Photon Source

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We will review experimental capabilities in Inelastic X-ray Scattering, past, present and future, and explore scientific opportunities for pioneering IXS studies enabled by improvements of synchrotron radiation sources, IXS instrumentation, energy resolution, and meaningful in-situ sample environments

IXS Opportunities offered by High Repetition Rate X-ray Free Electron Lasers

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High repetition rate XFELs are capable of delivering orders of magnitude average spectral brightness in addition to the high peak brightness. This presentation will provide an update of the status of LCLS-II and LCLS-II HE projects at SLAC, current plan for IXS instruments at LCLS-II and LCLS-II HE, as well as scientific opportunities associated with them. Novel ideas to further improve the performance of high repetition XFEL will also be discussed.

Looking at low energy excitations in the lanthanides with RIXS: current status and opportunities

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Over the last decade soft X-ray RIXS has established itself as a powerful probe of the low energy excitations in the 3*d* transition metal compounds, testified for example by its enorm impact in the field of cuprate high-T_c superconductors. A major reason for the success of the technique has been the steady push for higher resolution, better control of the scattering geometry and more advanced detection schemes, including polarization resolved RIXS. This has resulted in a new generation of extremely capable instruments that has become available now.

The most recent advances in soft X-ray instrumentation also created new opportunities in better understanding the equally diverse and complex physics of lanthanide compounds. Due to the very localized character of the 4*f* shell the relevant energy scales in these systems are typically an order of magnitude smaller than in transition metal compounds, which was until recently prohibitively small to seriously consider probing them with RIXS.

In this talk the first RIXS studies of lanthanides in the context of this new instrumentation will be reviewed and some important differences in the RIXS response of these systems compared to the 3*d* compounds will be highlighted. In a second part an outlook will be given where RIXS could be extremely suitable to grant truly new insights into the physics of 4*f* systems. I will finish with a brief discussion of further developments that could help this new field of research grow out of its infancy.

High Performance Soft X-ray RIXS and Applications to Magnetic Materials

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RIXS is a bulk-sensitive photon-in/photon-out spectroscopic and scattering technique to investigate the electronic structure and elementary excitations in materials, which offers considerable flexibility in terms of samples and sample environments. To keep step with the construction of a new 3 GeV ring supposed to be operational from 2023, we are planning to construct two high resolution soft X-ray RIXS beamlines/end-stations. One is a high energy resolution RIXS spectrometer which gives priority to detection efficiency with the total length of ~2.5 m and will be built as an upgrade of the HORNET station [1], and the other is a newly-developed ultrahigh energy resolution RIXS spectrometer with the total length of ~12 m. In the upgraded HORNET station, the resolving power will be improved from the present 8,000 to ~50,000 at 500 eV (~30,000 at 900 eV) with the vertical X-ray size of 0.5 μm, the grating slope error of 0.1 μrad, and the CCD spatial resolution of 5 μm. Since the dimension of the spectrometer is not changed from the original, the RIXS signal intensity will be enhanced more than 10 times higher according to the increase of the incident flux. In the ultrahigh energy resolution RIXS spectrometer, the resolving power is designed to be >150,000 at 1000 eV with the vertical X-ray size of 0.5 μm, the grating slope error of 0.05 μrad, and the CCD spatial resolution of 2 μm, and the combined resolving power with the beamline of >100,000 will be challenged. The complementary use of the high efficiency and ultrahigh energy resolution RIXS spectrometers will synergistically promote materials research and the characterization of physical properties. We have promoted several developments and research of the use of high performance RIXS using the HORNET station at SPring-8 BL07LSU: magnetic circular dichroism in RIXS (RIXS-MCD) under magnetic field [2], RIXS under electronic field, in-situ/operando RIXS [3,4], damageless RIXS measurement of Fe complexes [5], and RIXS of condensed matter [6,7] and solution [8,9]. By RIXS-MCD, we identified electronic structure which induces the weak ferromagnetism of α-Fe_2O_3 and obtained a clear evidence of half metallicity of Mn_2VAl.

In the presentation, I will talk about the conceptual design of the new beamline and spectrometers as well as recent activities at SPring-8 BL07LSU with a focus on magnetic materials revealed by RIXS-MCD.

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Spin and Charge Excitations in Stripe-Ordered Cuprates

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Recent resonant inelastic x-ray scattering (RIXS) for a stripe-ordered hole-doped cuprate has shown that spin excitations along the $(\pi,0)$ direction perpendicular to the charge stripe are strongly influenced by the emergence of a static charge stripe [1]. In contrast, RIXS in an electron-doped cuprate has shown a small downward shift in energy toward $(\pi,0)$ [2]. In an overdoped region of hole-doped cuprate, strong anisotropic spin excitation between the $(\pi,0)$ and (π,π) directions has been reported from RIXS [3]. These RIXS data have uncovered characteristic spin dynamics in doped Mott insulators. Therefore, it is crucial for theory to explain these spin excitations. We first examine spin excitations in a stripe-ordered doped Mott insulator using a 24×4 *t-t'-J* ladder whose ground state has a clear stripe order with a four-site charge period. Using dynamical density-matrix renormalization group (DDMRG), we find the influence of the stripe on spin excitations along the $(\pi,0)$ direction [4]. With increasing the number of leg on ladder toward square lattice, we find a moderate change of spin excitation around (π,π) with weakening charge stripe. We also find that the anisotropy between the $(\pi,0)$ and (π,π) directions becomes stronger with hole doping in an 8×8 *t-t'-J* lattice. In an electron-doped in the *t-t'-J* lattice, we find a clear downward shift in energy toward $(\pi,0)$ [4]. All these calculated behaviors are consistent with the recent RIXS experiment [1-3], indicating that RIXS is a powerful tool to observe both charge ordering and resulting spin dynamics expected from theoretical calculations.

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Unbiased simulations of the Hubbard model and how to see collective charge modes

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It is now well accepted that there are many competing and intertwined phases as "solutions" of the Hubbard model that are separated by very small energies, typically 1/100th of the hopping strength. Therefore, it is important to benchmark using unbiased methods which are the stable phases, and how they can be manipulated. In this talk I will present the results of quantum Monte Carlo and DMRG simulations that explore the relationship between stripes and superconductivity. We can show a strong interplay between CDW and long-range superconductivity that can be controlled by varying the next nearest neighbor hopping t'.

LDA+DMFT approach to calculation of RIXS spectra

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Density functional plus dynamical mean-field theory (LDA+DMFT) has become the method of choice to describe electronic correlations on a material specific level. While the valence one-electron (photoemission) spectra are the natural outcome of DMFT calculations, the method allows in principle access to any response function and its structure makes is particularly well suited for the description to core-level spectra such as XPS, XAS or RIXS. We will review LDA+DMFT results for core-level spectra of selected transition metal oxides such as NiO, Fe2O3, LaCuO3, NaCuO2 [1] or LaCoO3 obtained with our implementation [2,3] based of configuration-interaction solver. We will discuss the nature of Raman-like and fluorescence-like features in the RIXS spectra of the studied materials and their description by Anderson impurity model that sits at the heart of DMFT approach. We will also discuss observation of strong dispersion of spinful excitons in LaCoO3 that was theoretically predicted [4] and recently confirmed by RIXS experiments on Co L-edge [5].

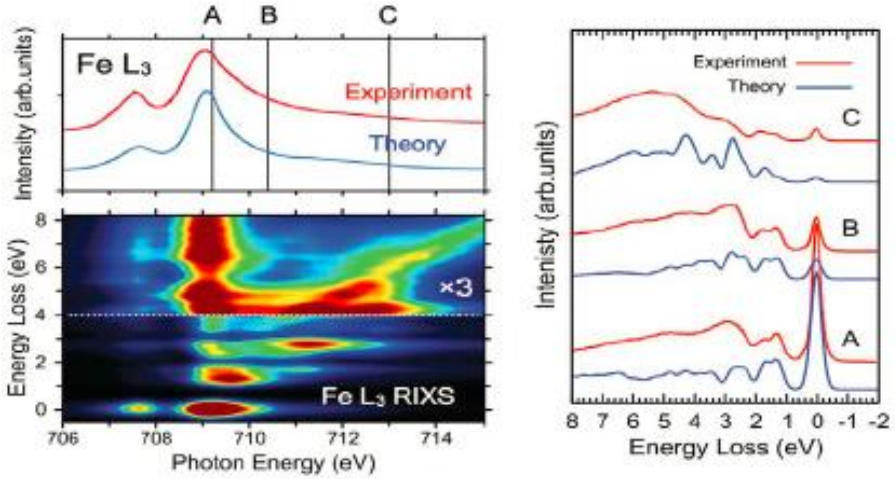

Fig.1 Calculated and experimental spectra of Fe2O3, XAS (top left) and RIXS for several incident photon energies (right). The scan of calculated RIXS spectra over incident photon energies showing the Raman-like and fluorescence-like features (bottom left).

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RIXS for 5d spin-orbit coupled oxides in the strong Mott-insulator limit

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RIXS has been a useful probe of magnetic excitations in spin-orbit coupled solids. In this talk, I will discuss aspects of RIXS for highly-localized electronic states in t_{2g} orbital Mott insulators, where we can model the spectrum at various electron fillings to extract the local spin-orbit coupling and Hund's coupling. Comparing theory with experiments on a wide class of double perovskite materials, we find a consistent description of these parameters in rhenates, osmates, and iridates. This work provides insights into the "lego" building blocks of such oxides, which can be used to construct frustrated fcc magnets with Kitaev interactions and magnetoelastic, and quantum anomalous Hall insulators in ultrathin half-metals.

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Estimation of phonon lifetime in epitaxial films

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Phonon properties in semiconductors have garnered a lot of interest in discussions of the heat properties, since phonon scattering is directly related to thermal properties. Especially, as thermal management of the next-generation devices (such as thermoelectric, high power, and photovoltaic devices) has become significant with continued advances in miniaturization to the nanoscale, the detailed phonon information of these materials is helpful to design the devices with low energy loss. In contrast to Raman scattering and IR spectroscopies, where the phonon information is confined at Γ in reciprocal space, inelastic neutron (INS) and X-ray (IXS) scattering spectroscopies measure momentum-dependent phonon energy and linewidth. The experimentally obtained detailed information can be compared, for example, to the DFT (density functional theory) calculations, that tie the microscopic phonon features to the macroscopic thermal properties.

Utilizing the techniques in grazing incidence X-ray diffraction (GIXRD) measurements, we have developed an IXS technique to measure the phonon dispersion and linewidth in thin films without artifacts from the substrate. Figure 1 shows the IXS spectra of ScN epitaxial film (40 μm thickness) on a sapphire substrate [1]. ScN is a promising thermoelectric candidate among nitride semiconductors. The phonon dispersion curves are clearly visible along the high-symmetry lines (Γ-X and Γ-K-X). The phonon lifetime can be evaluated from the linewidth of optical phonon modes. The obtained phonon dispersion and lifetime, which correspond to the harmonic and anharmonic contributions of phonon, are well reproduced by the DFT calculations. Moreover, the calculated thermal conductivity (43 W/m/K at T=300 K) agrees with the experimentally obtained value (36.4 W/m/K), that lends credence to the discussions of the microscopic (phonon) origin of the thermal properties.

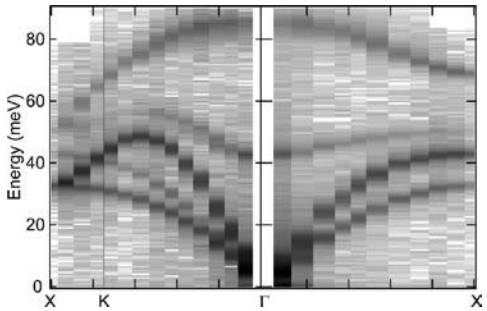

Fig. 1 Observed phonon dispersion curves (black) of ScN epitaxial film

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Spin and lattice correlations in quantum magnets and the full elasticity tensor from thermal diffuse scattering.

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Inelastic x-ray scattering (IXS) with meV resolutions is an ideal tool to study lattice vibrations in crystals. IXS can be also sensitive to other degrees of freedom if strongly coupled to the phonons. One intriguing possibility is when polar phonons couple to spin waves in an ordered magnet. In this presentation, I will show, that the strong magnon–phonon coupling in the triangular lattice Heisenberg antiferromagnets $LiCrO_2$ and $PdCrO_2$ enables the measurement of magnetic correlations throughout the Brillouin zone via IXS. Our studies reveal intrinsic details of the magnetoelastic excitation spectrum. We found single particle excitations with momentum dependent lifetime and continuum scattering at low temperature [1]. In a high-pressure experiment at cryogenic temperatures we furthermore show that tuning the lattice allows for efficient control of magnetic interactions.

The second part of my talk focuses on the quantitative analysis of thermal diffuse x-ray scattering (TDS). I will show that high-precision measurements of diffuse scattering intensities together with a rigorous data analysis allow the determination of the full elasticity tensor in a single crystal diffraction experiment [2]. Our approach enables a reliable and model-free determination of the elastic properties and can be performed together with crystal structure investigation in the same experiment.

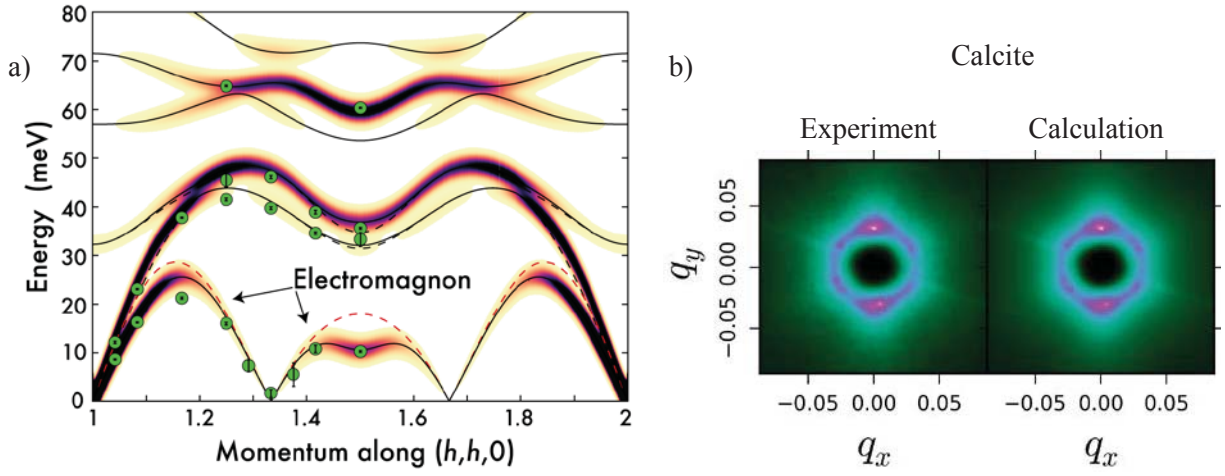

Figure 1. a) Phonon and electromagnon dispersion in $LiCrO_2$ measured by IXS. b) TDS in Calcite, measurement (left) and calculation (right) for the determination of the full elasticity tensor.

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Advanced magnetic spectroscopies for the fine characterization of magnetic nanomaterials

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In this talk I will show how the combination of XMCD and RIXS-MCD spectroscopies allows a deep understanding of the electronic and magnetic structures in complex magnetic nanomaterials such as bimagnetic core-shell nanoparticles and ferrofluids, and reveal emergent properties [1]. Questions related to core-shell interdiffusion and the distribution of magnetic anisotropies inside nanoparticles can be answered from measurements combined to Ligand Field Multiplet calculations and from XMCD / RIXS-MCD-detected magnetization curves [2,3]. In addition, I will discuss the possibilities offered by RIXS-MCD using a new liquid cell dedicated to the *in-situ* measurements of liquid and frozen ferrofluids, which allows preserving the nanoscale magnetic dipole interactions responsible for magnetically driven macroscopic properties used in numerous applications.

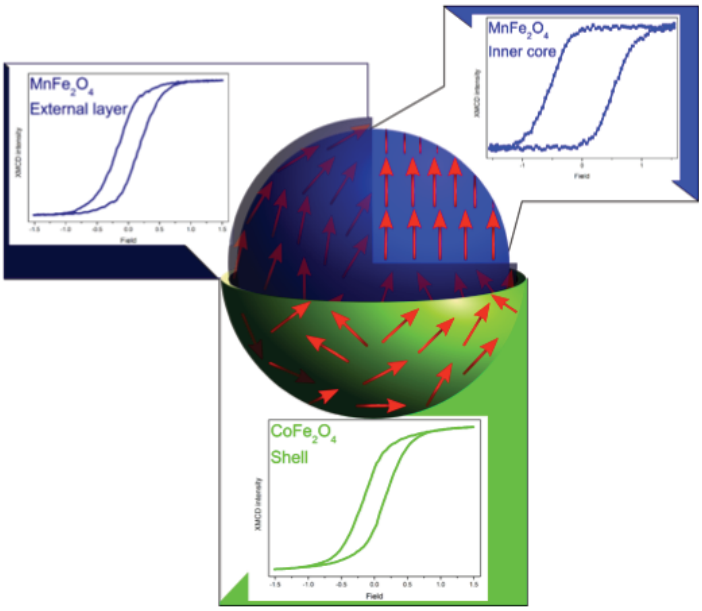

Figure 1: Gradient in magnetic coercivity measured in $MnFe_2O_4$@$CoFe_2O_4$ nanoparticles [3].

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Lattice dynamics of epitaxial strain-free interfaces

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The contact area between two dissimilar materials is very often a source of new physical phenomena that govern many modern technologies such as opto- and nanoelectronics, photovoltaics, and magnetic recording. Unlike the electronic and magnetic properties, which have extensively been investigated and well understood, the propagation of thermal lattice excitations (phonons) along and across epitaxial interfaces remains poorly understood despite decades of research. The importance of the interface lattice dynamics in lattice-matched heterostructures arises from the fact that it is intimately related to physical phenomena such as nanoscale thermal transport and phonon filtering, which are fundamentally important for waste heat harvesting, thermal management in nanoelectronics and are the basis for the development of new logic devices operating at THz frequencies.

To achieve a comprehensive understanding of the interface lattice dynamics, we performed a combined experimental and theoretical study of the strain-free Fe_3Si/GaAs heterostructure [1], which is a widely used model system for the investigation of semiconductor/ferromagnet interfaces for spintronic and magnetoelectronic devices [2]. The Fe-partial phonon density of states (PDOS) of Fe_3Si layers with various thicknesses was obtained from nuclear inelastic scattering experiment performed at room temperature at the Dynamics beamline P01 [3] of PETRA III with an energy resolution of 1.0 meV. The reduction of the layer thickness down to about 0.8 nm leads to a smearing of the phonon features and a drastic enhancement of the low-energy phonon modes. While the first effect arises from phonon confinement in the ultrathin layers, interface roughness and defects, first-principles calculations demonstrate that the observed anomalies at low energy originate from an interface-specific phonon density of states. The interface PDOS stems from the differences in the coordination spheres of the interface atoms compared to those in the bulk, which reduces significantly the atomic force constants of the former [4,5].

The reported findings pave the way for phonon nanoengineering in two-dimensional systems that will have implications on the design of efficient thermoelectric heterostructures and will stimulate further progress in thermal management and nanophononics.

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Magnetic circular dichroism in x-ray fluorescence

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X-ray fluorescence is a very basic phenomenon, in which the atom excited by an x-ray photon emits a secondary x-ray that corresponds to the energy transition between electron orbitals. Since fluorescence energies are element specific, x-ray fluorescence techniques, including x-ray fluorescence microscopy and imaging, are widely applied in trace element analysis.

The state of a photon is characterized by its wavevector and polarization. For x-ray fluorescence photons, however, the second feature has not received particular attention so far. To my best knowledge, there has been limited work reported on polarization analysis of inelastically scattered x-rays [1,2].

The reasons are probably followings; (i) crystal and multilayer optics used in polarization analysis require well collimated x-ray beams, while fluorescence x-rays are emitted in all directions. (ii) in the hard x-ray region, fluorescence x-rays emitted from shallow core or valence levels, which may be influenced by valence electrons or local symmetry, are usually weak.

Recently, I have searched for a new magnetic spectroscopy with a large magnetic effect for 3*d* transition metal elements in the hard x-ray region. Eventually, I planned to measure the degree of circular polarization of the *K*α emission emitted from a ferromagnetic material [3]. In the experiment, a fully magnetized iron single crystal was exposed by synchrotron x-rays. The degree of the circular polarization was evaluated by a diamond phase plate followed by a linear polarization analyzer. The phase plate converted the circular polarization of the *K*α emission to the linear polarization and vice versa. The converted linear polarization was then measured by the linear polarization analyzer, which was a Ge (400) single crystal.

The obtained $K\alpha_1$ spectra are shown in the figure. I^+ and I^- are spectra measured when $+\pi/2$ and $-\pi/2$ phase shifts were introduced by the phase plate, respectively. I^+ and I^- are given by $I_0(1+P_c)/2$ and $I_0(1-P_c)/2$, respectively, where I_0 is the total intensity and P_c is the degree of the circular polarization. P_c was 12% at 6.405 keV and would amount to 18% if corrections are taken into account. Large magnetic dichroism was, therefore, obtained in intense *K*α emissions. This is a new magnetic spectroscopy in the x-ray region and also a new magnetoptical effect in the x-ray region, which is called x-ray magnetic circularly polarized emission.

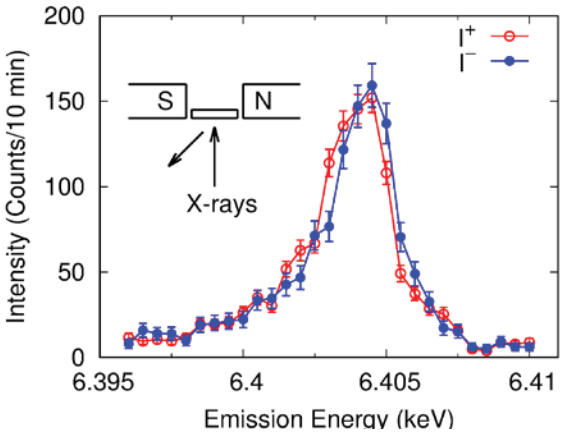

Fig. 1. Fe $K\alpha_1$ emission spectra I^+ and I^- separated by circular polarization analysis.

In the talk, I will show several fluorescence spectra calculated for a rare-earth element and will describe a new, highly efficient spectrometer with a collimating optics (Montel mirror). As a preliminary result, the magnetization map of an electrical steel sheet will also be presented.

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Resonant inelastic EUV scattering at a seeded free electron laser

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Free electron lasers promise to extend the resonant inelastic scattering to the ultrafast time domain offering the unique opportunity to investigate the dynamics of site and energy resolved elementary excitations of condensed matter. Here I will present recent pump probe and time resolved RIXS results about the dynamics of the orbital excitations of the quasi-one-dimensional $CuGeO_3$ obtained at the EUV M_{23} edges.

Ultrafast time-resolved x-ray scattering reveals diffusive charge order dynamics in $La_{2-x}Ba_xCuO_4$

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Charge order is universal among high-T_c cuprates but its relation to super- conductivity is unclear. While static order competes with superconductivity, dynamic order may be favorable and even contribute to Cooper pairing. Using time-resolved resonant soft x-ray scattering at a free electron laser, we show that the charge order in prototypical $La_{2-x}Ba_xCuO_4$ exhibits transverse fluctuations at ps timescales [1]. These sub-meV excitations propagate by Brownian-like diffusion and have an energy scale remarkably close to the superconducting T_c. At sub-meV energy scales, the dynamics are governed by universal scaling laws defined by propagation of topological defects. Our results show that charge order in $La_{2-x}Ba_xCuO_4$ exhibits dynamics favorable to the in-plane superconducting tunneling, and establish time-resolved x-rays as a means to study excitations at energy scales inaccessible to conventional scattering techniques.

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Time-resolved Resonant Inelastic X-ray Scattering on Quantum Materials

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Resonant Inelastic X-ray Scattering (RIXS) is a powerful probe of excitations from the electronic ground state of correlated materials involving lattice, charge, orbital and spin degrees of freedom. Owing to its momentum sensitivity, RIXS has evolved as an important spectroscopic tool for accessing the dispersion of collective excitations in a diverse set of quantum materials. During the last years this advanced spectroscopy has been extended to the time-domain, in particular in experiments following pump-probe schemes. In this talk we illustrate the scientific capabilities of time-resolved soft X-ray RIXS by summarizing the results obtained at SXR/LCLS during recent experiments on the prototypical Mott-Hubbard material V_2O_3 and the Spin-Peierls spin-chain $CuGeO_3$.

V L_3-edge RIXS experiments unveil the ultrafast dynamics of the orbital structure in V_2O_3 after transiently inducing an insulator-metal transition (IMT) by an fs laser excitation with an 800 nm pump. The probed orbital excitations within the d-shell of the V ion show an ultrafast sub-ps time response, followed by a transient recovery at intermediate (ps) timescale and a slower thermal-like IMT. By irradiating $CuGeO_3$ with an ultrashort laser pulse of 266 nm wavelength, we excited the electronic system across the charge transfer gap, thereby disturbing the antiferromagnetic correlations by removing spin-1/2 holes from the Cu sites. By monitoring the evolution of particular non-local charge transfer excitations, the Zhang-Rice singlet excitations, [1] probed with O K-edge RIXS as a function of time delay between pump and probe pulses, we observed ultrafast depletion of the short-ranged antiferromagnetic correlations within 2 ps, after which a slow recovery dynamics sets in persisting over several 100 ps depending on pumping fluence.

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Tracing the active atomic sites in photochemistry with soft X-ray resonant inelastic X-ray scattering

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Active atomic sites in photochemical processes are a central conceptual approach onto which governing principles of rate and selectivity are based. The optical radiation is behind both the desired processes of charge separation and photochemical heterogeneous catalysis but also requires efficient mechanisms of photoprotection to shut off unwanted radiation damage. Since they are highly disperse within the molecular moiety itself, but even so in the typical solvation environment, their observation requires ideally exquisite selectivity of the orbital based valence electronic structure at these active sites in real time only possible with femtosecond and picosecond FEL and Synchrotron soft X-rays. Having established the fingerprints of time resolved resonant inelastic X-ray scattering (RIXS) recently [1,2] a series of deep atomic insights to photochemical processes has been achieved: For saturatively coordinated transition metal complexes in heterogeneous catalysis, the competition between ultrafast intramolecular spin relaxation and solvent induced scattering in ligand substitution is established [3,4]. For photo-driven charge separation in spin-cross over complexes the additional importance of σ - π valence interaction is established [5,6,7]. Finally, the highly selective bond elongations in photoprotection through tautomerization has been observed and quantified [8,9]. An outlook is given, how at European XFEL time resolved RIXS at the transform limit (HeisenbergRIXS consortium) now makes these approaches available more widely.

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Dynamics study in time scales between nano-second and micro-second by quasi-elastic scattering based on synchrotron radiation

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Observation of microscopic dynamics in time scales between nano-second and milli-second is quite difficult by X-ray scattering and even neutron scattering experiments. In this presentation, we introduce quasi-elastic scattering technique using gamma rays with neV-energy resolution. The technique allows us for studying microscopic density fluctuations in time scales between nano-second and micro-second.

^{57}Fe nuclei can be excited to 14.4-keV energy level by synchrotron radiation. By the Mössbauer effect, the excited ^{57}Fe nuclei emit photons with neV-energy width, which is called as Mössbauer gamma rays. The Mössbauer gamma rays show high directivity as incident synchrotron radiation and are used for the probe beam of the unique quasi-elastic scattering experiment. By constructing so-called time-domain interferometer, microscopic dynamics is measured in time domain through observation of the intermediate scattering function. [1] Recently, we developed advanced time-domain interferometry using the Mössbauer gamma rays which show multi-peaks in energy spectra. [2] In addition, we considered an effect of a finite bandwidth of the incident synchrotron radiation on the gamma-rays spectrum. [2] These improvements largely increase measurement efficiency and help to construct really useful system for application studies. We introduce current status of quasi-elastic scattering technique using the Mössbauer gamma rays.

In the time scales between nano-second and micro-second, various important relaxation processes occur in condensed matters; however, these microscopic mechanisms still remain unsolved because, in the time scales, (i) direct observation of microscopic motions is very difficult, (ii) molecular dynamics simulation studies are still hard due to it large calculation cost, and (iii) often many-body relaxation processes, which make theoretical studies extremely difficult, play central roles. Therefore, direct microscopic dynamics study in the time scales is very challenging and is now feasible by using the quasi-elastic scattering technique using the Mössbauer gamma rays.

In the presentation, we also introduce some application results obtained so far. By cooling glass formers, the structural α-relaxation process starts to show many-body relaxation property, and additional local activated processes starts to be observed when the α-relaxation time reaches around 100 nano-second. The mechanism of the dynamical change in deeply supercooled glass formers is still not well-known despite its essential relation to nature of glass transition. The present technique showed a unique property of the local process and its relation to the structural process. Similar local activation processes were found to be elemental processes of the protein folding process by previous studies suggesting that the nano-second and micro-second dynamics is also essentially important in soft matters. We also introduce our result on soft matter systems. In smectic phase of liquid crystals, which shows a nanometric layered structure, we found the molecular translational motion across the layer occurs in a time scale of 100 nano-second. Such interlayer molecular motions were too slow to be observed by quasi-elastic neutron scattering due to limitation of its energy resolution.

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Nanoscale lattice dynamics probed by extreme ultraviolet transient gratings

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The availability of fully coherent extreme ultraviolet (EUV)/soft x-ray ultrafast pulses at the FERMI free electron laser (FEL) facility has permitted to develop the four-wave-mixing approach in a transient grating configuration [1,2]. Such a novel experimental tool allows probing several kind of dynamical processes and has been further developed with the implementation of FEL-pump/FEL-probe capabilities [3]. This enables access to the *mesoscopic* (10s of nm) length-scale range, which is hardly accessible by other means.

In the present contribution we present these new experimental capabilities, showing evidences for phonon and thermal transport dynamics in some prototypical samples (amorphous silicon nitride, crystalline silicon and silicon carbide, and an amorphous metallic alloy) down to 24 nm wavelength [4-6], corresponding to thermal transport distances shorter than 10 nm. In this regime we found that the thermal response of the crystalline samples shows a strongly non-diffusive behavior, while in amorphous silicon nitride the thermal transport process is still compatible with a diffusive trend.

We finally discuss on the potential that the EUV/soft x-ray TG approach holds in other fields, as for instance in the study of mesoscopic structural relaxations in liquids and ultrafast magnetic dynamics at the nanoscale. Furthermore, these are the first time-resolved four-wave-mixing experiments carried out exclusively using EUV pulses, hence representing an advance in nonlinear optics and potentially paving the way to a wider class of experiments, so far conceived only theoretically [7].

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Evolution of the spin and charge excitations in high T_c cuprates

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Spin fluctuations are considered as one of the essential ingredients for the pairing mechanism of high-temperature superconductivity in cuprates. From resonant inelastic x-ray scattering (RIXS) study we found that the larger long-range exchange interaction can lead to a higher $T_{c,max}$ [1]. Upon doping, neutron scattering experiments have indicated that the long-range spin correlations weaken significantly [2], whereas x-ray scattering experiments have found that high energy magnetic excitations persist across the entire superconducting dome [3,4]. To reconcile these two apparently incompatible experimental facts, I will present our comprehensive RIXS study of the doping and momentum evolution of magnetic excitations in the cuprate family $(Bi,Pb)_2(Sr,La)_2CuO_{6+\delta}$ [5]. We find that the short-range magnetic interaction is little affected within a large momentum region of Brillouin zone, but the collective spin excitations become increasingly decoherent and eventually loses their propagating character. Our results demonstrate that the relevance of spin fluctuations to superconductivity is momentum dependent and maximal near the antiferromagnetic point.

Charge density wave is a common feature of cuprate superconductors, but its relevance to superconductivity and the underlying origin remain unclear. I will present our recent RIXS study taken at SIX beam line at NSLS2 on the charge density waves in the stripe-ordered cuprate $La_{2-x}Eu_{0.2}Sr_xCuO_4$ and discuss about their interplay with phonons [6].

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Ubiquitous Charge Density Wave Excitation and Local Commensurability in Underdoped Cuprates

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Charge density waves (CDWs) are now established as a key component of the underdoped cuprate superconductors. While accumulating experimental evidence reveals a ubiquitous CDW dome centered near 1/8 doping in the phase diagram, a unified description of CDWs has not been reached due to different doping-dependence of CDW wavevector in different cuprate families. Here, we describe our recent inelastic x-ray scattering studies of canonical CDW system $La_{2-x}Ba_xCuO_4$ (x = 0.115 − 0.155) [1-3]. We discovered that the CDW develops in two stages with decreasing temperature. A precursor CDW with near-commensurate wavevector emerges first at high-temperature. This doping-independent precursor CDW correlation originates from the CDW phase mode couples with phonon excitations and "seeds" the low-temperature CDW with a strongly doping dependent wavevector. Our observation reveals the locally commensurate precursor CDW and its phase mode as the common thread of underdoped cuprates with important implications for the highly intertwined electronic ground state.

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Probing CDW phenomena and charge excitations in cuprates via RIXS

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Characterizing low energy excitations, in particular those associated with phonon and charge degrees of freedom, are essential to understand the rich phenomena in cuprate superconductors. In this presentation, I will first highlight our recent RIXS studies on the CDW phenomena in Bi-based double-layered cuprates. Intriguing interplay between CDW, CDW excitations, and phonon excitations as a function of temperature and doping were observed and will be discussed [1, 2]. In the second part of my talk, I will also highlight the observation of rapidly dispersed charge excitations in the electron-doped cuprates, which bear signature of plasmonic behaviours in layered 2D systems [3].

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Uniaxial pressure control of competing orders in the cuprates

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External control of electronic phases in correlated-electron materials is a long-standing challenge of condensed-matter research. Layered cuprates exhibit antiferromagnetic, charge-density-wave (CDW), and high-temperature superconducting ground states which can be tuned by doping and external magnetic fields. However, disorder generated by lattice defects and randomly pinned magnetic vortices greatly complicates the interpretation of these experiments. Here, we report a high-resolution inelastic x-ray scattering study of the high-temperature superconductor YBa2Cu3O6.67 under uniaxial stress and show that a three-dimensional long-range-ordered CDW state can be induced by pressure along the a-axis, in the absence of magnetic fields. The amplitude of the CDW is strongly suppressed below the superconducting transition temperature, indicating strong thermodynamic competition with superconductivity. We also show that the transition is driven by the complete softening of an optical phonon mode. The results provide new insights into the anomalous normal-state properties of high-temperature superconductors an illustrate the potential of uniaxial-pressure control of competing orders in quantum materials [1].

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High-resolution momentum-resolved RIXS at Taiwan Photon Source

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In this talk, we will present the commissioning status and the performance test of a new soft X-ray RIXS beamline at Taiwan Photon Source. This new RIXS setup exploits the energy-compensation principle of grating dispersion to improve its measurement efficiency. Its vertical focusing mirror and both varied-line-spacing gratings are incorporated with a special bender, permitting the adjustment of curvature and local profile in an active manner to eliminate the effect of thermal deformations. In addition, both the monochromator and the spectrometer gratings are equipped with a long trace profiler for measurements of the grating figure. Currently we have achieved a total energy resolution better than 25 meV at the O K-edge. Several examples of high-resolution momentum-resolved RIXS measurements will be prersented.

Resonant Inelastic X-ray Scattering at SIX of NSLS II:

An update on the beamline status and its performances

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The Soft Inelastic X-ray scattering beamline SIX at NSLS II, designed to achieve the ambitious combined resolving power of R~ 70'000 [1, 2], is currently operational and open to users delivering the possibility of performing resonant inelastic x-ray scattering experiments with a final resolving power around R~ 20'000.

In the first part of the talk we will provide an update on the beamline status and on the Centurion spectrometer performances, with reference to the results achieved so far and to the available experimental conditions.

In the second part of the talk, we will present the commissioning path undertaken to reach the design-performances of SIX, the planned instrumentation upgrades and the active scientific projects.

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High energy resolution RIXS study on orbital ordered $KCuF_3$

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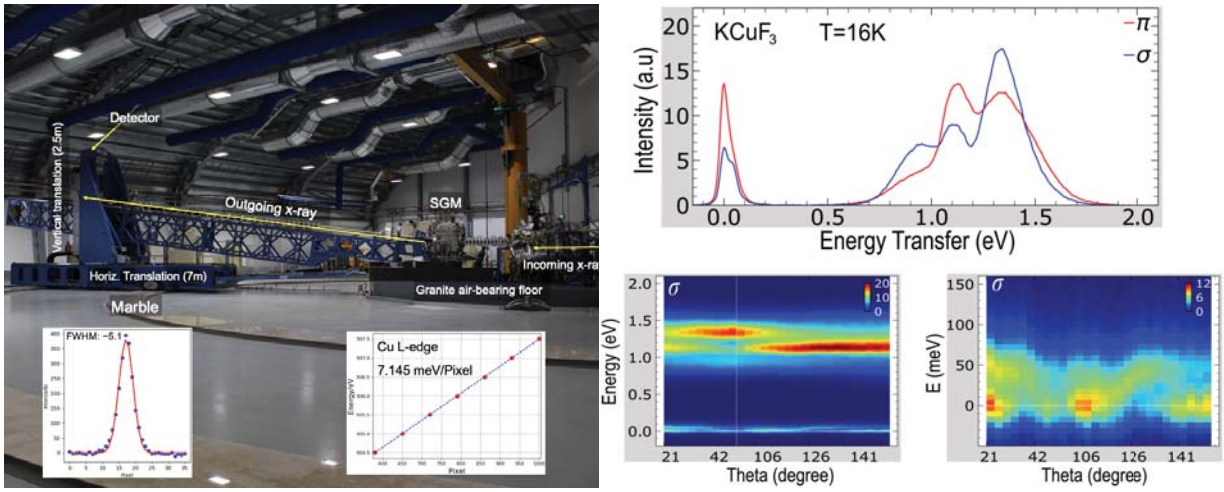
The I21 beamline is a dedicated Resonant Inelastic soft X-ray Scattering (RIXS) facility at Diamond Light Source in the UK. It covers an energy range from 250 to 1500 eV with a planned upgrade up to 3000 eV. Through the early X-ray commissioning and user operations, we obtained the energy resolution of about 35 meV at Cu L-edge (930 eV) and about 15 meV at Oxygen K-edge (532 eV) with high photon flux. Key technical achievements of the beamline will be presented.

As one of science examples, we performed RIXS study on orbital ordered $KCuF_3$. The high-temperature orbital order in $KCuF_3$ is an intriguing but long-debated phenomenon. Two possible underlying mechanisms were proposed: cooperative Jahn-Teller distortion and Kugel-Khomskii orbital superexchange. The Jahn-Teller distortion often yields local excitations while the superexchange scenario is signified with collective orbital excitations. With unprecedented energy resolution, orbital excitations among different Cu^{2+} 3*d* orbitals are resolved and compared with the *ab initio* calculation. At the low energy side, excitations below 100 meV present clear dispersion. We will discuss the key results in connection with spin and orbital physics in $KCuF_3$.



Opportunities for resonant elastic and inelastic x-ray scattering studies at the SCS instrument at the European XFEL

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European XFEL is a free-electron laser user facility located in Schenefeld, Germany, providing x-ray FEL pulses of extreme peak intensities. The unique features of European XFEL radiation are its high average brilliance, femtosecond pulse duration and high degree of coherence.

The Spectroscopy and Coherent Scattering (SCS) scientific instrument at European XFEL is located at the soft x-ray SASE3 source [1]. It aims for time-resolved spectroscopic and structural studies of strongly correlated and functional materials. It will implement among others the techniques of x-ray absorption spectroscopy (XAS), resonant inelastic x-ray scattering (RIXS), x-ray resonant diffraction (XRD) and coherent diffraction imaging (CDI) with two baseline setups for solid samples: XRD (optimized for reflection/backscattering geometries) and FFT (forward-scattering geometries). Moreover, the SCS instrument will host the User Consortium Heisenberg RIXS (hRIXS) spectrometer and an additional setup for liquid-jet chemistry experiments.

The hRIXS spectrometer is designed for high energy-resolution RIXS and flexible sample environments. At the SCS beamline it will enable momentum- and time-resolved RIXS studies close to the Heisenberg time-energy transform limit, which will open up a route to follow the dynamics of complex systems (solid and liquid) on their relevant momentum, energy and time scales.

The SCs instrument has started user operation in the fall of 2018 and the first successful experiments have been performed with the FFT setup. The XRD setup and hRIXS will open for users in 2020 [2].

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Soft X-ray scattering instrumentation at LCLS-II

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When fully commissioned LCLS-II is projected to deliver average flux 3-to-4 orders of magnitude higher than LCLS, courtesy of the superconducting accelerator technology, allowing the repetition rate to increase from 120 Hz up to 1 MHz. The NEH 2.2 beamline will deliver monochromatic, near-transform-limited femtosecond pulses suitable for a wide variety of ultrafast pump-probe studies. Time-resolved Resonant Inelastic X-ray Scattering (RIXS) is particularly well-suited for FEL-based experiments. While today most of the studies have been focused on demonstration experiments, limited by the low average flux, LCLS-II will bring transformational capabilities.

In this talk I will give a brief overview of our plans for the monochromatic soft x-ray beamline (NEH 2.2), including beam delivery and 2 endstations focusing on RIXS experiments: chemRIXS, targeting dilute systems in solution phase where moderate resolution and high throughput are needed, and qRIXS addressing the needs for a high-resolution, *q*-resolved RIXS of correlated material systems.

Time permitting, I will present 2 relatively new methods for collecting RIXS spectra: Transition Edge Sensors (TES), and Photoelectron Analysis of X-rays (PAX). While still in R&D phase these methods could provide exciting new alternatives to existing instrumentation.

I will conclude by presenting the projected timeline for machine operation at LCLS and the targeted dates for delivery of fully commissioned instrumentation.

High spectral and focal resolution RIXS with state-of-the-art flat crystal spectrometer

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State of the art resonant inelastic x-ray scattering (RIXS) spectrometer based on x-ray collimation and analysis features high spectral and focal resolution [1,2]. In this presentation, we introduce flat-crystal RIXS spectrometer with collimating mirror. We present measurements of magnon gap in Sr_2IrO_4 and magnetic quasiparticle excitation in Na_2IrO_3 using the 10 meV spectral resolution. In addition, we present scattered polarization analysis with high-resolution and high-efficiency which was carried out for the Majorana fermion scattering [3]. Further improved 6 meV spectral resolution measurement is shown, verifying that Quartz crystal quality can accommodate few meV spectral resolution. As a case of other absorption edges, 12 meV spectral resolution was achieved for the Ta L_2 edge. Measurement on Weyl semimetal TaAs is presented [4]. The collimating mirror limits the spectrometer field of view to less than 100 um, which is advantageous for probing low-lying excitations in a high pressure environment [5]. The flat crystal measurement on a sample in the high-pressure cell is compared with the diced, spherical analyzer measurement on the same sample, demonstrating that extrinsic scatterings from high pressure environments are mostly avoided in the flat crystal spectrometer.

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IRIXS: an intermediate x-ray energy RIXS spectrometer at P01, DESY

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The need to access the $L_{2,3}$ absorption edges of the chemically important 4d elements has driven most synchrotrons to develop x-ray instruments operating in the intermediate (tender) x-ray range (1-5 keV). Although techniques requiring moderate/low energy resolution (i.e. REXS and XAS) are readily available to users, major hurdles remain for methods relying on higher resolving power (i.e. RIXS). In this talk I will present an ongoing project at Petra III DESY where a collaboration between the Max-Planck Institute in Stuttgart and beamline P01 was established in order to create the first intermediate x-ray energy RIXS spectrometer (IRIXS). Our IRIXS instrument covers the $L_{2,3}$ absorption edges of Ru (2.840 and 2.970 keV), representing a huge scientific opportunity to look at the intriguing ruthenates (e.g. p-wave superconductivity in Sr_2RuO_4, Kitaev physics in alpha-$RuCl_3$, and excitonic magnetism in Ca_2RuO_4). I will discuss the technical challenges introduced by the use of intermediate x-rays as well as presenting the current status of our instrument. Finally, I will address the prospects of reaching resolution comparable to the Cu and Ir L_3-edge RIXS.

The Inelastic X-ray Scattering beam line ID20 at the ESRF

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Conceived and constructed in the framework of the ESRF Upgrade Phase I project, ID20 is a state-of-the-art hard inelastic X-ray scattering beam line dedicated to the study of electronic and magnetic excitations in condensed matter systems. We are interested in the investigation of strongly correlated electron systems, functional- and energy-materials, electronic excitations under extreme conditions, as well as the study of chemical reactions in liquids and gases.

The beam line hosts two spectrometers, one of which is optimized for studies with energy resolution up to 25 meV as often used for resonant inelastic X-ray scattering experiments of 5d element containing strongly correlated materials [1], and one end-station dedicated to studies with mediocre energy resolution yet processes with small cross sections such as non-resonant inelastic X-ray scattering from core excitations [2].

Here, we will present the instruments and show examples of recent studies that exploit the unique capabilities of the beam line and its end stations.

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Ultra-high-resolution Compton Scattering

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Compton scattering is an inelastic process in the very high ω and q transfer region. An electron acquires large kinetic energy from photons, then exiting the sample and behaving as a plane wave. The energy scale of recoiled electrons is much larger (~keV) than that in the initial states (~eV). This fact often allows us to neglect electron binding energy, so that the so-called impulse approximation (IA) is fulfilled. The distribution in p-axis in the initial state is projected onto ω-axis in the final state, meaning that the energy distribution of scattered photons represents the electron momentum distribution, involving information of Fermi surfaces and correlation effects.

For the unit for the momentum p, the atomic unit (au) is widely used, where p is indicated by a wavenumber k (= $p/\hbar$) multiplied by Bohr radius a_0 (1 au = 1.89 A^{-1} = 1.99 x 10^{-24} kg·m·s^{-1}). The momentum resolution of Compton scattering is commonly not so high: General high-resolution Compton scattering only provides a 0.1 – 0.2 au resolution while a radius of Fermi sphere or a size of the first Brillouin zone is typically ~1.0 au.

If x-ray Raman spectrometers are utilized, one could easily get a ~0.01 au instrumental resolution but the problem is that they are commonly optimized for x-rays ≤ 10 keV. The error of IA becomes significant in such an energy region, smearing out the most of fine features. A true high-resolution experiment requires higher energy x-rays (ideally, as high as 50 keV) so as to make the final states effect negligible.

In the presentation, ultra-high resolution Compton scattering studies will be discussed. Using x-ray Raman spectrometer optimized at 25.5 keV, we obtain an instrumental resolution of 0.016 au while an estimated final state effect 0.013 au, providing the overall resolution is 0.024 au. Two experiments will be discussed: (i) Momentum distribution near Dirac cone in graphene and graphite [1] and (ii) Renormalization of momentum distributions in liquid Li [2].

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Recent Progress in Non-Resonant Inelastic X-Ray Scattering

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Recent progress in inelastic x-ray scatting as carried out at RIKEN's Quantum NanoDynamics Beamline (BL43LXU) of SPring-8 will be discussed [1]. Several upgrades to the IXS program will be mentioned including practical sub-meV resolution [2], measurements at very low momentum transfers using analyzer masks and a Soller Slit [3], measurement of bulk liquids at temperatures approaching 2000K using carbon based heaters [4], installation of a new KB system that efficiently provides a 5 micron focal spot [3], use of a Soller screen for background reduction in measurements from a DAC [3] and a high-flux medium resolution setup [5]. Some specific science examples will be discussed, with a probable focus on measurements of the vibron in liquid hydrogen [6].

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Ex-situ and in-situ scattering and spectroscopic study of oxygen evolution catalysts

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Non-noble-metal, thin-film oxides are widely investigated as promising catalysts for solar to fuel conversion devices relying on catalysts of the oxygen evolution reaction (i.e., $2H_2O \rightarrow O_2 + 4H^+ + 4e^-$) mediating four protons and four electron oxidations of waters to dioxygen. To develop highly performative catalysts, structural dynamics and electronic state, and reaction mechanism underlying solar to fuel catalysis must be understood. In this presentation, recent studies will be presented by utilizing multimodal approaches of *ex-situ* and *in-situ* x-ray scattering (PDF, RIXS) /spectroscopies (XAS, RXES, Raman) on oxygen evolving amorphous thin films (cobalt and iridium oxygen evolving catalysts) and their molecular and heterogeneous analogue and difference.

High-Energy X-ray Compton Scattering for Non-destructive and Quantum Characterization in Batteries

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X-ray Compton scattering has become as a unique technique to characterize energy materials [1-7]. An advantage of this technique is the high penetration power in materials of high-energy X-rays (over 100 keV) used in the measurements. Therefore, it enables to visualize lithium reactions and to monitor lithium concentration on real batteries nondestructively. Moreover, since the Compton profile, obtained from this technique, is directly related to the wavefunction of the electrons, by comparing the experiment with first-principles simulations, we can extract the orbital characters and the occupation numbers of chemically active electrons. The orbital characters and the occupation numbers provide a fundamental understanding of electrode reactions. In this talk, we present our recent studies for a commercial coin-type lithium rechargeable battery VL2020 and for the positive electrode material $Li_xMn_2O_4$. The Compton scattering experiment was carried out at BL08W of SPring-8.

In the studies of a VL2020 coin battery, we developed a quantitation method of lithium from the line-shape of Compton scattered X-ray energy spectrum and we successfully obtained the lithium concentration at the positive and the negative electrodes during a cycle. Moreover, by comparing the experimental spectrum and calculated one, we can reveal phases in the electrodes responsible of the battery degradation.

In the studies of $Li_xMn_2O_4$ positive electrode material, by comparing the experiment with first-principles calculation, we clarify that the redox orbital involved in lithium insertion and extraction process is mainly oxygen 2*p* orbitals. We also found that this material has a ferrimagnetic phase, which is stable for slightly non-stoichiometric $Li_xMn_2O_4$ samples. This phase is beneficial for the stability of the battery since the it reduces the octahedral distortion of the $Li_xMn_2O_4$ positive electrode material. The above methodology based on quantum mechanical approach paves the way for advanced characterization of the energy materials.

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The Evolution of Electronic Complexity in Biology: 2p3d and 1s3p RIXS of Iron Sulfur Clusters

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Iron sulfur proteins are ubiquitous in nature, performing essential roles in electron transfer processes, redox chemistry, regulatory sensing and catalysis. The metal active sites of these proteins range from simple single iron sites to complex eight iron clusters. Perhaps the most complex iron sulfur cluster that has been identified to date is the iron molybdenum cofactor (or FeMoco) of nitrogenase, which is capable of cleaving the strong triple bond of dinitrogen. The fundamental question that arises is how does nature evolve complexity in order to enable challenging transformations? In our view, a deeper understanding of the complex geometric and electronic structure of iron sulfur clusters requires the pursuit of novel experimental approaches for integrating their electronic structure in a detailed and quantitative fashion. To this end, we are applying both 2p3d and 1s3p resonant inelastic X-ray scattering (2p3d RIXS), in order to obtain deeper insights into the electronic structure of these important clusters. These data provide an experimental measure of the d-d transitions and allow for more detailed insights into the nature of the multiplet structure. The utility of these methods for understanding the electronic structure of nitrogenase will be highlighted. The challenges that RIXS spectroscopy presents for theoretical modeling will also be discussed.

NRIXS studies of halide perovskites

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Halide perovskites [1] is a class of new optoelectronic materials for photovoltaics, photodetectors, phototransistors, light emitting diodes, lasers, etc. They are semiconductors with rich and complex evolution of structure and dynamics, where dynamic instabilities, electron-phonon coupling, anharmonic effects, and unusual thermal behaviours are implicated.

NRIXS (Nuclear resonant inelastic x-ray scattering) [2] is a unique synchrotron radiation method to study lattice dynamics and thermodynamics. The result from NRIXS can also be used to validate materials simulations, as demonstrated in the applications of NRIXS in biophysics and catalysis researches.

We have for the first time applied NRIXS method to study this class of materials via Sn-119 nuclear resonant scattering. Here we present NRIXS measurements and results for inorganic tin halide perovskites and related compounds [3]. Lattice dynamics properties are inferred and discussed.

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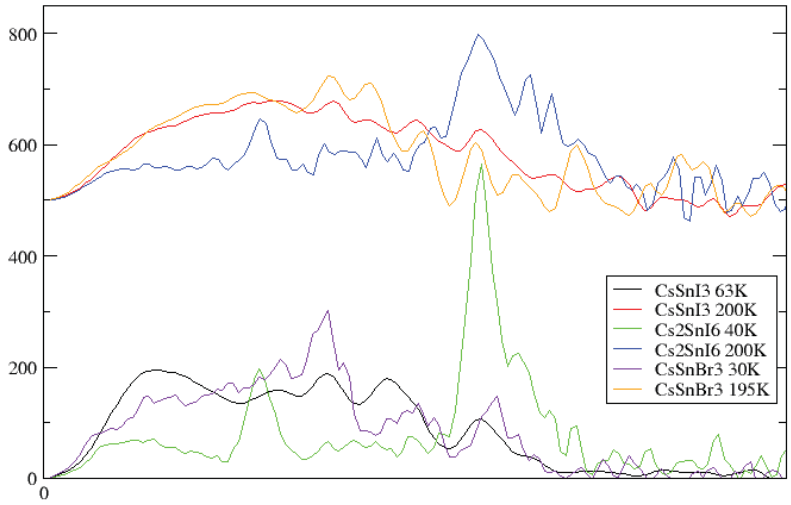

What experiments can we do with 2p3d RIXS?

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In this contribution I give an overview of the progress in 2p3d RIXS experiments. The energy ***resolution*** has been improved to 20 meV which allows accurate determinations of energy dependent phonon, multi-magnon, electronic and charge transfer excitations.

The use of ***angular variations*** allow a large range of different experiments including
(a) the variation of the detection angle between forward (or backward) scattering, 90 degrees horizontal, 90 degrees vertical and all intermediate angles, These experiments can be performed on non-oriented or oriented samples.
(b) variations in the momentum transfer in (θ,2θ) experiments on an oriented sample, for example a single crystal. This allows the detection of the dispersion of magnons and phonons, albeit that the q-range is limited by the x-ray energy. Detection of the polarization dependence of the emitted photon further enhances the information.
(c) azimuthal angular dependence can reveal the combined effects of the exchange interaction, the spin-orbit coupling and the specific (distorted) crystalline site on the angular momentum.
Angular dependent experiment can be combined with the use of ***circular dichroism*** that allows the site and valence specific measurement of the orbital moments, where due to the possibility of specific enhancements, 2p3d RIXS is more sensitive as 2p XMCD.

In addition to high-resolution experiment, ***medium resolution experiments*** can provide useful additional information. For example, the use of efficient x-ray emission detection schemes, both low-resolution monochromators or bolometric detection, will make 2p3d RIXS experiments applicable to low concentration samples. For example metal centers in proteins and other metal impurities. Such experiments would also be useful for ***operando experiments***. RIXS is a photon-in photon-out experiment and as such it should be ideal for operando experiments on systems in action. Applications include batteries, heterogenous (electro)catalyst and other energy systems. The measurement of the full 2p3d RIXS plane with medium resolution can reveal details regarding the specific radiative and non-radiative core hole decay mechanisms, including a better understanding of the super Coster-Kronig Auger channel. Related issues are the study of (inverse) partial fluorescence yield and pre-resonance excitations in relation to a better understanding of saturation and self-absorption effects.

A possible extension of RIXS using diffraction limited lightsources is the combination with ***microscopy***. For example studying the angular dependence and/or circular dichroism can enhance the information on the magnetic ordering with respect to x-ray PEEM experiments.

Femtosecond RIXS experiments are possible at x-ray FEL sources. Time-resolved RIXS has a large advantage over time-resolved XAS. Time-resolved XAS is (as yet) not very specific regarding the nature of the transient states at different time delays. RIXS can greatly enhance the details of the electronic structure of the transients, for example by measuring the dd-excitations at a few x-ray excitation energies.

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Directional cuts through potential energy surfaces

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Potential energy surfaces determine the energy of a system in terms of its structure, determining material functionality. In a multidimensional system the potential energy surfaces of the electronic groundstate rationalize how distortions along defined degrees of freedom connect structural information and conformation to energy. To access the local potential energy surface at an atomic center is now possible using sub-natural linewidth RIXS. It is demonstrated how this technique is used to extract ground state local potential energy surfaces along different molecular coordinates.

Here, present gasphase water is presented as a showcase of how to extract the potential energy surface of the electronic ground state along different molecular coordinates and compare to the case of liquid water where the hydrogen bond network adds environmental effects.

The instrumental developments in high resolution RIXS are accelerating and will offer broader application of the presented approach to access collective phenomena in functional molecules or high-temperature superconductors.

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High-resolution Inelastic X-Ray Scattering as a probe of the dynamic in complex and hybrid materials*

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One of the most exciting opportunities disclosed by modern Nanotechnology is the opportunity to grow complex structures effectively interacting with high frequencies (terahertz) phonons. Indeed, phonons of this frequency are the main conveyors of heat flow in insulators and their control is of paramount importance to implement heat management based upon nanostructure design. To shed further light into this topic, we used Inelastic X-Ray Scattering to investigate the phonon propagation in various prototypical soft mater systems. Samples include liquid crystals in various mesogenic phases[1], suspensions of nanoparticles[2] and carbon nanotubes[3]. Results obtained point towards the possibility of effectively manipulating sound propagation at nanometer and picosecond scales, which might inspire new avenues in next generation phononics.

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Inelastic X-ray scattering at extreme conditions and its geophysical applications

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Density and sound velocity are important observational data for the Earth's interior. The density of the Earth's inner core is smaller than that of hcp-iron nickel alloy. Therefore, the core may be composed of metallic iron-nickel alloy with small amounts of light elements such as Si, C, S, H and other elements. However, the lack of the precise sound velocity data of the core forming materials under the conditions of the core makes it difficult to clarify the chemical and physical characteristics of the inner core. Therefore, we applied inelastic X-ray scattering technique to measure the sound velocity of various iron-light element alloys and compounds relevant to the Earth's core at extreme conditions [1-5].

We measured the compressional velocity at high pressure and temperature by using the inelastic X-ray scattering combined with a double sided laser heated diamond anvil cell [6] at the beamlines BL35XU [7] and BL43XU [8] of SPring-8. We developed a portable double sided laser heating and temperature measuring system for the diamond anvil cell, COMPAT [6], to be installed at the IXS beamlines, which enabled us to conduct high temperature experiments to 3000 K at ultrahigh pressure when coupled with a beam focused by KB Mirrors [9].

We have determined the compressional velocity of the polycrystalline hcp-iron up to 163 GPa and 3000 K [1], which is the highest pressure and temperature conditions ever achieved by using IXS method. Additionally, we measured the compressional velocities of hcp iron-silicon and iron-nickel alloys, and iron-light element compounds such as Fe_3C [2], Fe_3S [3], and FeH [4] at high pressure and temperature. Based on these sound velocities of the candidate materials of the inner core, we can estimate the composition of the inner core relevant to the seismological observations. We also determined the sound velocity of rhenium, which is one of the best materials for the pressure standard, at room temperature and ultrahigh pressure up to 286 GPa [5] of which pressure was based on the equation of state of rhenium by Anzellini et al. [10]. The sound velocity of rhenium measured here by IXS combined with the density data obtained by X-ray diffraction can provide important constraints for the pressure scale in the multi-megabar pressure range.

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New Insight into Biophysics of Lipid Membranes with High Resolution IXS

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Biological membranes exhibit a great deal of compositional and phase heterogeneity due to hundreds of hemically distinct components that form them. Their phase behavior is enormously complex and, as a result, structural and dynamic processes in cell membranes are extremely difficult to study, especially at the molecular level. In the present work, we argue that high resolution inelastic X-ray scattering (IXS) can provide valuable insight into biophysics of lipid membranes. In the series of IXS experiments on single, binary, and ternary lipid mixtures, we show that such processes as passive transport, lateral lipid diffusion, and formation of phase separated domains can be quantitatively studied. Using high resolution IXS in combination with Molecular Dynamics simulation, we obtain experimental evidence of propagating in plane transverse acoustic (TA) phonon modes in a single component DPPC membrane and optical (OP) phononic modes in binary (DPPC-Cholesterol) and ternary (DPPC-DOPC/POPC-Chol) systems. These phonon modes exhibit low-Q/low-E phononic gaps (the absence of phonons within a particular Q- or E-range) as a function of temperature and compositional variation in those systems. In single-component systems [1], the phononic gap in the TA mode is a direct signature of the existence of transient voids caused by short-lived lipid density fluctuations that mediate solute permeation across the membrane. In more complex systems, the observation of OP modes that exhibit phononic gap is a direct result of the nm sized lipid patches formed by strongly coupled lipid "pairs"[2]. The concentration- and temperature-dependent nucleation of these stable entities leads to different phases on a phase diagram. We highlight that the results provide insights into the biophysical properties of rafts, including their size, life time and structural complexity.

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RIXS characterization of the giant crystal field, multiplet mixing and magnetism in $CeRh_3B_2$

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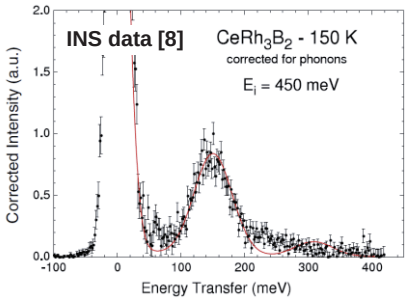
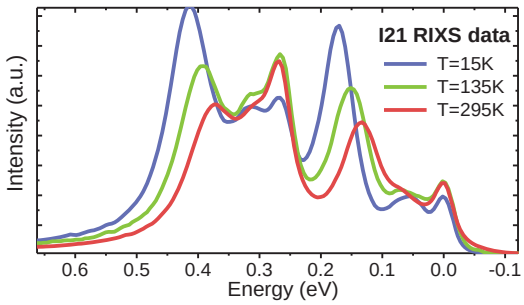
The physics of intermetallic cerium ($4f^1$) compounds is driven by the competition two phenomena, both functions of the exchange interaction J_{ex}. For small J_{ex} a long range magnetic order (RKKY) forms whereas for large J_{ex} the material is intermediate valent, with a partial delocalization of the 4*f* electrons in the conduction band (*cf*-hybridization). In between, a Kondo screened or so-called heavy fermion state is formed, alongside a reduction of magnetic ordering temperature due to the *cf*-hybridization. There are some puzzling exceptions with surprisingly high ordering temperatures, despite the strong presence of *cf*-hybridization.

The so-called giant crystal-field compound $CeRh_3B_2$ is one of these exceptions. Hexagonal $CeRh_3B_2$ orders ferromagnetically at T_{Curie}=116 K [1,2] although the DeGennes scaling from $GdRh_3B_2$ suggests a T_{Curie} two orders of magnitude lower and although the Ce configuration is highly intermediate valent (4*f* occupation<0.9), as found by PES and HAXPES [3,4]. The small saturated magnetic moment of $0.4\mu_B$ and the magnetic anisotropy cannot be explained within a simple crystal-field (CF) model [5,6]. The first CF transition detected by inelastic neutron scattering (INS) at 150meV (Fig.1) shows that the CF splitting is very large. Consequently, exchange interaction [6,7], crystal field [7,8] and spin-orbit (280meV) interactions act on an equal footing. Hence, for a deeper theoretical understanding, the CF scheme must be fully determined.

We present high resolution soft-RIXS data of $CeRh_3B_2$ at the Ce M_5 edge taken at I21 (DLS) and ID32 (ESRF). We obtained the first direct mapping of the full CF level scheme of $CeRh_3B_2$ above and below T_{Curie}. RIXS accesses excitations unreachable by INS thanks to a larger energy range and different selection rules (Fig.1,2). Comparison with full-multiplet calculations allows for a full description of the CF problem.





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Nematic Fluctuations and Lattice Coupling in $Ba(Fe_{0.97}Co_{0.03})_2As_2$ and FeSe

C-2

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We have conducted a detailed comparison of the transverse acoustic (TA) phonon dispersion in $Ba(Fe_{0.97}Co_{0.03})_2As_2$ (underdoped, T_S=95K) and FeSe (T_S=90K, T_c=9K). This TA phonon connects to the shear modulus near the zone center. Previously published data have shown a significant decrease in the shear modulus upon approaching the structural or superconducting transition. Nematic fluctuations couple to the lattice and soften the phonon energy in a q-dependent manner. Our work connects the zone-center shear modulus data with the finite-q phonon softening, with the goal of determining the role of nematic fluctuations in the phase transitions.

We made measurements of the phonon dispersion using the BL43LXU beamline (RIKEN, SPring-8) and HERIX Sector 30 beamline (APS, Argonne National Laboratory). F.W. and D.R. have published a paper showing how to fit the phonon dispersion and extract the nematic correlation length, ξ (Ref. [1]). We have applied this technique to our data with mixed results. The Lorentzian-form correlation function used in Ref. [1] fits our data poorly. Instead we used a Gaussian-form correlation function which significantly improved the quality of our fits although it does not match existing theory. Fig. 1 shows an example fit to the phonon dispersion.

Using the fit phonon dispersions, we extracted the nematic correlation length ξ (Fig. 2). The maximum ξ is on the order of 4 unit cells in both materials. ξ does not diverge upon approaching the structural phase transition. Implication of these results for our understanding of Fe-based superconductors will be discussed.

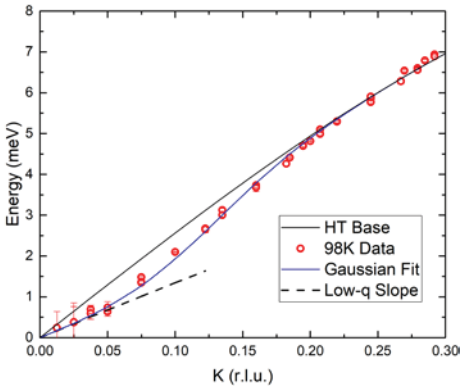

Fig. 1. Phonon energies and fits in UD Ba-122 (T_S=95K) at 98K. The black curve is the expected dispersion in the absence of nematic fluctuations. The blue curve shows fits of the phonon energy using a Gaussian nematic correlation function. The dashed line shows the expected low-q slope from published 3-point bending data.

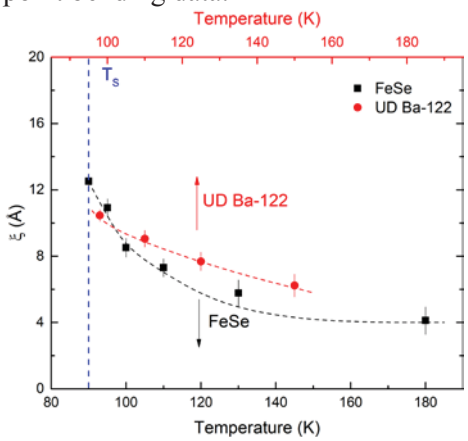

Fig. 2. Nematic correlation lengths ξ in FeSe and 3% Co-doped (UD) Ba-122. T_S shown by vertical dashed line; other dashed lines are guides to the eye. X-axes for the two compound have been translated relative to each other to match structural transition temperatures.

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Topological Phononic Excitations Revealed by Inelastic X-Ray Scattering

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Condensed matter systems have now become a fertile ground to discover emerging topological quasiparticles with symmetry protected modes. While many studies have focused on fermionic excitations, the same conceptual framework can also be applied to bosons yielding new types of topological states. Here we use inelastic x-ray scattering (IXS) to perform the first experimental measurement of the THz topological phonon dispersion in parity-breaking crystalline FeSi. By directly tracking the bulk phonon dispersion near the high symmetry points, we demonstrate the existence of the theoretically predicted double Weyl points, which have not yet been observed experimentally in related Fermion systems. Our results thus establish transition metal monosilicides as new model systems to explore topological bosonic excitations and symmetry protected properties such as the topological nontrivial edge modes[1,2].

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First-principles modeling of RIXS in liquid water: Core-excited nuclear dynamics and the distribution of local potential energy surfaces

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Most of chemistry and biology takes place in aqueous environments. Therefore large efforts are continuously devoted to the study of hydrogen bonded systems and specially ambient water. However, understanding the quantum nuclear motion and vibrations in liquid phase remains a non-trivial task in condensed phase physics. Hence, the development of local probes that deliver information about the intra- and inter-molecular interactions and dynamics around centers of interest is vital.

In this contribution, we carry out a comprehensive analysis of the resonant inelastic X-ray scattering (RIXS) in liquid water where the final state is the electronic ground state. When the photon energy is tuned to the pre-edge feature of the X-ray absorption spectrum, a long vibrational progression is observed, which arises due to the dissociative character of the core-excited state. Previously, the long progression had been analyzed empirically[1,2] but no strict calculations had been performed until now[3].

We present a quantitative analysis of the vibrational RIXS spectrum by a combination of ab-initio molecular dynamics simulations, local potential energy surfaces calculations and quantum nuclear wave packet simulations of the cross-section. Our model stands on a robust framework which was carefully established for the gas-phase[4-6]. We effectively demonstrate that RIXS constitutes a complementary probe to traditional infra-red spectroscopy, while being more sensitive to the local hydrogen bond (HB) environment since it gives direct access to highly excited vibrational states, in which the OH bonds stretch far from the equilibrium covering a large range of ground state potential energy surface.

For completeness, we also analyze the lowest electronic inelastic channel, which displays the well known $1b_1$ splitting[7]. We do so by looking at its excitation energy dependence across the pre-edge. We see that this splitting arises due the near-parallel behavior of the potential energy curves involved in the process, thus leading to the formation of a non-energy-dispersive pseudo-atomic peak and a molecular band.

In summary, here we show how vibrational RIXS spectroscopy can shed light on the local environment variations in liquid water and are able to analyze HB strength via the distribution of local potential energy curves along the OH bonds extracted from experimental data. In contrast, we see that the $1b_1$ peak at the pre-edge arises from the nuclear dynamics and due to the non-bonding nature of the $1b_1$ and 1s orbitals involved in the transition, being less sensitive to the local structure than the quasi-elastic channel.

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Systematic Calculations of RIXS for the 3d Transition Metal Oxides by the *Ab-initio* Multiplet Methods

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The spectral shapes of resonant inelastic x-ray scattering (RIXS) at the *K*-pre-edge and $L_{2,3}$-edges of 3d transition metals are dominated by the multiplet structures which originate from the strong electronic correlations between 3d electrons and 2p core-hole. Therefore, the single-particle approximation including the density functional theory (DFT) cannot reproduce the experimental spectra in general. To treat the electronic transition from the core 2p level, relativistic effects such as the spin-orbit coupling should be considered. In addition, the ligand-to-metal charge transfer (CT) plays an important role in determining the spectral shapes of the *K*-pre-edge and $L_{2,3}$-edge RIXS. Thus, the contribution of ligand oxygen ions should also be included in the calculations. The semi-empirical charge transfer multiplet (CTM) method has commonly been used for simulating the RIXS spectra on transition metal compounds [1]. However, the method cannot be used to predict the spectral shapes *a priori*, because of the use of adjustable parameters such as the ligand-field splitting (10*Dq*) and charge transfer energy (Δ). An *ab-initio* approach that takes both the multiplet and CT effects into account is strongly desirable.

In our research group, the *ab-initio* multiplet method for x-ray absorption (XAS) has been developed [2], and it has also been extended to simulating RIXS spectra, recently [3]. The method is based on the relativistic configuration interaction (CI) theory in the quantum chemistry using cluster models. The electronic correlations between 2p and 3d electrons are rigorously taken into account by expressing the wavefunctions corresponding to initial, intermediate, and final states as linear combinations of all possible Slater determinants. The ligand-to-metal CT can also be included in the same theoretical framework by introducing the additional electronic configurations in the CI.

In this study, the *K*-pre-edge and $L_{2,3}$-edge RIXS of transition metal oxides with various oxidation states and crystalline structures were calculated systematically by the *ab-initio* multiplet method. The effects of covalent bonding between transition metal and ligand oxygen ions, and the ligand-to-metal CT are investigated in detail.

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Computing Resonant Inelastic X-Ray Scattering Spectra Using The Density Matrix Renormalization Group Method

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In recent years, the continued improvement in instrumentation is making resonant inelastic x-ray scattering (RIXS) a powerful spectroscopic tool to study elementary excitations of strongly correlated materials [1]. On the theory side, the computation of RIXS spectra starting from model Hamiltonians is often a formidable task because of the absence of accurate many-body tools, particularly when many orbitals are active. In most cases, exact diagonalization (ED) techniques are used which restricts clusters to a relatively small size that limits momentum resolution. In addition, the interpretation of the RIXS cross section in terms of easier dynamical correlations functions is often a subtle theoretical problem [2]. For these reasons, alternative computational methods that can overcome these limitations are highly demanded.

In this talk, I will present a novel method for computing RIXS spectra in quasi-one-dimensional quantum systems using the Density Matrix Renormalization Group Method (DMRG). The DMRG is a powerful numerical method based on a variational ansatz for the many-body wave function in terms of matrix product states [3]. It is able to treat strongly correlated electronic systems with hundreds of lattice sites, obtaining the ground state as well as the dynamical response functions with very high accuracy. I will show how to apply the DMRG method to compute the RIXS scattering cross-section within the Kramers-Heisenberg formalism. With this new procedure, I computed the low-energy magnetic excitations observed in Cu L-edge RIXS for the challenging corner shared CuO_4 chains on cluster sizes well beyond state-of-the-art ED techniques, using both multi-orbital and downfolded t-J model Hamiltonians [4]. Finally, I will discuss the implications of the results for experiments and outline future directions of research.

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Magnetic Dynamics of $Sr_2IrO_4/Sr_3Ir_2O_7$ Superlattices

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Spin-orbit entangled magnetic moments, or pseudospins, are predicted to express a wide array of novel magnetism ranging from isotropic (Heisenberg) to extremely anisotropic (Kitaev) interactions [1]. The Ruddlesden-Popper (R-P) iridate series $Sr_{n+1}Ir_nO_{3n+1}$ is an ideal platform to investigate isotropic pseudospin physics that underlie a plethora of unconventional electronic phases [2]. Sr_2IrO_4 (n=1), on one side, is well described by a spin-1/2 Heisenberg model in a square lattice in promising parallel to cuprate phenomenology [3,4,5]. $Sr_3Ir_2O_7$ (n=2) on the other side, is an Ising-like system that has been identified to exhibit a charge-density-wave-like instability [6, 7].

We have grown pure single-phase iridates with an ideal alternate stacking of the Heisenberg-like single layer Sr_2IrO_4 and Ising-like bilayer $Sr_3Ir_2O_7$ units. This superlattice combines multiple distinct two-dimensional phases in one material system, which we exploit to identify two-dimensional electronic instabilities that require the integrity of a three-dimensional lattice structure. Resonant X-ray Diffraction shows that the finite-temperature magnetic orderings of the single layer and bilayer units are largely unaffected, indicating that the static order is governed primarily by anisotropic interactions. To study the pseudospin dynamics in this complex system, we utilized state-of-the-art High-energy-resolution Resonant Inelastic X-ray Scattering [8], which reveals that within the two-dimensional framework the pseudospin dynamics around the magnetic zone center display a behavior distinct from the parent systems, including a vastly enhanced spin wave velocity.

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Imaging sub-micrometer length scales with RIXS on complex oxides

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Complex oxides can display inhomogeneities on sub-micrometer length scales [1], partially through chemical disorder, partially through the spontaneous formation of domains. Some of these fluctuate, while others can be fixed in space through pinning. In any case, soft X-ray resonant inelastic X-ray scattering (RIXS) has been shown to directly access the excitations relevant for materials functionality, but has so far been applied without spatial resolution, limited by the beam line spot size and available beam time.

With an imaging RIXS spectrometer though, we can combine the ability to obtain spatial resolution and acquire a large set of spectra in parallel at the same time without increasing damaging X-ray flux densities on the sample. This increases the throughput of such a device by around two orders of magnitude. Such a spectrometer provides direct access to materials' inhomogeneities but also allows RIXS to be applied to confinement effects at the edges of micro-structured materials or e.g. real semiconductor devices in operando.

We demonstrate here the application of an off-axis transmission zone plate as an analysing element [2] and show how we achieve resolution on the order of 2µm in both spatial dimensions in the surface plane of a sample, limited only by the spectrometer length and detector pixel size [3]. One of the axes is scanned, while the other is imaged in parallel to recording RIXS spectra for a range of excitation energies. The full dataset then allows to extract complete RIXS maps (naturally including absorption spectra) for each point on the sample surface.

The demonstrated spectrometer concept using a single optical element though has strong chromatic aberrations which could be alleviated by novel detector concepts that work at extremely grazing incidence angles. Easier to implement is a combination of two elements, where the chromatic aberrations can cancel each other. Proof-of-concept experimental results will be shown alongside with ray-tracing simulations.

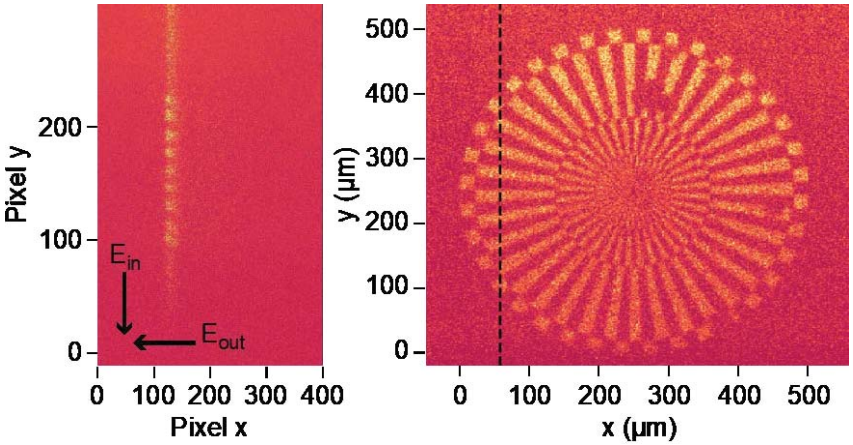

Figure:
Left: Detector image, x encodes the emitted energies, different incoming energies illuminate also different points on the sample.
Right: Scanning in x allows to reconstruct an oxygen fluorescence map of a test pattern. Structure at the dashed line is extracted from the detector image on the left.

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Stimulated X-ray Emission Spectroscopy with hard X-rays

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Current XFEL spectroscopy experiments are using primarily the linear interaction of matter with the X-ray pulses taking advantage of their very short duration. However, given the ultrahigh intensities that can be obtained when focusing these pulses, nonlinear X-ray matter interaction can occur. An exciting future perspective is the transfer of nonlinear spectroscopy from the optical to the hard X-ray spectral region, which is known for its sensitivity to the electronic structure of *e.g.* 3d transition metal ion complexes.

For this, a highly focused X-ray beam tuned to above the Mn 1s ionization energy creates a population inversion along its beam path, allowing stimulated emission that leads to massive intensities mainly in the forward direction from a single X-ray shot.

For amplified spontaneous emission processes at Mn $K\alpha_1$, we find spectra at amplification levels extending over four orders of magnitude until saturation, and observe bandwidths below the Mn 1s core-hole lifetime broadening in the onset of the stimulated emission. In the exponential amplification regime the measured spectral width is constant over three orders of magnitude, pointing to the build-up of transform-limited pulses of $\sim$1 fs duration. Driving the amplification into saturation leads to broadening of the line. Importantly, the chemical sensitivity of the stimulated X-ray emission to the Mn oxidation state appears to be preserved.

Using a second color as a seed, the amplified spontaneous emission process can be overcome and weaker emission lines are amplified. Kb spectra are known for their high chemical sensitivity and dependence on the effective spin state of the transition metal ion. Stimulating the Kβ emission lines (which are ~ 8-10 times weaker than $K\alpha_1$) is therefore an important and exciting step towards a broader applicability of stimulated emission spectroscopy.

In this presentation, spontaneous and seed amplified stimulated emission experiments will be presented and discussed together with a comparison to conventional emission spectroscopy taken at a synchrotron.

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Resonant Inelastic X-ray Scattering on ultrathin FeSe grown on $SrTiO_3$

C-10

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The recent discovery of enhanced superconductivity in FeSe monolayer grown on $SrTiO_3$ spurred a lot of interest in the condensed matter community. When grown in the ultrathin limit FeSe displays a T_c of about ~60 K, an order of magnitude higher than its bulk counterpart (~8 K). However, the limited amount of material precludes many spectroscopic investigations leading to the lack of experimental data able to proof or disregard theories explaining the enhanced superconductivity.

Resonant Inelastic X-ray Scattering (RIXS) is a photon-in photon -out technique able to probe bosonic modes in thin materials. We successfully performed Fe-L edge RIXS on FeSe bulk and monolayer. The Fe resonance assures that the measured signal is originating from the FeSe layer and not from the $SrTiO_3$ substrate. I will present high resolution RIXS data revealing a remarkable evolution of the low energy spectrum between the bulk and monolayer FeSe. A dispersion mode resembling the spin excitations observed in other Fe-based superconductors is observed in the bulk [1-4], but in the thin limit a flat optical mode of enhanced bandwidth is detected.

I will discuss the interpretation of the low energy spectrum and some possible mechanisms to explain the evolution of the excitations from the bulk to the thin limit. Finally, I will mention the implications of our evidence and how they can help to shed light on the high T_c of FeSe monolayer.

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Direct imaging of orbitals using inelastic X-ray scattering

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It is generally accepted that the correlated motion of electrons is at the origin of many interesting physical properties including unconventional superconductivity, colossal magnetoresistance, and multiferroicity, just to name a few. These spectacular properties emerge through intricate interplay of charge-spin-orbital degrees of freedom of valence electrons, so their characterization is an essential ingredient for modeling these curious behaviors and revealing the underlying mechanisms. This is a very challenging and delicate undertaking, which often requires the knowledge of the active valence wave function.

We developed a new experimental method that directly images the active orbital in solids, without advanced calculation or spectroscopic analysis [1]. The method, s-core-level non-resonant inelastic X-ray scattering (s-NIXS), relies on high momentum transfer in the inelastic scattering process, which is necessary for dipole-forbidden terms to gain spectral weight. To demonstrate the strength of the technique, we imaged the text-book example, x^2-y^2/3x^2-r^2 hole orbital of the Ni^{2+} ion in NiO single crystal (see Figure). We will present the basic principles of s-NIXS and its experimental implementation.

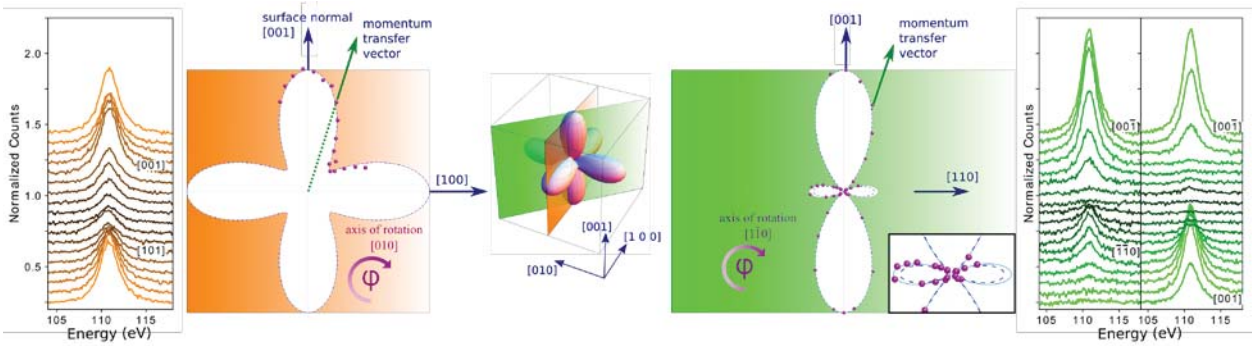

Figure: High momentum transfer allows the dipole-forbidden 3s→3d transition to gain spectral weight. Directional dependence of the spectral intensity associated with the 3s→3d transition directly maps the local hole charge distribution.

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Double dispersion RIXS (QERLIN) at the ALS

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A novel momentum-resolved resonant inelastic x-ray scattering spectroscopy (RIXS) instrument using the double-dispersion concept is under construction at the ALS. The beamline (QERLIN) utilizes the Hettrick-Underwood optical scheme to produce an erected focal plane where the sample will be placed. The scattered photons over the illuminated area on the sample, which is 2 microns (width) by 2 mm (height), will be re-imaged vertically by a Wolter type 1 mirror pair in the spectrometer. In the meantime, the spectrometer dispersive assembly comprised of an elliptical cylinder and varied line spacing plane gratings disperses these photons horizontally. In the detector plane, one can then record the RIXS spectra over a wide-range excitation photon energy in single acquisition (RIXS map). This setup, taking advantage of the off-axis x-rays typically blocked by beamline exit slit, will give enhanced throughput over the conventional RIXS setup. The science drivers, challenges, and current status will be reported.

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Hyperspectral Imaging with X-ray Raman Scattering

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X-Ray Raman Scattering (XRS) has been widely used to investigate the K-edge of light elements. XRS combines the chemical sensitivity of XANES with the benefits of hard x-rays for bulk sensitive spectroscopy. We will present here a novel extension of XRS towards hyperspectral imaging (XRS-I) with a focus on ancient materials [1,2] as performed at the GALAXIES beamline at SOLEIL Synchrotron [3] in collaboration with IPANEMA, SLAC and ESRF. XRS-I provides 'chemical images' with micron resolution. The hyperspectral images produced by XRS-I can be analyzed via segmentation algorithms to extract spatial distribution of different chemical classes. Perspectives in the view of SOLEIL upgrade program will be discussed.

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CF scheme of UO_2 measured with NIXS

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Core-level non-resonant inelastic x-ray scattering (NIXS), also known as x-ray Raman scattering, is a powerful tool for investigating the orbital occupation in uranium systems. Although the crystal field is known from neutrons in UO_2 [1], the crystal field schemes of metallic actinide systems have been generally too broad to determine with neutrons.

In core-level NIXS high momentum transfer of the order of $|\vec{q}| \approx 10\text{Å}^{-1}$ gives rise to terms in the double-differential cross section that are higher than dipole transitions. The spectral features of these multipole transitions are more excitonic and show well defined multiplet structures also for actinides [2] so that multipole selection rules give access to the ground-state symmetry when studying the vector $\vec{q}$ dependence of the scattering function $S(\vec{q},\omega)$ [3,4]. These multipole selection rules give, in contrast to the dipole case, also insight into symmetries higher than twofold so that also cubic materials can be investigated. Another important advantage for actinides is the bulk sensitivity of NIXS; eliminating *in-situ* cleaving.

Here we present NIXS data of cubic UO_2 at the U $O_{4,5}$ edges ($5d \rightarrow 5f$) that were taken at the RIXS end station on ID20 at the ESRF (see Fig. 1(a)). UO_2 is a well-studied uranium compound because of its importance as nuclear fuel. UO_2 is one of the very few uranium compounds where inelastic neutron scattering [1] could propose a crystal-field scheme. It is therefore ideally suited as a benchmark material for demonstrating the power of NIXS [5].

The Hund's rule ground state of the $5f^2$ configuration of U in UO_2 splits into 1 singlet, 1 doublet, and 2 triplets by the cubic crystal field. Figure 1(b) shows the experimental directional dependence of $S(\vec{q},\omega)$ together with a simulation for the Γ_5 triplet ground state. The calculation was performed with the full multiplet code *Quanty* taking into account a realistic CF splitting that is based on neutron scattering [1]. We show that the size of the CF splitting has a large impact on the NIXS spectra so that we are able to distinguish between a weak and strong CF scenario as was discussed for UO_2. This experiment demonstrates that the directional dependence in NIXS can be modeled quantitatively and that the branching ratio/lineshape is strongly influenced by the size to the CF splitting. In our previous work, this was not taken into account [4].

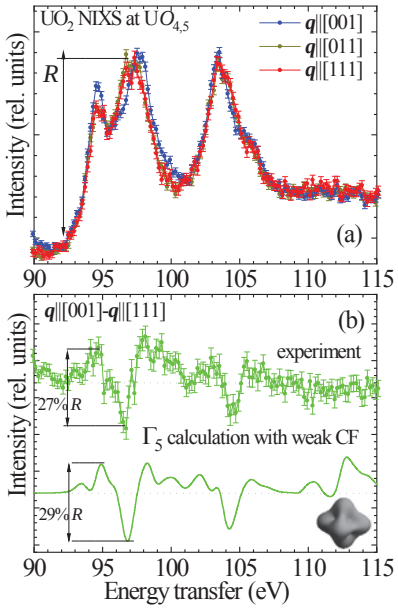

Figure 1: (a) Experimental NIXS data at $|\vec{q}| \approx 9.1\text{Å}^{-1}$, (b) $\vec{q}$-directional anisotropy of $S(\vec{q},\omega)$ of the UO_2 U $O_{4,5}$ edges adapted from [5].

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Thermal acoustic excitations with atomic scale wavelengths in amorphous silicon

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The vibrational properties of glasses remain a topic of intense interest due to several unresolved puzzles, including the origin of the Boson peak and the mechanisms of thermal transport. Inelastic scattering measurements have revealed that amorphous solids support collective acoustic excitations with low THz frequencies despite the atomic disorder, but these frequencies are well below most of the thermal vibrational spectrum. Here, we report the observation of acoustic excitations with frequencies up to 10 THz in amorphous silicon using inelastic X-ray scattering. The excitations have atomic-scale wavelengths as short as 6 Å and exist well into the thermal vibrational frequencies. Simulations indicate that these high frequency waves are supported due to the high group velocity and monatomic composition of a-Si, suggesting that other glasses with these characteristics may also exhibit such excitations. Our findings demonstrate that a substantial portion of thermal vibrational modes in amorphous materials can still be described as a phonon gas despite the lack of atomic order.

Tracking Collective Spin and Charge Excitations through Time-Resolved Raman and RIXS Spectra

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Ultrafast characterization and control of elementary excitations are critical to understanding and manipulating emergent phenomena in strongly correlated systems. In particular, spin plays an important role in unconventional superconductivity, but efficient tools for probing spin dynamics out of equilibrium are still lacking. In this talk, I first introduce the theory for non-resonant time-resolved Raman scattering as a way to detect spin dynamics. Different ultrafast processes are shown to exist in the time-resolved Raman spectra simulated in a Hubbard model and dominate under different pump conditions. Particularly, for high-frequency and off-resonance pumps, we show that the Floquet theory works well in capturing the softening of bimagnon excitation. We next discuss the theory of its resonant correspondence, the time-resolved RIXS. With a focus then on the charge excitations, we simulate the time-resolved XAS and indirect RIXS in a Mott insulator. Using various resonances, we decompose the charge excitations with various intermediate states. While the Stokes response reflects the photoinduced low-energy modes, the anti-Stokes response in the "well-screened" and "double-screened" resonances reveals the propagating and local charge excitations, respectively. Through the resonance with intermediate states, we show that trRIXS can decipher charge excitations from different origins in a nonequilibrium state.

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Commissioning of New CdTe Pilatus Area Detector for High-Energy Resolution Inelastic Scattering Spectrometer at Sector 30 Advanced Photon Source

PT-1

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We have installed and commissioned a new Pilatus area detector for the high energy resolution inelastic x-ray (HERIX) spectrometer at sector 30 at the Advanced Photon Source (APS). The detector is a replacement for the single CdTe single element detector system. Pixelated detectors in principle, offer different advantages over single element detectors including the ability of analyzing the energy as a function of pixel position, also it allows for viewing the image formed by the analyzer which in turns helps in optimizing the focus of the analyzers. In this work, we present the technical details of the detector as well as test results. The CdTe pixelated detector has an area of 83.8x33.5 mm^2 with pixel size of 0.172x0.172 mm^2 and a thickness of 1 mm. We also present the challenges caused by the cosmic rays and solutions to mitigate this problem.

Operando RIXS studies of electrode materials for Li-ion batteries to understand the redox reactions

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Improving the energy and power density of electrode materials for Li-ion batteries (LIBs) is highly important to further develop electric and hybrid-electric vehicles. For the improvements of battery performance, understanding the charge (Li-extraction) and discharge (Li-insertion) mechanisms from a viewpoint of the electronic structure is indispensable. Recently, electronic-structure analyses of the electrode materials using soft X-ray spectroscopy have been of particular importance. While many soft X-ray absorption spectroscopy (XAS) studies of the electrode materials have been reported, we have focused on the development of *operando* resonant inelastic soft X-ray scattering (RIXS) techniques and have revealed the relationships between the performances for several electrode materials and their electronic-structure changes during charge-discharge [1,2].

We demonstrated *operando* Mn L_3-edge RIXS for $LiMn_2O_4$. The *operando* cell consists of the $LiMn_2O_4$ thin film, a counter electrode and an electrolyte solution [3]. The *operando* RIXS experiments were carried out at BL07LSU of SPring-8 [4]. The RIXS spectra were analyzed by theoretical analyses based on the configuration-interaction full-multiplet calculation [2,5]. A strong charge-transfer (CT) effect from the O 2*p* to Mn 3*d* orbitals has been found for the Mn^{4+} state. In parallel, the electronic structures and CT effects for several cathode materials like $LiFePO_4$ have been studied by means of XAS and RIXS [6-8]. We explained that the changes of CT effects during charge-discharge could be related to the corresponding cycle performances.

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Evidence for MARX-Raman(Multi Atom Resonance X-ray Raman)

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X-ray absorption fine structure (XAFS) is a powerful tool to investigate the local structure around the catalyst active site especially under the working conditions. However, it is very difficult to apply XAFS to light elements such as O, C, and N because (1) X-ray is easily absorbed by the coexisting reactant gases. (2) The XAFS signal from the light elements at the active sites is hindered by the other coexisting light elements. Here we propose a new method to identify the light element under the real catalytic conditions, called as MARX Raman [1] which is a bond-specific method. The theory of MARX Raman is similar to that of MARPE(Multi-Atom Resonant Photoemission) .[2, 3] Although there are experimental and theoretical evidence for MARPE[4, 5], yet many people considered it is due to the experimental artifact[6] which may arise from the large background. In case of Raman, the background signal is small so that we can detect the signal more easily.

Experiment was carried out at BL36XU of SPring-8.[7] Figure 1 shows the MARX-Raman signal from $Er(cp)_2$.[7] The excitation energy was 9752 eV just at the L1 Er edge. We found the abrupt change of emission intensity at 287 eV which was corresponding to C K-edge. It could be assigned to the MARX-Raman signal. Theoretical investigation indicated the MARX-Raman signal was proportional to R^{-2} (R was bond distance) and it appeared in the lack of spatial inversion center in the light elements. We would like to discuss the MARX-Raman possibilities.

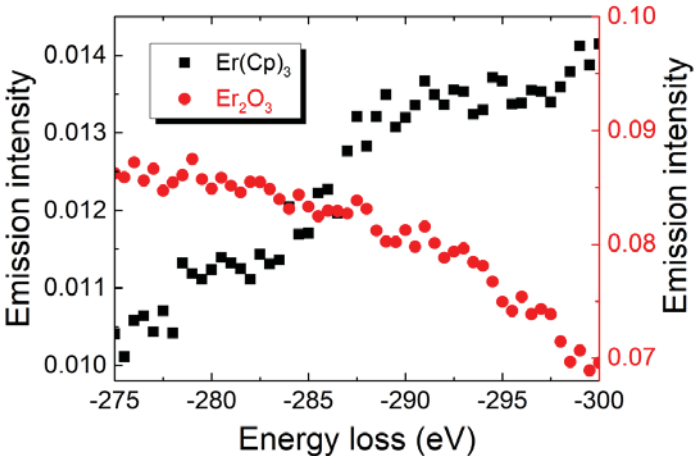

Figure 1 Raman spectra at 9752 eV of $Er(C_5H_5)_3$ (a line formed by black squares) and Er_2O_3 (by red circles).[7]

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Bimagnon tail in the magnetic excitations of $Sr_2CuO_2Cl_2$ and La_2CuO_4

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The spin ½ antiferromagnetic (AFM) two-dimensional (2D) square lattice is one of the most well studied quantum systems and represents a benchmark for quantum magnetism. In particular, a square AFM lattice in the CuO_2 planes represents the building block of all layered high-temperature cuprate superconductors. The parent compounds La_2CuO_4 (LCO)[1] and $Sr_2CuO_2Cl_2$ (SCOC)[2] are well known examples of 2D AFM systems and their spin wave excitations have been investigated by inelastic neutron scattering (INS). However, thanks to great improvements in the experimental energy resolution, resonant inelastic x-ray scattering (RIXS) has proven to be a particularly useful technique to study a great variety of excitations in strongly correlated systems. In particular, it has also been possible recently to analyze the linear polarization of the scattered light (σ and π, polarimetric analysis). Recently, Plumb *et al.*[2] pointed out some discrepancies between INS and RIXS measurements in the magnetic excitation dispersion of SCOC close to q = (½, 0) r.l.u. They attributed them to the presence higher-energy electronic excitations invisible to INS.

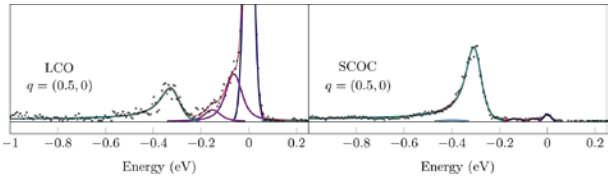

Figure 1: Low-energy part of the spectra at (½, 0) for LCO and SCOC. The various spectral features are fitted, showing the Fano lineshape of the magnon peak (teal curve) around -0.3 eV. Black dots: experimental data, red = fit, navy curve = elastic line, purple curve = phonons, teal curve = single magnon, blue curve = bimagnon.

Here, we report state-of-the-art ultra-high resolution RIXS measurements on LCO and SCOC. Interestingly we find that the single magnon excitations always show an asymmetric lineshape (Fig. 1); this asymmetry increases moving from Γ to (½, 0). We interpret this as a signature of a Fano-like process induced by the interaction between the single magnon and the very broad bimagnon continuum of excitations. We show that a Fano lineshape produce a much better fitting of the single magnon peak than more conventional damped oscillator functions. Considering this, the LCO spin wave dispersion matches the one derived from INS, while the discrepancy in the SCOC dispersion is reduced to ~15 meV and can now be easily attributed to errorbars and artifacts. This effect is more pronounced in SCOC than in LCO because in the latter the bimagnon excitations are at higher energy. In addition, we performed the polarimetric analysis for SCOC at three different q points (Fig. 2). This allows us to assign the single magnon peak (ΔS = 1) to the crossed channels (πσ' and σπ'). Contrary to what previously assumed, the bimagnon peak (ΔS = 0) does not completely belong to the non-crossed channels (σσ' and ππ') but instead shows a mixed character at all q values. In addition, the energy of the bimagnon peak varies for different incoming polarization at (½, 0). These surprising results can be well reproduced with interacting spin wave theory, which has already successfully explained the multi-magnon Raman and IR absorption spectra of cuprates[3].

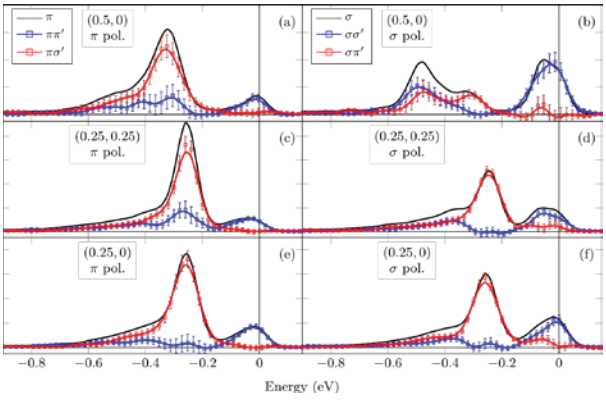

Figure 2: Polarimetric analysis on SCOC at various points in reciprocal space. The bimagnon shows mixed character.

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Options for Inelastic X-Ray Scattering at PETRA IV

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PETRA III is operating as a synchrotron radiation source for about 10 years now. It is diffraction limited in the vertical plane, and delivers high coherent flux in the hard x-ray range, however, demand for smaller foci and even higher coherent flux increased in recent years. Therefore, DESY decided to investigate options for an upgrade of the accelerator in order to create a diffraction limited storage ring for synchrotron radiation. This new project PETRA IV [1] started in 2016. The current plan is to rebuild the storage ring in 2025, and start operation of PETRA IV in 2027.

I will present the present status of the design studies of the PETRA IV project, and focus on properties of the radiation which will be delivered by undulators. Based on experience, which we gained in recent years with the operation of the new x-ray spectroscopy beamline P64 at PETRA III [2], I will present parameters like flux and beam-size, which we can expect for a new beamline at PETRA IV, and discuss possible applications in the field of inelastic x-ray scattering, which will profit from the low emittance of PETRA IV.

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Momentum-resolved lattice dynamics of parent and electron-doped Sr_2IrO_4

PT-6

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The delicate balance of spin-orbit coupling (SOC), crystal fields and electron correlations (U) in the *5d* iridates makes them a fruitful class of materials in the search for novel electronic and magnetic phases[1]. Most prominently, the layered perovskite Sr_2IrO_4 has been shown to be a spin-orbit Mott insulator where the orbital degeneracy of the Ir^{4+} t_{2g} levels is lifted by SOC, enabling a moderate U~2eV to open a charge gap[2]. Moreover, it has striking structural, electronic and magnetic similarities to the parent of the cuprate high-temperature superconductors La_2CuO_4[3], which on doping hosts many broken symmetry phases such as charge density waves that appear to be intimately linked to high-temperature superconductivity[4].

One crucial difference from the cuprates is the orbital character of the $j_{eff} = 1/2$ wavefunction, which results in the couplings between the pseudospins being highly sensitive to lattice geometry[1]. Recent theoretical[5] and experimental[6] work has shown that this coupling is essential to understanding the ground-state magnetic structure and in-plane magnon gap of Sr_2IrO_4. Lattice distortions can result in significant admixture of the $j_{eff} = 3/2$ wavefunction into the $j_{eff} = 1/2$ ground state, which has led to predictions of strong interactions between lattice, orbital, and magnetic excitations in the layered iridates[7].

These phenomena can, in principle, be sensitively probed through studies of the lattice dynamics, which we have conducted for both parent and electron-doped Sr_2IrO_4 through a combination of high-resolution, momentum-resolved inelastic x-ray scattering measurements (both non-resonant and tuned to the oxygen *K* edge) and density functional theory calculations. Despite the predictions, in the parent compound we find no clear evidence for the coupling of lattice excitations to those in the spin sector within the ~1.5meV experimental energy resolution, and in the doped compound the phonon anomalies expected if cuprate-like charge density wave order were present are not observed.

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Fabrication of Single Perovskite Solar Cells and Projection to Increase V_{OC} via SnO_2 Experimentation

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Since its discovery as both a hole-transport medium (HTM) and electron-transport medium (ETM), the organic-inorganic halide perovskite has been revolutionary in the research and development of solar cells with ever-increasing power conversion efficiency (PCE). Further research of the effects of introducing mixed-halide and lead (Pb) and tin (Sn) blends in the ABX_3 molecular composition of perovskite has led to published results displaying the increases in electrical parameters such as efficiency (η), open-circuit voltage (V_{OC}), short-circuit current density (J_{SC}), and fill factor (FF). Experimentation focused on perovskite molecular composition has also led to increased tolerances to exposure to moisture and high temperatures.

Our primary research is focused on developing an optimal methodology to fabricate competitive thin-film perovskite solar cells. Our design includes devices with areas of 0.096 cm^2, a glass superstrate coated with fluorine-doped tin oxide (FTO), a solution-processed perovskite with HTM and ETM heterojunctions consisting of spiro-OMeTAD and tin oxide (SnO_2), respectively, followed by a silver (Ag) rear contact. Our ABX_3 perovskite composition includes a blend of formamidinium (FA) and cesium (Cs) cations as the A molecule, B is a Pb metal cation, and X_3 is a mixed halide. Thus far, our fabrication methods have yielded complete solar cells that provide η ranging from 10.7 to 15.5%, as well as V_{OC} and J_{SC} values from 1.095 to 1.110 V and 21.7 to 23.2 mA/cm^2, respectively. Literature proves that by including antireflection coatings (ARC), wavelength (λ) selective gratings, and light trapping structures into our fabrication process, an overall increase in our device performances can be achieved.

A future note of particular interest is attempting to increase V_{OC} to at least 1.50 V via fabricating solar cells with varying thicknesses of the ETM layer. Controlled experimentation will include performing SnO_2 layering via spin-on and / or chemical bath deposition (CBD) processes for varying amounts of time, applied temperatures, and layer thickness. A related interest includes observing device performance with ETMs processed with slight changes in their recipe (e.g. a volumetric change in the Sn-base solution or other components of the material).

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Time-resolved high-resolution resonant inelastic soft X-ray scattering at the 3d transition metal M-edge: the NiO case

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The development of high brilliant, high-repetition rate free-electron lasers (FELs) and the advances of X-ray spectrometers makes time-resolved high-resolution RIXS studies with sub-picosecond time resolution possible. Ultrashort dynamics of quasiparticle interactions in functional materials as well as life science related questions can be studied now within reasonable experimental time, however, still putting challenging demands on the experiment.

Here, we report on a time-resolved study of the *dd*-excitations in NiO at the Ni M-edge, performed at the TRIXSS (**t**ime-**r**esolved high-resolution **i**nelastic soft **X**-ray **s**cattering **s**pectrometer) end-station at FLASH at DESY in Hamburg. This imaging spectrometer installed at the PG1 monochromator beamline currently covers a photon energy range of 36 - 210 eV with an energy resolution of 55 - 90 meV and a time resolution better than 300 fs fwhm [1, 2].

For the presented experiment, NiO has been optically pumped and high-resolution RIXS data at the Ni M-edge resonance with 65 meV energy resolution have been recorded for several pump-probe delays (100 fs to several ps). We discuss the experimental setup, timing diagnostics and show first (preliminary) RIXS data.

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X-ray Raman scattering study of hydrothermal fluids: The case of aqueous sodium chloride solutions

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Aqueous solutions play a key role in many natural and industrial processes. Particularly, hydrothermal fluids (i.e. hot aqueous fluids with dissolved electrolytes, silica, CO_2, or other solutes) are essential in mass and energy transfer in the Earth's crust and upper mantle, for instance in subduction zones [1]. These fluids are also involved in the formation of most ore deposits. The system H_2O + NaCl is of special interest in geosciences as NaCl is long known to be the major solute in aqueous fluids in Earth's crust [2]. Interestingly, the effect of electrolytes on the water structure at sub- and supercritical conditions has only been investigated for a handful of chemical compounds. The role of additional protons and hydroxide ions in aqueous solution is still debated [3,4,5] and the spatial extend of the ion's influence on the local water structure is controversially discussed [6]. In order to reveal the microscopic structure of this fascinating liquid and to investigate the influence of the alkali halide on the hydrogen bond network up to the supercritical regime, we present an in situ x-ray Raman scattering study of aqueous sodium chloride solutions at high pressure and high temperature. Probing the oxygen K-edge of the salt solution yields unique information about the oxygen's local coordination around the ions, e.g. solvation-shell structure and ion pairing. Using x-ray Raman scattering in combination with ab initio modelling of the liquid's molecular structure we aim to constrain the structural effects of sodium chloride on the hydrogen-bond network providing unique understanding of NaCl solvation in hot aqueous fluids. A first qualitative analysis of the spectra exhibit the increase of spectral weight in the pre-edge region accompanied by a decrease in intensitiy in the post-edge region. This finding points towards a loss of tetrahedral order accompanied by a reduction of the number of hydrogen bonds in the system.

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Combining x-ray emission and x-ray Raman spectroscopy for the study of Earth materials at high pressure and high temperature

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X-ray emission and x-ray Raman scattering spectroscopy are powerful tools to investigate the local electronic and atomic structure of high and low Z elements in situ and can be applied at high pressure and high temperature conditions which are present in e.g. the Earth's mantle. We developed a setup for simultaneous x-ray emission (Fe $K\beta_{1,3}$ and valence to core) and x-ray Raman scattering (low Z elements' absorption edges) studies at beamline P01 of PETRA III synchrotron radiation source using a wavelength dispersive von Hamos spectrometer together with the existing multiple-analyzer Johann-type spectrometer in combination with a laser heating device. It's capabilities are demonstrated by investigating the iron spin crossover in siderite ($FeCO_3$) and bridgmanite ($(Mg,Fe)SiO_3$). This setup provides a unique combination in order to achieve new insights into the spin transition and compression mechanisms of mantle materials which is of importance for the understanding of the macroscopic physical and chemical properties of the inner Earth.

Trimeron correlations above the Verwey temperature in Magnetite?

PT-11

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Introduction

The origin of the metal to insulator transition in magnetite (Fe_3O_4) remains a mystery due to the complexity of the system: it is a mixed valence, strongly correlated system where many interactions such as Jahn-Teller (dynamical and static), Kugel-Khomskii and phonons are very close in energetics. A recent interpretation of the Verwey transition as an ordering of a three-site magnetic polaron, the trimeron, has been put forward. It has remained a challenge to establish the existence of the high temperature trimerons because of their dynamical nature and associated small local Jahn-Teller distortion which renders standard techniques as X-ray diffraction, NMR and UV-Vis spectroscopy ineffective. In order to shed light on the high temperature trimeron correlations, we measured for the first time high resolution (~76 meV) Fe 2p3d RIXS magnetic circular and linear dichroism (RIXS-MCD and RIXS-MLD).

Results and discussion

We measured high resolution Fe 2p3d RIXS on a (001) Fe_3O_4 synthetic single crystal at the ADRESS beamline of Swiss Light Source. A permanent magnet (B = 0.4T) was used to saturate the magnetization of Fe_3O_4. We observed a strong RIXS-MLD signal at the low energy-loss spin-orbital excitations. The dichroic response enables us to unambiguously deduce the ground state symmetry of the nominal Fe^{2+} ions. Guided by theoretical simulations [2], we reveal that the polarization dependence of the low energy spin-orbital excitations is incompatible with tetragonal Jahn-Teller trimeron type distortion (compare Fig. 1b to c, d and e).

Conclusion

We conclude that the high temperature ground state can be best interpreted to arise from a strong interplay between trigonal crystal-field, magnetic exchange and spin-orbit interaction. The Verwey transition cannot therefore be viewed as an order-disorder transition of the trimeron quasi-particles.

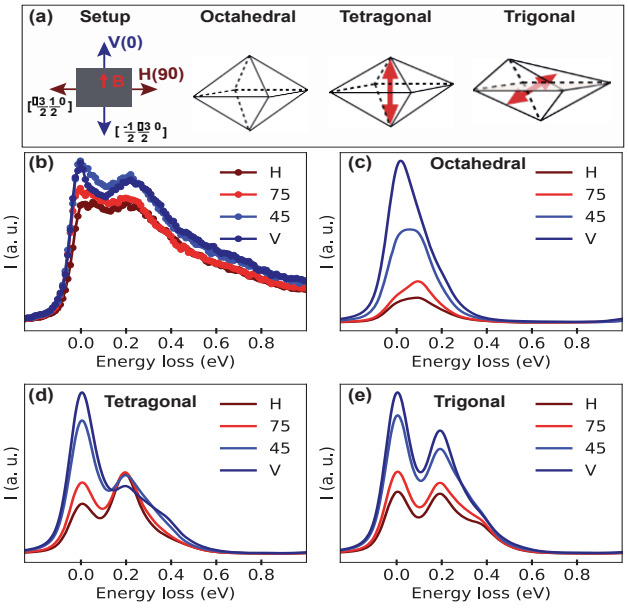

Figure 1: Fe 2p3d RIXS-MLD at E =706.1eV selective to Fe^{2+} ions. (a) Sketch of the setup and the three-local site-symmetries for the simulations presented in c, d and e. (b) Experimental RIXS-MLD. The angle is defined from the vertical polarization direction labeled V. Calculations of nominal Fe^{2+} ion in: (c) octahedral symmetry, (d) tetragonal symmetry according to the low temperature trimeron structure by [1] and (e) in trigonal symmetry.

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Magnetic contrast at spin-flip excitations: An advanced X-ray tool to study magnetic-ordering

PT-12

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Introduction

Magnetic phenomena associated with spin-orbit coupling, Dzyaloshinskii–Moriya interaction, exchange bias and magnetic anisotropy have led to a wide variety of emergent spin textures. The determination of the local orientation and magnitude of the magnetization in such systems plays a pivotal role in understanding and harnessing magnetic properties for technological applications. Here we employ the polarization dependence of resonant inelastic X-ray scattering (RIXS) to directly probe magnetic ordering with chemical and site selectivity by studying spin-flip excitations [1] in magnetite (see Fig. 1b).

Results and discussion

We measured high resolution Fe 2p3d RIXS on a (001) Fe_3O_4 synthetic single crystal at the ADRESS beamline of Swiss Light Source. A permanent magnet (B = 0.4T) was used to saturate the magnetization of Fe_3O_4. The incident polarization was rotated a full 180° as shown in Fig.1a in addition to circular. We observed strong polarization dependence at the spin-flip excitation (see Fig. 1c). Applied to ($[Fe^{3+}]_A[Fe^{3+},Fe^{2+}]_BO^{2-}{}_4$), we can distinguish spin-flip excitations of the anti-ferromagnetically coupled Fe^{3+} ions at the A and B sublattices. This is confirmed by the sign reversal of the RIXS magnetic circular dichroism (RIXS-MCD). Furthermore, by determining the angle at which the spin-flip excitation shows maximum intensity, we quantified the orbital contribution to the magnetic moment at the A sites. The angular dependence follows that expected for $\mathbf{l_z} \approx 0$ case as can be seen by comparing the experiment to the calculations (Fig. 1c and d) unlike previously proposed [2].

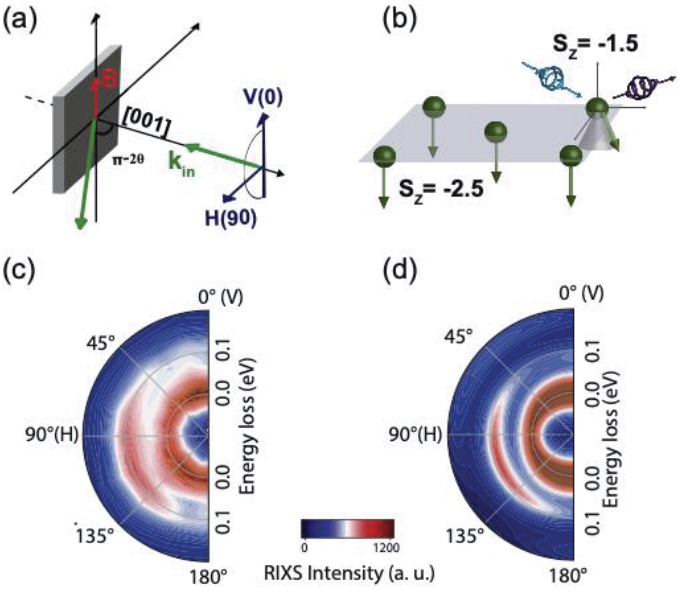

*Figure 1: Fe 2p3d RIXS-MLD at E =708.8eV selective to the Fe^{3+} A-sites. (a) Sketch of the setup. (b) A cartoon of a single spin-flip excitations at the A sites. (c) Experimental RIXS data measured as a function of the rotation angle of the incident polarization vector. The angle is defined from the vertical direction labeled **V**. The external magnetic field is aligned parallel to **V**. The data is presented in a polar contour plot where the radial axis gives the energy loss and the polar angle gives the angle be- tween **εin** and the vertical direction. (c) Calculations of the Fe 2p3d RIXS magnetic linear angular*

Conclusion

The magnetic contrast obtained at spin-flip excitations is an element and site selective probe of the exchange interaction. A thorough analysis of the angular dependence of spin-flip excitations can be used to identify orbital polarization. The potential to use this magnetic contrast in a RIXS-microscopy setup with nanometer resolution will open doors to study many interesting physics.

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Non-collinear ordering of the magnetic orbital moments in magnetite

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The orbital degree of freedom has been recognized as a key player in the determination of the ground state of many transition-metal oxides, yet its role in magnetite (Fe_3O_4) is still actively debated [1-3]. Here we investigate the orbital magnetic moment of Fe_3O_4 using a combination of Fe K-edge X-ray magnetic circular dichroism (XMCD) and 1s2p resonant inelastic X-ray scattering magnetic linear dichroism (RIXS-MLD). The reason we are able to shed new light is that we use the accurately measured full angular distribution of the RIXS signal to extract the mean square magnetic moment in addition to the mean quantity.

We investigated stoichiometric (001), (110) and (111) Fe_3O_4 single crystals. Based on the K-edge XMCD measurements, we quantified the average magnetic orbital moment of Fe in Fe_3O_4 to be $0.26 \pm 0.03\mu_B$. Comprehensive 1s2p RIXS angular dependence distribution revealed a subtle phase shift of the spectral intensity as a function of the magnetic field orientation. Fig.1a shows the 1s2p pre-edge RIXS dichroism revealing the *1s* to *3d* quadrupole pre-edge transitions in Fe_3O_4. Theoretical calculations revealed that the only possibility to explain our experimental results is the existence of a finite non-collinear magnetic orbital ordering that can tilt the orbital magnetic moments as large as 82° away from the spin magnetic moments (see Fig. 1c and d). We proposed a model for this non-collinear magnetic orbital ordering and inferred the presence of a dynamical distortion related to the X_3 phonon mode in the high temperature phase. Our findings suggest that Fe_3O_4 thin films will potentially key players in the emerging field surface orbitronics.

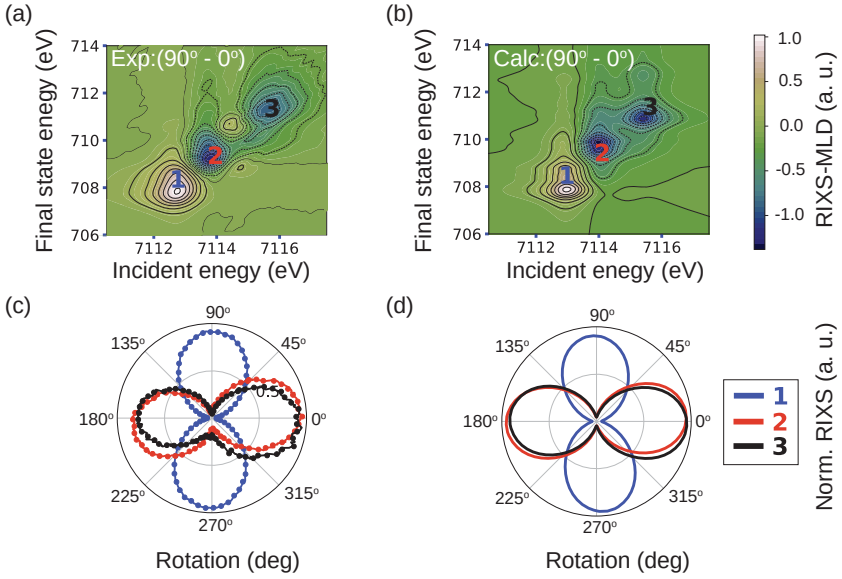

Figure 1: *Fe 1s2p RIXS dichroism map with vertical (90°) and horizontal (0°) linearly polarized beam. a) Experimental dichroism map. b) Theoretical dichroism map calculated taking into consideration the static and dynamical local distortion. The full 360° experimental and calculated angular dependence of the three main dichroic features (labelled 1, 2 and 3 in the RIXS map) are shown in panels c) and d).*

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PEAXIS – The new RIXS and XPS endstation at BESSY II

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The electronic states and their dynamics determine important functional material properties such as charge and, via electron-phonon coupling, also heat transport. Thus a profound understanding of the microscopic mechanisms underlying electrical conductivity and its coupling to thermal conductivity is gained by probing the electronic structure.

The new endstation PEAXIS built at BESSY II combines two important experimental methods for electron spectroscopy in a single UHV system. Resonant Inelastic X-ray Scattering (RIXS) and X-ray Photoelectron Spectroscopy (XPS) allow wave vector-resolved band mapping of electronic states in a broad range of functional materials. Research on PEAXIS focuses on solid state samples as the instrument covers a large wave vector range accessible by a continuous rotation of the RIXS spectrometer arm and the available solid-state sample manipulators cover a temperature range from 10 to 1000 K.

In this presentation we report on the capabilities of PEAXIS and show the first results of commissioning experiments. The beamline covers an incident energy range from 200 eV to 1200 eV and provides a beamspot size as small as 3.8 μm x 12.4 μm at the sample position. PEAXIS provides excellent energy resolution with an experimental resolving power exceeding 104.

Dispersing orbital excitations in the quasi-1D and 2D cuprates

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In correlated oxides orbital excitations tend to have a localized character and are usually described by orbital and spin quantum numbers in a symmetry adapted atomic picture: thus *dd* excitations do not disperse, irrespective of their spin character. However, theory predicts that at low dimensionality, *dd* excitations can split their orbital and spin components, giving rise to complex dispersion in momentum space [1]. Indeed, orbital excitations with sizeable dispersion, called orbitons due to their collective character, were observed with Resonant Inelastic X-rays Scattering (RIXS) on quasi-1D cuprates [2,3]. On the other hand, in 2D a sizeable dispersion was observed in RIXS *only* on the 2-1-4 iridium oxides, i.e. once a large spin-orbit coupling strongly modifies the energy scales [4,5].

Here, we present the RIXS measurements of dispersing orbital excitations on both quasi-1D and 2D systems. Regarding the quasi-1D Ca_2CuO_3, our results are consistent with those of Sr_2CuO_3 [2] but they are analyzed with an improved theoretical model. In particular, we verify how the Hund's exchange affects the observed spectra and whether a predicted interaction between spinon and orbiton can be observed [6]. For the quasi-2D $CaCuO_2$ and Nd_2CuO_4 cases, our RIXS experiments show a clear dispersion in the *dd* spectral region, which resemble the one of Ca_2CuO_3, but with different symmetry and periodicity (in particular for the *xy* and *xz/yz* orbitals). It had been suggested [1,7] that orbital excitations in a quasi-2D cuprate might still be dispersive, even though their dispersion is expected to be different to that in a quasi-1D cuprate. In order to get more insights and a better understanding of the orbiton dispersion, we additionally performed polarization-resolved RIXS at selected in-plane transferred momenta on both Ca_2CuO_3 and CaCuO) and we employ this enhanced experimental basis to refine the theoretical model of orbitons in one and two dimensional cuprates.

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Probing the RIXS Raman-to-fluorescence crossover in $BaFe_2As_2$

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Resonant inelastic X-ray scattering (RIXS) studies have significantly enhanced our understanding of correlated materials in recent years. However, experimental and computational efforts have been largely focused on insulating materials. An early exception was the investigation of the Raman-to-fluorescence cross-over in FeTe [1] for which experimental results were analyzed in the framework of the threshold singularity theory of Nozières and Abrahams [2]. We have performed related RIXS measurements on $BaFe_2As_2$ at the Fe L_3 edge. Band structure based Bethe-Salpeter equation calculations of the RIXS signal capture the incident energy dependent Raman-to-fluorescence cross-over as well as the main features observed in the experiment. These calculations account only for direct RIXS processes and neglect indirect RIXS processes, nevertheless they are able to reproduce the Raman-to-fluorescence cross-over. This indicates that at all incident energies the RIXS loss spectrum is effectively fluorescence-like and can be understood through comparison to the non-resonant emission. The apparently Raman-like spectra originate from the energy uncertainty of the intermediate state due to the core-hole lifetime. These calculations improve our ability to quantitatively and qualitatively separate simpler band structure contributions from more complex many-body effects in the RIXS spectra of correlated metals.

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The first experimental measurements of in-house X-ray resolution tests on a RIXSCam

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The resolution performance of RIXS experiments are limited by a combination of the source, the error on the grating, and the detector. Improvements in the spectrometer have resulted, in some cases, in the limiting factor being the spatial resolution of the detector. The use of CCDs in RIXS has been shown to have an intrinsic limit to the resolution obtainable due to the charge spreading in the detector material, which is linked to device active thickness and is independent of pixel size [1]. Using an EMCCD in "photon counting" mode, in combination with centroiding algorithms applied to single X-ray events, means that a sub-pixel resolution of 2-3 μm FWHM can be achieved from the 16 μm pixels [2] in the soft X-ray energy range. This has led to the development of the *RIXSCam*™ containing up to 3 large-area EMCCDs to increase the resolution performance of RIXS experiments as well as the photon throughput.

We have developed an in-house test system to explore aspects of the performance of the *RIXSCam*, particularly the spatial resolution performance. A resolution test system has been designed and constructed which allows us to conduct experiments to investigate the resolution performance of the *RIXSCam* detectors under various different operating conditions. We will present the experiment design and the first results exploring the effective resolution performance as function of CCD incidence angle and X-ray energy.

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Ab initio simulation of 1s2p resonant inelastic X-ray scattering spectroscopy

PT-18

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With resonant inelastic X-ray scattering (RIXS) experiments reaching high resolution (0.1 eV) in the energy transfer direction,[1] it becomes important to describe both multiplet effects and charge-transfer states in the hard energy region. A detailed interpretation and understanding of the X-ray spectra require accurate simulations, which can unravel subtle spectral features. To model the X-ray spectra, one high-level method that can describe important spectral effects, e.g, 2p and 3d spin orbit coupling (SOC), multiplet structures, selection rule, and charge transfer between metal and ligands is required. The *ab initio* restricted active space (RAS) method has been successfully used to simulate and interpret the different types of X-ray spectra.[2-6] In calculations, the most important orbitals are included in the active space. Not only the metal character core orbitals and 3*d* molecular orbitals, but also the important ligand molecular orbitals. The RIXS event can be thought of as a two-step process, which can be simulated by including different types of core orbitals into two sub-spaces in RAS calculation. The first application of RAS on 1s2p RIXS is performed,[4] see Figure 1. The ratio between cost and accuracy can be optimized by a proper selection of active space, basis set and computational algorithms, and the method can potentially be applied to both small and medium-sized systems.

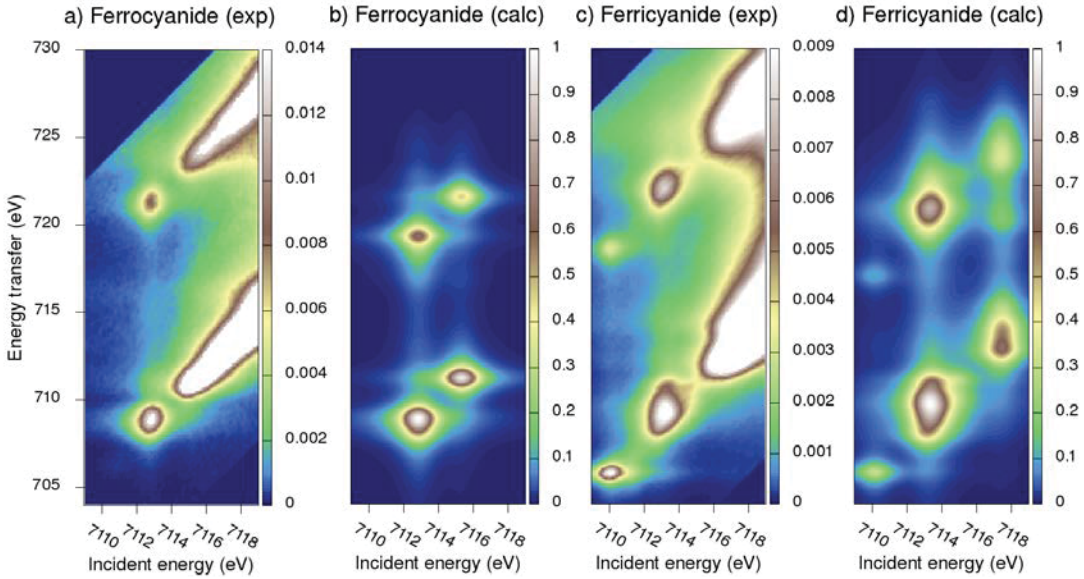

Figure 1. Experimental and RAS calculated 1s2p RIXS planes of $[Fe(CN)_6]^{4-}$ and $[Fe(CN)_6]^{3-}$. Experimental spectra are taken from Ref [3]. The rising edges are not described in the RAS spectra.

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Sum-rules in resonant and non-resonant IXS

PT-19

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On this poster, we will present several sum rules. We will relate the integrated intensity over resonant edges in non-resonant inelastic x-ray scattering to ground state expectation values. We will furthermore relate the integrated intensity over the resonant spectrum to an absorption spectrum.

Synchrotron and optical probing of hybrid organic-inorganic perovskite halides for photovoltaics

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The Goldschmidt's Tolerance Factor is a reliable figure of merit or empirical index to forecast the formation of preferred and stable structures and phases with ABX_3 hybrid organic-inorganic perovskite tolerance factors in the range of 0.9 to 1. Here, we probe perovskites of compositional variations ABX_3 with tolerance factors in the range of 0.9 to 1.0, and a large effective ionic radius greater than 200 pm. We report on the structural and optical properties of hybrid perovskite halides for photovoltaic (PV) applications, $MAPbI_3$, $FA_{0.8}MA_{0.1}Cs_{0.1}Pb(I_{0.86}Br_{0.14})_3$, and $FA_{0.8}Cs_{0.2}Pb(I_{0.86}Br_{0.14})_3$, with tolerance factors of 0.91, 0.964 and 0.967 respectively. PV devices were fabricated using these high tolerance factor hybrid perovskite halides. We report we have achieved power conversion efficiencies (PCEs) greater than 21% using the high tolerance factor perovskites investigated. Here we use synchrotron radiation to study and characterize the most predominant form of hybrid organic-inorganic perovskite halides, $MAPbI_3$ using spatially resolved X-ray absorption near edge structure (XANES) spectroscopy at the National Synchrotron Light Source (NSLS) II submicron resolution X-ray (SRX) beamline. XANES spectra were recorded with a double crystal Si (111) monochromator at the SRX synchrotron beamline 5-ID at NSLS II at Brookhaven National Laboratory (BNL). XANES is used here to probe the data on the electronic structure including the oxidation state and the chemical coordination. We have demonstrated a wide-bandgap (1.6 eV) perovskite solar cell with a PCE of 21.2% and a V_{oc} of 1.088 V, using formamidinium (FA) and cesium (Cs) cations, Pb, and mixed halide perovskites. The FEFF calculated orbital angular momentum projected local density of states (*l*DOS) provides detailed information on the origin of the spectral features. At the L_3 edge we probe $2p_{3/2}$ to 6*d* transitions in the presence of a core hole. The Pb *s*, *p* and *d* DOS contributions in $MAPbI_3$ at the Pb L_3 edge. The Pb *p* DOS is an intense sharp multipeak overlapping with Pb *d* DOS. The overlapping Pb *p* - *d* DOS contributions suggesting the presence of orbital mixing, and dominance of the Pb *d* DOS beyond the Fermi level, with spectral features dominated by the Pb *d* DOS, and resembling those in the Pb reference edge data. We observe that the $MAPbI_3$ system has a large resonance in the density of *p*-states before the Fermi level.

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Lattice dynamics evidence for a structural instability involving the conduction electrons in the unconventional superconductor $K_2Cr_3As_3$

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$K_2Cr_3As_3$ features a highly uniaxial crystal structure with quasi-one-dimensional electronic bands. The one-dimensional building blocks of the structure are stacked triangular clusters of Cr_3As_3. The arrangement of the large number of K guest ions in the unit cell breaks the inversion symmetry. The superconductivity observed below temperatures of T_c = 6.1 K [1] is of unconventional nature because of both, the broken inversion symmetry as well as the one-dimensional electronic structure.

In this study we present the lattice dynamics of $K_2Cr_3As_3$ as observed by high resolution inelastic x-ray scattering. Vibrational modes are identified by comparison to ab-initio calculations that match the data very well. The data were obtained in a temperature range between 25 K and room temperature. The highest energy Cr-derived mode with longitudinal polarisation displays a striking increase of the peak width at high temperature. While the increase is modest near the Brillouin zone centre Γ (l = 4.96 in Fig. 1) it is very pronounced in near the zone boundary point A (l = 4.65 and l = 4.5). This peak broadening gives mode- and momentum-resolved direct evidence of the suspected structural instability that could restore the inversion symmetry at high temperatures [2].

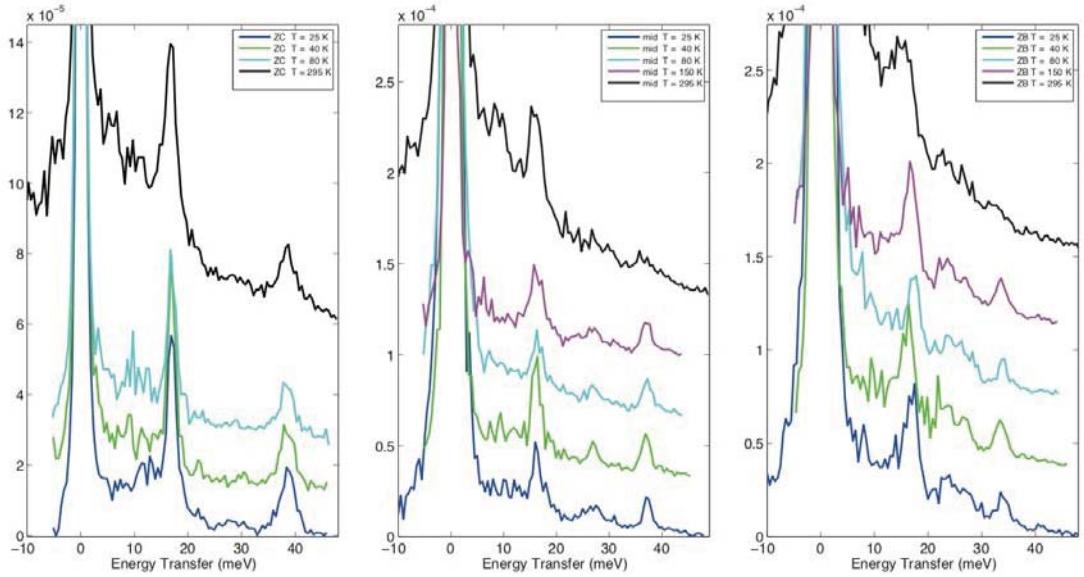

Figure. 1 IXS spectra as a function of temperature at three selected l = 4.96 (left), l = 4.65 (centre) and l = 4.5 (right). The spectra are offset by arbitrary amounts for graphical clarity.

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Spin fluctuations of multiferroic collinear magnet $Fe_2Mo_3O_8$ revealed by RIXS

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RIXS is a powerful experimental technique to explore charge and spin excitations of transition metal compounds. However, whether RIXS cross section truly reflects spin fluctuations across a phase transition is not etestablished yet. Here we demonstrate that soft X-ray RIXS reveals spin fluctuations across the phase transition of collinear magnet $Fe_2Mo_3O_8$. This system, which contains tetrahedral Fe^{2+} and octahedral Fe^{2+}, is a promising multiferroic material [1-2]. Below $T_N \sim 60$ K, $Fe_2Mo_3O_8$ exhibits a collinear antiferromagnetic order with spins parallel to the *c* axis. There is a simultaneous change of dielectric constant and electric polarization with the temperature induced magnetic transition across T_N. We present a study of polarization and temperature dependent RIXS on this collinear magnet. Through a special cross-polarized geometry, the temperature dependence of the quasi-elastic RIXS intensity in $Fe_2Mo_3O_8$ resemble that of the magnetic susceptibility of the system.

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Spin excitations in $La_{2-x}Sr_xCuO_4$: comparison of temperature dependence between RIXS and INS

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In hole-doped high-T_c cuprates, pseudogap in the range of temperatures just above the superconducting transition temperature (T_c) is an unsolved issue in addition to the mechanism of superconductivity [1]. In the pseudogap regime, electronic spectral weight in a part of the Fermi surface is suppressed below a certain temperature T^* ($> T_c$), which is quite unusual in normal metals. In a recent inelastic neutron scattering (INS) study of the hole-doped cuprate $La_{2-x}Sr_xCuO_4$ ($x = 0.075$) [2], high-energy spin excitations are found to show temperature evolution across T^*; a localized spin-wave-like dispersion of the paramagnon is observed up to 300 K ($< T^*$) and it changes to a broad ridge centered at the magnetic Brillouin zone center $\mathbf{q}_{AF} = (1/2,1/2)$ above T^*.

Encouraged by the possible relation between high-energy spin excitations and pseudogap, we performed a Cu L_3-edge RIXS experiment of $La_{2-x}Sr_xCuO_4$ ($x = 0.075$ and 0.12) focusing on the temperature dependence. The experiment was performed using the AGM-AGS spectrometer at beamline 05A1 of the National Synchrotron Radiation Research Center, Taiwan and we measured the spin excitations from 80 K to 400 K. The spin excitations in RIXS are found to soften with increasing temperature in both compounds. This unexpected softening is a stark contrast to the INS result because the aforementioned spectral change in INS is regarded that the spectral weight at a certain momentum away from $\mathbf{q}_{AF}$ moves toward higher energy with increasing temperature. It may imply that physical quantities observed with RIXS is not necessarily equivalent to those with INS even in the qualitative level. We note that the energy-momentum dependence above T^* in INS is similar to that of electron-doped cuprates [3,4] and high-energy shift of the spin excitations is consistently observed as an effect of electron doping in RIXS [4,5]. Possible origin of the contrasting temperature dependence between RIXS and INS will be discussed.

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RIXS Study of Uniaxially Strained Sr_2IrO_4

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It has been shown that the structural details, such as the Ir-O-Ir angle and the Ir-Ir distance, play an important role in influencing the magnetic properties of spin-orbital entangled iridates. However, microscopic understanding of how the electronic structure and magnetic interaction in the iridates respond to structural distortion is still lacking. The application of uniaxial strain within the ab plane can break tetragonal symmetry and induce the lattice distortion. In addition, the distortion can be continuously tuned, allowing one to examine the pseudospin-lattice coupling [1]. Using a piezoelectric-based strain apparatus [2], we have performed Ir L_3-edge RIXS measurements on a Sr_2IrO_4 single crystal under uniaxial tensile/compressive strain applied along the in-plane (1 0 0) and (1 1 0) directions. In both cases, we are able to strain the sample up to 10-3, and tune the magnetic domain structure in Sr_2IrO_4 accordingly. However, the maximum strain was still not large enough to change the magnetic excitation spectra observed with RIXS. We found that both zone-boundary energy and the in-plane magnon gap in Sr_2IrO_4 is relatively robust with respect to the uniaxial perturbation.

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RIXS investigations correlated electron materials at high-resolution beamlines

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Resonant Inelastic X-ray scattering (RIXS) is a powerful spectroscopy tool to investigate low energy excitations in correlated materials. Excitations of charge, spin, orbitals and lattice give rise to low energy excitations and interplay of these leads to emergent properties. Some of these emergent properties are metal to insulator transition, superconductivity, topological insulators and many more. During the past decade there has been numerous RIXS studies done on correlated materials especially high temperature superconductors. High resolution RIXS makes it possible to study these low energy excitations such as magnons [1, 2] and orbitons, which gives information about emergent properties. VERITAS beamline at MAX IV synchrotron (Lund, Sweden) has been designed with the aim to further refine the RIXS energy resolution. The beamline and spectrometer resolution (>35 000 resolving power), the capability to cool down to 4K (cryo-cooler) and the ability to change the scattering angle in the horizontal plane from 30 to 150 degrees (Q-chamber) [3] will able to access low-energy excitations in correlated materials with unprecedented quality. Furthermore, a goniometer supplement the Q-chamber, which permits crystal planes orientation and the mapping of dispersions over Brillouin zones.

Several projects on correlated materials will be investigated included the study of the hollandite K2Cr8O16 [4], spinel superconducting thin-films LiTi2O4 [5] and quasi two-dimensional Sr2Cu(BO3)2 [6]. K2Cr8O16 exhibits a metal to insulator (MI) transition preserving the ferromagnetic phase above and below the MI transition. RIXS will be used to investigate magnetic excitations, and orbital hybridization in this system. Molecular hybridization in LiTi2O4 will also be investigated below and above the superconducting transition. Sr2Cu(BO3)2 is a quasi two-dimensional material with an antiferromagnetic ground state characterized as Shastry-Sutherland square lattice. Preliminary M-edge RIXS (74 eV) investigations on Sr2Cu(BO3)2 show *dd* at 2 eV and charge transfer excitations at 6 eV. Due to the different configuration of the Cu ions in crystal there is a splitting in *dd* excitation. Further RIXS investigations of these compounds will be carried out.

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High resolution hard x-ray spectroscopy at PETRA III beamline P64

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PETRA III beamline P64 [1] is dedicated to advanced x-ray absorption and emission spectroscopy techniques. The beamline is based on the 2m undulator x-ray source, providing up to 10^{13} photons/s on a sample. Beamline's optics allows beam focusing down to 150x50 µm (HxV). These beamline's characteristics are the perfect basis for high energy resolution (non-)resonant hard x-ray emission (XES) spectroscopy. In recent years this technique is more and more used to determine the electronic and even geometric structure of matter by means of core-to-core (CtC) and valence-to-core (VtC) XES. Additionally, it gives an opportunity to perform resonant inelastic x-ray scattering studies (RIXS) and high energy resolution fluorescence detected XAS (HERFD-XAS) measurements.

For high energy resolution XES wavelength-dispersive spectrometers are used. They employ spherical, toroidal or cylindrical curved crystal geometry. In either case, it is possible to achieve a spectral resolution better than 1 eV. Such a resolution opens up new perspectives for investigations of materials in different sample environments at different conditions. As an example, hydrogen ligands in photochemical water splitting or other catalytic transformations can be investigated under reaction conditions, which is impossible with most other techniques.

In this contribution, we will present XES end station at PETRA III beamline P64, which is based on wavelength-dispersive von Hamos type spectrometer [2]. We will demonstrate its performance and capabilities on selected scientific examples. Additionally, an overview of ongoing projects at the beamline, related to the high-resolution XES will be covered. Particularly advancement of the new focusing spectrometer implementation and a work in progress towards time-resolved pump-probe XES experiments will be discussed.

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Flat-crystal based Resonant Inelastic X-ray Scattering Spectrometer for Measurements under High Pressure

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While recent advances in resonant inelastic X-ray (RIXS) scattering have led to various research achievements especially in iridates systems [1-3], there have also been theoretical and experimental predictions that some iridates systems will have novel magnetic physics upon applying pressures [4-6]. However, there has been a lack of any clear evidence for these predictions because of difficulties to measure their low-energy magnetic excitations. The difficulties stem from the fact that sample environments such as a diamond anvil, Be gasket, pressure media give strong noises around elastic signal and hinder us from obtaining a clean low-energy spectrum [7]. Here, we show that these extrinsic signals can be effectively removed by using a RIXS spectrometer based on flat-crystal optics at the Ir-L3 absorption edge (11.215 keV). This spectrometer, which was originally designed to achieve an energy resolution of sub-10 meV [8, 9], actually can be used as a spectrometer for investigating pressured samples as well, thanks to its collimating nature and limited field of view (FOV). Furthermore, we compare the efficiency of multiple options for RIXS spectrometers based on flat-crystal optics.

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Accessing fractionalized quasiparticle excitations in one-dimensional antiferromagnets using resonant inelastic x-ray scattering

PT-28

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One-dimensional (1D) magnetic insulators have attracted significant interest as a platform for studying quasiparticle fractionalization, quantum criticality, and other emergent phenomena. The spin-1/2 Heisenberg chain with antiferromagnetic nearest neighbor interactions is an important reference system; its elementary magnetic excitations are spin-1/2 quasiparticles called spinons. However, while the excitation continuum associated with two-spinon states is routinely observed, the study of four-spinon and higher multispinon states is an open area of research. In this talk, I will discuss how four-spinon excitations can be accessed directly in Sr_2CuO_3 using resonant inelastic x-ray scattering (RIXS) at its oxygen K-edge in a region of phase space clearly separated from the two-spinon continuum, both from a theoretical and experimental point of view [1]. These findings are made possible by the fundamental differences in the correlation function probed by RIXS in comparison to other probes and can be expressed in terms of effective correlation functions [2]. Finally, I will briefly present theoretical predictions for the excitations in the oxygen K-edge RIXS spectra of doped Sr_2CuO_3. The evaluated RIXS spectra are found to be rich, containing distinct two- and four-spinon excitations, dispersive antiholon excitations, and combinations thereof, and therefore motivate RIXS experiments on the doped 1D antiferromagnets [3]. All these findings improve the understanding of the RIXS as a probe for observing fractionalized quasiparticles and hold great promise as a tool in the search for novel quantum states and quantum spin liquids.

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RixsToolBox – An open source software for the analysis of RIXS data acquired with 2D position sensitive detectors

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A software with a graphical user interface has been developed, initially with the aim of facilitating the data analysis for users of a new resonant inelastic X-ray scattering (RIXS) spectrometer installed at the ESRF beamline ID32. The software is organized in modules covering all relevant steps in the data reduction from a stack of several hundred two-dimensional CCD images to a single RIXS spectrum. It utilizes both full charge integration and single-photon centroiding to cope with high-flux and high-resolution requirements. Additional modules for further data analysis and the extraction of instrumental parameters are available. The software has been in routine use for three years now and in that time many additional features have been incorporated. Currently, it can be used to with data acquired at the ESRF ID32 and Diamond I21 RIXS spectrometers, but support for other instruments/data formats is easily added. We hope to provide the user community with an easy-to-use data analysis tool that allows analyzing data quickly as it is acquired and thus helps users to conduct their experiments more efficiently.

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The RIXS spectrometer at the ESRF soft X-ray beamline ID32

PT-30

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The RIXS spectrometer at the ESRF beamline ID32 has marked a leap forward in soft X-ray RIXS instrumentation. As the first of its kind it implemented several advanced features for the first time which are now becoming more and more available in several other places, too: continuous in-vacuum rotation of the scattering arm allowing for complete control of the scattering geometry, combined resolving powers E/dE in excess of 25,000, and polarization analysis of the scattered X-rays [1, 2]. After three years of successful user operation during which all design goals had been reached, operation stopped at the end of 2018 with the beginning of the ESRF EBS storage ring upgrade. Here we give a status report of the instrument and its performance during the first three years of operation, and present plans for an approved upgrade to further advance the spectrometer in terms of resolution and throughput. The instrument will become available again with the restart of ESRF user operation in August 2020, most likely already in fully upgraded form.

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RIXS investigation of dd excitations in α-$RuCl_3$

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α-$RuCl_3$ is drawing much attention as a candidate material for Kitaev quantum spin liquid, and its magnetic Hamiltonian is being scrutinized carefully [1]. One of the most important experimental parameters to be determined is the hierarchy of various energy scales, including spin-orbit coupling and crystal field splitting. We probed these energy scales directly using resonant inelastic x-ray scattering (RIXS) at the Ru M edge. The sharp A1 peak in Fig. 1 at 235 meV originates from local J_{eff} 3/2→1/2 transitions which are predicted at 195 and 234 meV for a t_{2g}^5 final state [2]. The energy dependence supports this conclusion since A1 resonates below the main peak on a shoulder, whereas the higher energy peaks with e_g states, i.e. $t_{2g}^4e_g$ or $t_{2g}^3e_g^2$ final state, resonate at the main peak. The A1 peak is not split, indicating very little trigonal distortion. If we take 39 meV from theory [2] as an upper limit and 157 meV SOC from our results, the ratio between the energies still supports a J_{eff} picture in α-$RuCl_3$. Our results are extremely promising and establishes M-edge RIXS in the 4d transition metals.

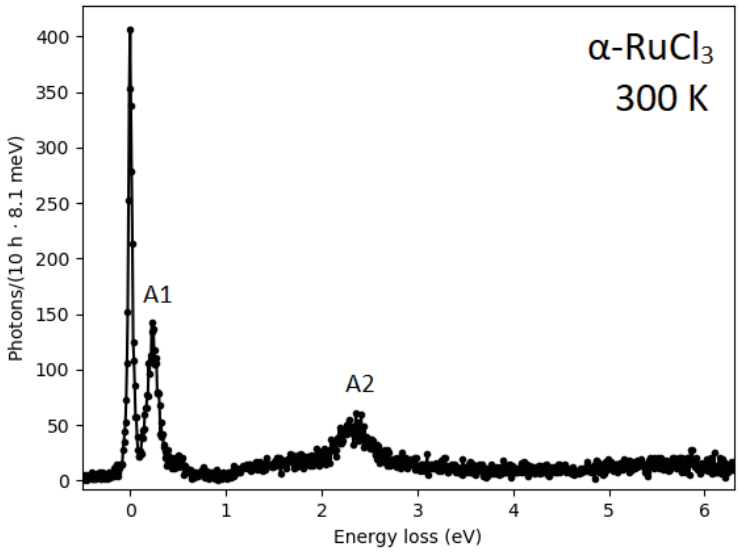

Figure 1: Ru M_3-edge RIXS taken on SIXS spectrometer of 2-ID at NSLS-II. A 500 l/mm monochromator grating, 20 um exit slits, and 1250 l/mm spectrometer grating are used for a total energy resolution of 37 meV (resolving power of ~12500 at 461 eV resonance). The sample was mounted with (100) and (001) laying the horizontal scattering plane which was set at 90 degrees. This spectrum was measured at 20 degrees grazing incidence and with LH polarization.

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Resonant inelastic x-ray scattering study of charge density wave in optimally doped $La_{2-x}Ba_xCuO_4$ using transition edge sensor detector

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Charge density wave (CDW) order in cuprate families has been of great interest due to its intimate connection with superconductivity (SC). In order to understand the interplay between CDW and SC, it is crucial to determine how the emergence of the SC phase affects charge ordering behavior when the two phases coexist with similar energy scale. However, detecting charge order in optimally doped materials with energy-integrating x-ray scattering techniques has been very challenging since the charge scattering is distributed over a large range of momenta, making it difficult to discern from the fluorescence background. Here, we present resonant inelastic x-ray scattering measurements of CDW order in optimally doped $La_{2-x}Ba_xCuO_4$ ($x = 0.155$, $T_c = 30$ K) at the Oxygen K edge with an innovative transition edge sensor (TES) array detector. Utilizing the energy resolving capability and the unprecedented high sensitivity of the TES [1], we detected weak and short-ranged charge correlations up to T = 115K, which is much higher than the reported CDW onset temperature [2], while rejecting the inelastic fluorescence background. On cooling, we observed that the correlation length of CDW order increases as SC emerges, suggesting CDW and SC may have positive correlation. Our observations open up possibilities of using TES for studying short-ranged orders in other optimally doped cuprates.

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The Origin of Ising Magnetism in $Ca_3O_2O_6$ Unveiled by Orbital Imaging

PT-33

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One-dimensional CoO_6 chains in $Ca_3Co_2O_6$ give rise to a unique Ising-like magnetism that shows intriguing quantum tunneling phenomena in the magnetization [1]. During the previous two decades a considerable debate has emerged regarding the underlying local electronic configuration of the Co ions in this material. To resolve the issue, we applied *s*-core-level non-resonant inelastic x-ray scattering (*s*-NIXS), a new technique that allows the direct imaging of the shape of the *hole* density of the 3*d* orbitals (Fig.1), as our group has recently demonstrated [2]. Concerning $Ca_3Co_2O_6$, we were able to segregate the spectral features that belong to both the prismatic trigonal coordinated, and the octahedral coordinated Co ions (Fig.2). The orbital shapes that we found (Fig.1) established unequivocally that both Co sites are in the 3+ valence state (i.e. $3d^6$), with the prismatic Co having the high-spin state and the octahedral Co the low spin. Interestingly, we 'see' that it is the complex d_2 orbital that is stabilized by the prismatic trigonal coordination, which naturally explains the Ising magnetism in the system. Harnessing this ability to directly relate the orbital occupation with the local crystal structure—without the need for theoretical modeling—is essential for modeling reliably the magnetic properties. Conceivably, this technique opens new avenues for the prediction and design of novel materials with unusual or optimized properties.

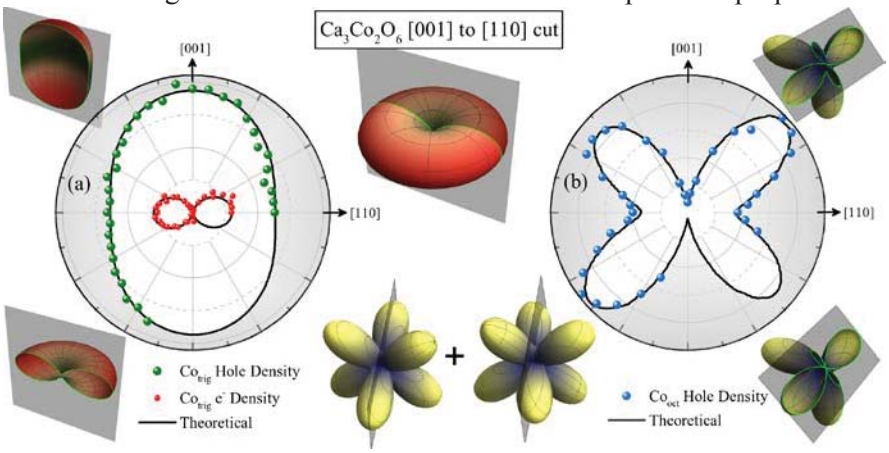

Figure 1. (a) The experimental electron density (red dots) in the [001] to [110] plane of $Ca_3Co_2O_6$ displays the donut-like cross sectional shape of the complex d_2 orbital (shown in the red three-dimensional images). **(b)** The total hole density for the octahedral site consists of two equivalent sites that are rotated 45° from one another about the *c*-axis. The 3D images show the overall hole densities of the two sites, and the corresponding cut we measure in our experiment.

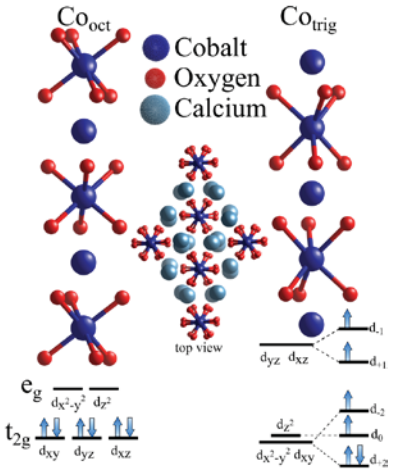

Figure 2. The low spin d^6 orbital occupation of the octahedral site (left) allows for only e_g holes. While the high spin magnetically active trigonal site (right) that leads to the Ising magnetism has its sixth electron in the d_2 orbital.

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Ultrafast dynamics of spin and orbital correlations in layered iridates

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Many remarkable properties of quantum materials emerge from states with intricate coupling between the charge, spin and orbital degrees of freedom. Ultrafast photo-doping of these materials holds great promises for understanding and controlling the properties of these states [1, 2]. Here I will present our recent time-resolved X-ray scattering results on the layered iridate $Sr_3Ir_2O_7$, where we have studied the time evolution of its magnetic properties after excitation with a 2 um optical laser pump. The material reveals an intermediate Coulomb repulsion and strong spin-orbit coupling that generates an effective J_{eff} =1/2 antiferromagnetic Mott ground state on the verge of a paramagnetic phase. The strongly reduced insulating Mott-gap leads to appreciable deviations from the isotropic Heisenberg model that is applicable for the related compound Sr_2IrO_4. As a result, stark differences in the ultra-fast dynamics of the photo-excited transient state are observed between the two materials. This behavior will be discussed in the context of a proposed "spin bottleneck" effect.

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Spin excitations in ultrathin superconducting cuprates

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The physics of quasi-two-dimensional high-T_c superconducting cuprates is enclosed in the CuO_2 planes but, due to technical limitations, the experimental investigation of these materials is normally carried out on bulk 3D samples. Here we study a few isolated layers of doped $La_{1.84}Sr_{0.16}CuO_4$ (LSCO) down to 1 unit cell thickness by means of momentum-resolved Resonant Inelastic soft X-ray Scattering (RIXS) at the Cu L_3 edge. We find that the RIXS spectra of the ultrathin films show dispersing spin excitations comparable to the ones in bulk [1] but with reduced dispersion dynamics, and hence diminished magnetic coupling, for decreasing thickness. We combine the RIXS data with high-resolution Scanning Transmission Electron Microscopy (STEM) measurements [2] that show, with picometric accuracy, a decreasing distance of the apical oxygens from the CuO_2 planes for decreasing number of cuprate layers. The reduced magnetic coupling in thinner films can therefore be ascribed to a stronger degree of hybridization of the $3d_{z^2}$ orbital with the $3d_{x^2-y^2}$. This is induced by the shorter apical oxygen distances at the interface with the substrate and it is detrimental for the in-plane hopping integrals that govern the magnetic super-exchange J. Our observations within one single cuprate family complement previous works comparing different undoped cuprates [3] and are well in agreement with the most advanced *ab initio* quantum chemical calculations reporting the correlation between J and apical oxygen distance [4]. Our work thus sets a new approach for the study of extremely thin layers of materials based on the combination of state-of-the-art RIXS and STEM.

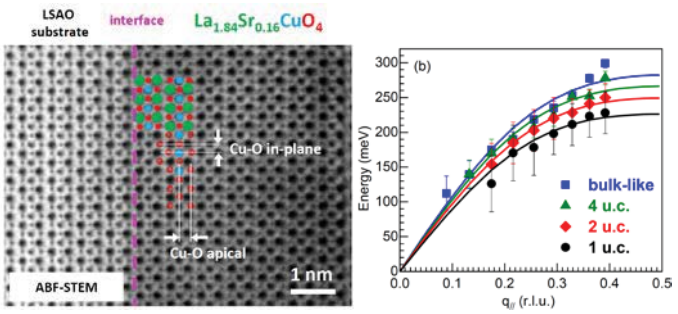

Figure STEM image in Annular Bright Field acquisition mode for a 4-unit-cell-thick LSCO film (left) and dispersions of magnetic excitations extracted from RIXS for ultrathin films with different thickness (right).

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Resonance behaviour of magnon and double-magnon excitations in NiO

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Abstract

In the early days of development of resonant inelastic x-ray scattering (RIXS) technique, possibility of probing spin-flip excitations at $L_{2,3}$ absorption edges of transition metals was predicted in the light of strong spin-orbit coupling present in the intermediate core-hole state [1]. Experiments have demonstrated *collective* nature of spin-flip excitations for both S=1/2 and S=1 spin systems [2-5]. NiO, an antiferromagnetic insulator, owing to $3d^8$ electronic configuration of Ni^{2+} ions, stabilizes in a high-spin S=1 ground state. Significant amount of work has been done over the years using RIXS to reveal high energy multiplet states in this prototype material as well as low energy magnetic excitations [3,5]. Unlike S=1/2 systems where spin-flip ΔS can at most be 1 (S=1/2→-1/2), in S=1 systems ΔS can go upto 2 (S=1→-1) resulting in the observation of double-magnons along with magnon excitations [3,4].

Since RIXS proceeds via a virtual intermediate state which is the final sate of XAS, it is expected that the inelastic features show strong resonance nature with the incident energy across the absorption edge. For example, it was predicted recently that dynamical charge structure factor can be enhanced by tuning not to the Cu L_3-edge but to its sattellite [6]. Bi-magnons at Cu K-edge have also been predicted to show different resonance based on effective core-hole screening of the intermediate states [7].

From Ni L-RIXS (~45 meV energy resolution) at I21 beamline, we observe a radically different resonance behaviour of magnon and double-magnon (both spin-flip excitations) in NiO as shown in Fig. 1(a). The plots show integrated intensity values of the magnon and double-magnon inelastic features in RIXS spectra obtained for incident energies varying across the Ni L_3-edge. Inset to Fig. 1(b) shows a representative RIXS spectrum used to extract the intensity values. These experimental observations are well-reproduced by crystal-field full-multiplet calculations using Quanty where RIXS transition probabilities were found to follow fundamental x-ray absorption spectral functions. [8,9]. A prior knowledge of incident energy suited to enhance an inelastic feature of interest shall benefit future experiments.

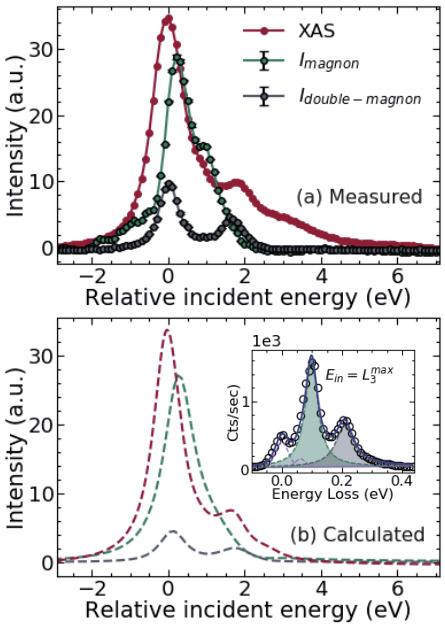

Fig. 1: Resonance of magnons and double-magnons as a variation of incident energy relative to NiO L_3*-edge electron yield maxima.*

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Analysis of magneto-orbital excitation from iridates via resonant inelastic scattering in a weak-coupling itinerant electron approach

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Recently we have developed an effective formalism to investigate low energy excitation spectra detected by *L*-edge resonant inelastic x-ray scattering (RIXS) [1]. The formalism, which was constructed on the basis of the calculation of the density-density correlation function, was applied to some iridates and succeeded in elucidating how J_{eff}=1/2 physics works (or breaks) to some degrees in such systems [2, 3]. Through the results of these works, we come up with the conclusions that (i) nearest neighbor hopping models give rather good results with the help of the itinerant electron treatment and (ii) magnetic excitations consist of several dispersion bands, the number of which are more than detected within the present experimental energy resolution.

In this work, we study another iridates such as $CaIrO_3$ and $Sr_3Ir_2O_7$ and try to confirm whether the above conclusions are valid in these systems. To this aim, by using the anisotropic effective hopping integrals between the nearest neighbor Ir sites, we first derive an effective Hamiltonian to describe the low energy physics of the material. Then, we calculate excitation dispersions and RIXS spectra. Since our treatment utilize the mean field approximation to provide the ground state and random phase approximation to evaluate the excitation states, our results have weakness quantitatively. However, semi-quantitatively, we are confident to assert that the calculated results are in accordance with the above conclusions for $Sr_3Ir_2O_7$ while not so simple for $CaIrO_3$, probably, due to low symmetry crystal structure (*Cmcm*).

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Temperature and doping dependence of the crystal field excitations in $La_{1-x}Sr_xVO_3$ family of compounds near the Mott insulator limit

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Bulk $La_{1-x}Sr_xVO_3$ is a Mott insulator for $x<0.18$. For $x=0$ it exhibits a structural phase transition from an orthorhombic to monoclinic structure at 140 K and a paramagnet to antiferromagnet transition accompanied by spatial ordering of orbital occupations [1]. For $0.18<x<0.3$, the system is metallic with low temperature antiferromagnetic ordering. Long-range magnetic order is suppressed for $x>0.3$. The importance of spin and orbital fluctuations on the charge transport properties in the vicinity of the metal-insulator phase boundary has been discussed in prior a study [2]. In order to understand the rich phase diagram of this family of rare-earth vanadates electronic, lattice, spin and orbital degrees of freedoms have to be taken into account. We have therefore investigated the doping and temperature dependence of the low energy crystal field excitations in high quality $La_{1-x}Sr_xVO_3$ thin films near the Mott insulator limit ($x=0$, $x=0.1$) with resonant inelastic X-ray scattering (RIXS) at the O K- and V L- edges. RIXS is a bulk-sensitive element-specific technique that can be used to probe the neutral excitations of a system. In particular, RIXS is able to directly probe spin and lattice dynamics simultaneously with the orbital excitations, and therefore offers an unprecedented view on the low energy properties of $La_{1-x}Sr_xVO_3$. This technique was used to study the closely related vanadate compound YVO_3, in which a possibly dispersive low energy peak near 200 meV was found and interpreted as arising from a collective orbital excitation [3].

Our doping dependent study reveals how the Sr to La chemical substitution affects essentially only the oxygen site local electronic structure of this doped Mott insulator, while our temperature dependent study of the pristine $LaVO_3$ compound connects the evolution of the low energy orbital excitations with the phase transitions occurring at 140 K.

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Lattice tuning and magnetic frustration in Sr_2IrO_4 and $Sr_3Ir_2O_7$ as seen by RIXS and Raman scattering

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Exploring the physics of the iridates is expected to shed light onto high-temperature superconductivity as well as provide valuable insights into the interplay of spin-orbit coupling (SOC), Hund's and Coulomb interactions. Of particular interest are low-energy magnetic excitations in one- and two-layer perovskites Sr_2IrO_4 and $Sr_3Ir_2O_7$. In this talk, I will first discuss how magnetic degrees of freedom in iridates can be manipulated by applying external pressure and strain. Since SOC-mediated magnetic momentum locking to the lattice has been reported in iridates in 2018[1], tuning local crystal environment has emerged as a new route to control structural and physical properties. Due to this momentum locking, pressure and strain in iridates are believed to have direct impact on magnetic excitations, which I will address using exact diagonalization in connection with recent discoveries by resonant inelastic x-ray scattering (RIXS) and x-ray Raman scattering. I will next discuss the effect of frustration on two-magnon excitations. For $Sr_3Ir_2O_7$, two-magnon Raman spectra have been reported to vary and lack simple correlation with single-magnon bandwidth [2]. We show that magnetic and finite spin gap are responsible for the unexpectedly low two-magnon peak in $Sr_3Ir_2O_7$. We also present a modified spin wave approach for an intuitive explanation of the numerical results. Our analysis highlights magnetic frustration as one of the important differences between single- and double-layer perovskites iridates.

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Development of X-ray Crystal Analyzers

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X-ray crystal analyzers are the key for success in application of XIS and XES, as well as XAFS with laboratory x-ray sources. In the last 16 years, we developed most analyzers to cover such applications, which include spherically bent analyzers, cylindrically bent analyzers, both with diced, striped and stand free type, and also few Johansson type of cylindrical and spherical curvature. The energy resolution can be down from meV of stand free type to few eV for very short radius analyzers.

Our patented spherically striped-bent crystal analyzers open a door to very short curvature radius analyzers, both in silicon and germanium. Our anodic bonding stand free flat-diced analyzers is also proved its value in supper high energy resolution for XIS.

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Cooperative valence dynamics in Anderson lattices observed by resonant inelastic x-ray scattering

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In rare earth intermetallics with weakly bound *f*-electrons and a Kondo energy scale much larger than magnetic exchange interactions or crystal field splittings, the screening of local moments may result in a non-magnetic Fermi liquid ground state [1]. At low temperatures, the quantum fluctuations between magnetic and non-magnetic valence configurations can then acquire a cooperative (lattice) character. On a phenomenological basis, a sound understanding of this *Anderson Lattice* phenomenon has been achieved. On the other hand, the microscopic description of the coherent coupling between Kondo-screened sites remains an outstanding theoretical challenge [2]. In experiment, the cooperative character of Anderson Lattices has only recently become directly accessible. Momentum-resolved spectroscopies, such as angle-resolved photoemission and inelastic neutron scattering, reveal the emergence of characteristic low-energy quasiparticle dynamics at low temperatures [3]. These methods probe single-particle excitations in the charge and magnetic channels, respectively. By contrast, high-resolution resonant inelastic x-ray scattering (RIXS) experiments couple to both charge and spin degrees of freedom in a non-trivial way and thus provide a more subtle point of view. We used this method to probe the elementary excitations in an archetypal intermediate valence material, $CePd_3$. At the crossover into the low-temperature cooperative state, we observe a change-over in spectral weight between different interband transitions. At low temperatures, dispersive trends in the spectra reveal the strange band-character of these quasiparticle states, which is clearly at odds with the traditional impurity approach to the Anderson model. If calculations of the underlying Kramers-Heisenberg term on a basis of strongly correlated *f*-electronic bands are achieved, RIXS may unlock unprecedented microscopic insights into the entanglement of local and itinerant charge and magnetic degrees of freedom. This would address a fundamental mechanism of quantum matter, with relevance far beyond lanthanides and actinides.

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Resonant Inelastic X-ray Scattering investigation of itinerate states in Rare Earth Hexaborides

PT-42

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We present X-ray Resonant Inelastic X-ray Scattering (RIXS) data collected at the L edges of divalent hexaborides YbB_6 and EuB_6. Our incident-energy-, momentum-, and polarization-dependent measurements display two features separated by 10 eV in incident energy, which appear to be well described using density functional theory calculations related to unoccupied 5d bands in these materials. Polarization-dependent data shows anomalies particularly at high incident energy in the divalent materials and we propose a process which describes the origin of this RIXS intensity. Our results suggest far-reaching utility of L-edge RIXS in determining the itinerant state properties of f-filling materials at a microscopic level.

Aberration-free imaging of inelastic scattering spectra with x-ray echo spectrometers and spectrographs

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X-ray echo spectroscopy, a space-domain counter-part of neutron spin echo, was introduced recently to overcome the limitations in spectral resolution and weak signals of the traditional inelastic hard x-ray scattering probes. X-ray echo is refocusing of the defocused x-ray source image. X-ray echo spectroscopy relies on imaging IXS spectra and does not require x-ray monochromatization. Due to this, the echo-type IXS spectrometers are broadband, and thus have a potential to simultaneously provide orders-of-magnitude increased signal strength, reduced measurement times, and higher resolution compared to the traditional narrow-band scanning-type IXS spectrometers. The detailed theory and optical designs of x-ray echo spectrometers were presented in [1,2].

Here we study conditions for aberration-free imaging of IXS spectra with x-ray echo spectrometers [3]. Aberration-free imaging is essential for achieving instrumental functions with high resolution and high contrast. Computational ray tracing is applied to a thorough analysis of a 0.1-meV-resolution echo-type IXS spectrometer operating with 9-keV x-rays. We show that IXS spectra imaged by the x-ray echo spectrometer that uses lenses for the collimating and focusing optics are free of aberrations. When grazing-incidence mirrors (paraboloidal, parabolic Kirkpatrick-Baez, or parabolic Montel) are used instead of the lenses, the imaging system reveals some defocus aberration that depends on the inelastic energy transfer. However, the aberration-free images can be still recorded in a plane that is tilted with respect to the optical axis. This distortion can be thus fully compensated by inclining appropriately the x-ray imaging detector, which simultaneously improves its spatial resolution. A full simulation of imaging IXS spectra from a realistic sample demonstrates the excellent performance of the proposed designs.

We will give an overview of the basic principles of IXS imaging with x-ray echo spectrometers, explain the choice of their design parameter, and provide examples of numeric simulations results of aberration-free imaging of IXS spectra with the 0.1-meV resolution. The results are equally applicable to x-ray echo spectrographs [4], which are a subsystem of x-ray echo spectrometers.

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Electronic and Magnetic Excitations of Cuprates $La_{2-x}Sr_xCuO_4$ Revealed by Soft X-ray RIXS

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The study of elementary excitations in cuprates is of great importance to reveal the spin fluctuations and the electron-phonon coupling in high-temperature superconductors. Here we report O K-edge resonant inelastic scattering X-ray scattering (RIXS) measurements to reveal charge-transfer, bi-magnon and phonon excitations in undoped and underdoped $La_{2-x}Sr_xCuO_4$. With an improved energy resolution of 25 meV, we observed charge-transfer excitons of Zhang-Rice spin singlet and bi-magnon excitations, which consist of coherent excitations involving spin flips of two neighboring sites with opposite direction. The energy and dispersion of the observed bi-magnon excitations lend support to the existence of magnon-magnon interaction is the undoped cuprate. Implications of these observed electronic excitations will be discussed in detail.

Numerical study of crystal optics misalignment effect on the performance of high energy-resolution focusing monochromator.

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Combination of x-ray focusing and crystal optics introduced a new concept of focusing monochromator into the IXS instrumentation [1]. The principle of operation of the x-ray focusing monochromator is very similar to that of the Czerny-Turner monochromator [2] used in infrared, visible, and ultraviolet spectroscopies. The general idea behind it is to provide spatial separation of the focused beams according to their energies using angular dispersion of the diffracting optic. Thus, the beam with the desired energy can be selected by an aperture in the image plane of the focusing optic. Here, a further development of a focusing monochromator concept for X-ray energy resolution of 0.1 meV and below is presented. The main accent of the research was done on the investigation of the focusing monochromator tolerance to the alignment imperfections of crystal optics, like non-coplanarity of the diffraction vectors of the constituent crystals. The numerical simulations revealed that the energy bandwidth of the focusing monochromator is much more sensitive to the angular misalignment of the monochromator crystals than that of a conventional high energy resolution monochromator. It is very important to consider this factor in designing IXS instruments like echo spectrometer [3].

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Pseudospin energy levels in the Kitaev spin liquid candidate α-$RuCl_3$ revealed by resonant inelastic x-ray scattering at the Ru L_3 edge

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α-$RuCl_3$ is one of the major candidate materials for the realization of Kitaev spin liquid state [1]. With increasing experimental evidence for fractionalized excitations, it is pivotal to quantify the energy scales of microscopic parameters. We studied the charge and spin excitation spectra of α-$RuCl_3$ by means of resonant inelastic x-ray scattering (RIXS) at the Ru L_3 absorption edge. The RIXS experiment was performed with the intermediate-energy RIXS (IRIXS) spectrometer at the P01 beamline of PETRA III, DESY. We successfully observed a sharp peak originating from the ionic transition from the J_{eff} = 3/2 to the J_{eff} = 1/2 states, thereby obtaining a quantitative estimation of the spin-orbit coupling constant. This transition does not show a splitting within our experimental error and the peak intensity is weakly dependent on momentum, indicating the highly cubic crystal field conditions around the Ru^{3+} ions. Furthermore, the quasi-elastic intensity composed of the J_{eff} = 1/2 sector has a broad intensity maximum at the Γ point, consistent with the inelastic neutron scattering data [2]. These findings provide strong evidence that the magnetism of α-$RuCl_3$ is in close proximity to the Kitaev model.

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2p3d RIXS as a complement to L-edge XAS

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XAS experiments at the metal L-edges can be simulated successfully via multiplet calculations, which provide a general framework to understand the spectra of both molecular and solid samples. Although XAS has been an important technique for both qualitative and quantitative analysis of electronic structure, there are significant disadvantages to measuring a one-dimensional spectrum. Because XAS results from an integration over all emission energies and decay channels, the measured spectrum is insensitive to changes in the emitted energy. Two simulations which would produce different RIXS planes may integrate to the same XAS spectrum, introducing uncertainty into analysis. Many models, especially based on multiplets, do not include either the continuum background or excitations to 4s states, but in measured XAS, these are simply added together with the excitonic states. Additionally, PFY-XAS often suffers from a combination of saturation and self-absorption, which can make analysis difficult.

The situation is radically different for RIXS, where measurement of the emission energy immediately provides enhanced separation between different states. Rather than measuring a high-resolution emission spectrum at a selection of incident energies, we focus on low-resolution RIXS measurements of the entire L3-L2 edge as an enhancement to XAS and PFY-XAS. In the RIXS plane, the onset of continuum emission, which is non-resonant, can be clearly identified. Coster-Kronig decay is also visible at the L2 edge. Furthermore, simulations of the RIXS plane can be corrected for both saturation and self-absorption, unlike XAS, where energy dependent re-absorption of the emitted energy is not considered.

We will present a series of RIXS experiments on nickel oxide and nickel hydroxide that demonstrate the utility of this approach when fitting a series of similar compounds. Our measurements were performed using a spectrometer on SSRL BL10-1 based on superconducting Transition-Edge Sensors (TES) [1], which have an energy resolution of ~2 eV. The TES is a broadband instrument capable of measuring energies from 200 eV to 1600 eV simultaneously, so our approach couples 2p3d RIXS with 2p3s PFY-XAS and Inverse Partial Fluorescence Yield to fully understand the measured spectrum. We find that RIXS is useful for qualitative fingerprinting, interpreting the PFY-XAS spectrum, and quantitative fitting via charge-transfer multiplet codes.

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Determining the Ground State and the Crystal Field Splitting of MnS by Orbital Imaging

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The often spectacular properties observed in materials containing transition metal (TM) elements continue to challenge our comprehension of solid state physics. In order to develop adequate models we need to determine the charge, spin, and orbital degrees of freedom of the TM ions.

We are exploring the opportunities offered by non-resonant inelastic x-ray scattering (NIXS) with high-momentum transfers, by which we can observe dipole-forbidden transitions. Focussing on transitions involving an s-core level (s-NIXS), we have recently shown that the shape of the TM 3d hole density can be directly imaged by mapping out the orientation dependence of the TM 3s→3d integrated intensity [1]. We are now taking the next step, namely to also investigate the orientation dependence of the spectral line shape. Using MnS as an example, we can resolve two final state features: one can be identified directly as e_g and the other as t_{2g} from their orbital shapes (see Fig.1), while the sum of the two yields a spherical shape confirming the high-spin $3d^5$ configuration for the Mn. We can also directly determine the octahedral crystal field splitting to be about 0.6 eV.

We conclude that s-NIXS has the ability to unveil the local electronic structure without the need for calculations. The information is given directly by the orbital maps. This unique power of s-NIXS is related to the fact that the s-core hole is spherical symmetric, which does not alter the orbital part of the multiplet structure of the final states.

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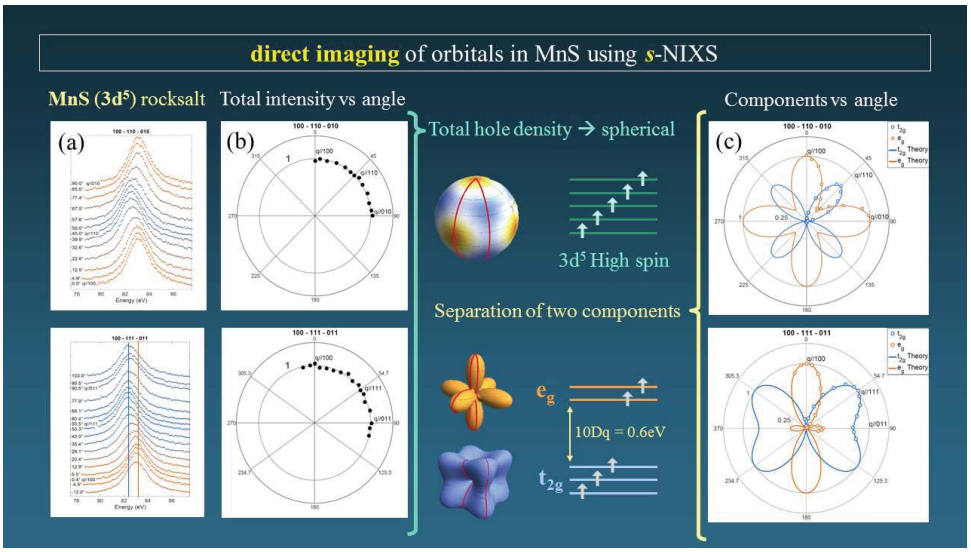

Figure 1. (a) Mn 3s→3d (dipole forbidden) inelastic scattering spectra taken for a series of angles between the momentum transfer direction and the crystallographic directions of MnS. **(b)** A polar plot of the energy-integrated Mn 3s→3d spectra. **(c)** A polar plot of the high and low energy components of the Mn 3s→3d spectra. **Middle panel**: total hole density of the Mn 3d and the 3D images of the high and low energy components of the Mn 3s→3d spectra.

Observing dynamical critical scaling with RIXS

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Various thermodynamic quantities, including the magnetization, susceptibility, and heat capacity, exhibit power law scaling in the vicinity of a second-order critical point at T_c. This is a consequence of the magnetic correlation length $\xi \sim |T\text{-}T_c|^{-\nu}$ and response time $\tau \sim |T\text{-}T_c|^{-z\nu}$ both diverging at T_c, where ν and z are examples of critical exponents. The values of these critical exponents directly reflect the spin and lattice dimensionality of the magnetic fluctuations. In particular, they are sensitive to the relevant interactions driving the phase transition, and weak anisotropies that may not be directly observable with other methods.

The resonant x-ray scattering (RXS) cross-section is (approximately) proportional to the spin-spin correlation function in reciprocal space. In the elastic channel, this enables the determination of the static critical exponents β, ν, and ξ, related to the magnetization, susceptibility, and correlation length respectively. For instance, magnetic critical scattering measurements on Sr_2IrO_4 have shown the presence of weak easy-plane anisotropy [1]. Meanwhile the interactions in $Sr_3Ir_2O_7$ fall into the 3D Ising universality class, and are perturbed by disorder [2].

The dynamic critical exponent z is typically difficult to measure, yet is a key quantity for determining whether a system exhibits a quantum critical point (QCP) or has some degree of itinerant magnetism. I will show that it is possible to determine z using resonant inelastic X-ray scattering (RIXS). I will compare the results for different 5d transition metal oxides, in particular $(Sr_{1-x}La_x)_3Ir_2O_7$. I will also show how it should be possible to extend the technique to other systems.

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Analysis of Device Fabrication's Process for Optimization of Perovskite Solar Cells

PT-50

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The External Quantum Efficiency (EQE) is a reliable factor to measure the quality of a perovskite solar cell as it forecasts the efficiency of the cell at a range of wavelengths of light. Absorbing light at different wavelengths is a fundamental property perovskite solar cells must present. Here, we probe a hybrid organic-inorganic perovskite cell structure formed by a fluorine-doped tin oxide (FTO), cassiterite ($Sn0_2$), perovskite, Spiro OMETAD and silver layers. We report we have achieved power conversion efficiencies (PCEs) greater than 21% using the device fabrication's process investigated. Photovoltaic (PV) devices were fabricated using $MAPbI_3$ as the dominant form of hybrid organic-inorganic perovskite halides due to its low price and manufacturing simplicity, making $MAPbI_3$ a potential light harvester. Chlorobenzene (CBZ) was used to increase crystal size of the perovskite helping carrier transport. Here we used an FTO glass as the transparent conducting oxide as it provides low resistance properties and its transparency makes it adequate for absorbing light at different wavelengths by the perovskite absorber layer. Spiro OMETAD was deposited using spin coating, helping to an increase of efficiency as it enhances hole transportation. We used a silver layer as a reflective back contact helping any fraction of light to be reabsorbed by the device. Here we used X-ray diffraction (XRD), photoluminescence (PL), dark and light current-voltage (I-V), and EQE measurements to characterize the quality of the junctions, power efficiency, fill factor, series and shunt resistance. We have demonstrated a PCE of 21% and a V_{oc} of more than 1.1 V for a wide-bandgap (1.6 eV) perovskite solar cell. We also observed the importance of an even coating of CBZ as crystals generated helps the creation of a successful p-n junction capable of capturing light at different band gaps. We observed that photocurrent increases with temperature until 333 K, but suffers a phase transition at higher temperatures due to a large resonance in the p-states density.

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2p3d RIXS of Ferric Porphyrin Complexes

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Iron porphyrin systems are essential to all life and are involved in numerous biological functions including oxygen transport, small molecule activation, and redox reactions. The electronic structure of the iron atom controls its ability to perform specific chemical functions. Despite many years of study, understanding of the reactivity of iron porphyrins is complicated due to the presence of overlapping high-intensity bands in the UV/Vis spectra originating from the porphyrin ring. In this study 2p3d RIXS has been used to directly probe the electronic structure at the iron center in these Fe complexes. Comparison of high and low-spin Fe(III) tetraphenyl porphyrins (FeTPP) shows that the spin state can directly be determined from the 2p3d RIXS spectra. These model complexes are compared with the RIXS spectrum of the metalloenzyme cytochrome c whose distinct spectrum show shows how the protein environment tunes the electronic structure of the Fe atom.

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The ESRF Crystal Analyser Laboratory (CAL), a laboratory completely dedicated to the development of spherically (cylindrically) bent analyser crystals for spectroscopic studies, has been operational since 2015. The CAL covers an area of roughly 120 m^2 including a "clean room" of 60 m^2 where the main processing tools are installed. At the moment the CAL is developing analyser crystals for 9 multi-analyser spectrometers at the ESRF but also provides optics to other X-ray light sources around the world. The domain of applications for such analyser crystals is quite wide including chemistry (energy storage materials, batteries), physics (correlated electron systems, liquid and glass dynamics), earth science, environmental science, catalysis, materials science and biology.

The laboratory is equipped with a combination of commercial instruments (wafer grinder, wafer dicing saw) as well as in-house designed fabrication and characterisation systems (for anodic bonding, cylindrical surface polishing, glue dispersion and inspection). The CAL manufactures essentially 3 types of analyser crystals (bent, bent-striped [1] and diced) mainly using Si, with the different types satisfying the various experimental conditions required by our scientists in terms of energy resolution, intensity and collected solid angle.

This talk will give an overview of the different technologies used for crystal analyser manufacture, describe the different processing capabilities of the CAL and describe the main development activities which are currently in progress. In particular, the latest improvements for the production of Von Hamos cylindrical analysers with radius R = 0.5 and 0.25 m will be shown and for the fabrication of both "single side" and "double side" machining of Johansson cylindrical analysers with radius R = 1 m.

A new system of vacuum-clamped analyser devices developed at the ESRF will also be presented. The system is intended primarily to host Ge bent and bent striped analysers and is similar to that presented by Jahrman [2] but much more compact in order to install many chambers in one line or in array. The preliminary results on Ge bent analysers indicate good performance and we intend to install this system in many ESRF multi-analyser spectrometers.

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Structural properties of actinides applying $M_{4,5}$ edge high resolution XANES and 3d4f RIXS

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We have recently demonstrated, also for elements beyond uranium (U) such as neptunium (Np) and plutonium (Pu) that high-energy resolution X-ray absorption near edge structure (HR-XANES) and resonant inelastic X-ray scattering (RIXS) spectroscopy at the actinide (An) $M_{4,5}$ absorption edge (3-4 keV) provide new insights into the An electronic structure and speciation.[1-2] The potential of these methods and an overview of recent applications will be presented. The experiments were performed at the ACT (actinide) experimental station of the CAT-ACT beamline for catalysis and actinides at the Karlsruhe Research Accelerator (KARA), Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany.[3] A Johann-type X-ray emission spectrometer with vertical Rowland circle geometry comprising five analyzer crystals with 1 m bending radius operated in He environment was applied.[3]

We will discuss the role of the 5f valence orbitals of U, Np and Pu in the chemical bonding. The level of localization and participation of the actinide (An) 5f valence orbitals in covalent bonds across the actinide series is one of the long-standing debates in actinide chemistry. An $M_{4,5}$ HR-XANES and 3d4f RIXS results reveal that the 5f orbitals are active in the chemical bonding for U and Np, shown by significant variations in the level of their localization evident in the spectra. In contrast, the 5f orbitals of Pu appear localized and less sensitive to different bonding environments. It will be shown that the An $M_{4,5}$ absorption edge HR-XANES and 3d4f RIXS can help to distinguish between the classical notion of overlap-driven covalency and energy-driven covalency. Apparently our results suggest that the overlap-driven covalent character of the An-O bonding in the actinyl cations decrease within the U-Pu series, while the energy-driven covalent character increases without increasing the electron density of the bonding.[1]

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Energy and angular dependence of saturation and self-absorption effects in resonant inelastic X-ray scattering

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Resonant Inelastic X-ray Scattering (RIXS) is a developing technique to investigate the ground and excited states with its angular degrees of freedom. Its scattering cross-section is based on the Kramers-Heisenberg formula which can be explained from two processes: absorption (photon-in) and emission (photon-out). The absorption process excites the electrons to the higher energy levels and the emission process emits the photons when the electrons decay to the lower energy levels. These scattered photons from the ions will be reabsorbed in the emission process. Consequently, the emitting intensity might be distorted according to the photon energy and system geometry. This saturation and self-absorption effect is a well-known behavior which introduces the energy dependence or angular dependence in fluorescence yield X-ray absorption spectroscopy (XAS)[1-3]. As such, it also distorts the RIXS spectra.

We focus on the saturation and self-absorption effect in the *2p3d* RIXS. We show that the effects distort the spectra not only as function of the incident photon energy but also as function of the emitted photon energy. For the *2p3d* RIXS, the emitted signal can be classified into two parts: the *dd* excitations and the fluorescence. When excited at the L_3 edge, the emitted photons' energies of *dd* excitations are close to the L_3 edge absorption, where strong angular dependence due to the saturation and self-absorption effects is presented. In contrast, the fluorescence feature is ~2.5 eV higher than the elastic peak in L_3 edge absorption, i.e., the emitted photons' energies are 2.5 eV smaller than the main line of the L_3 edge. This implies that the absorption factor is considerable smaller than for the main line of L_3 edge.

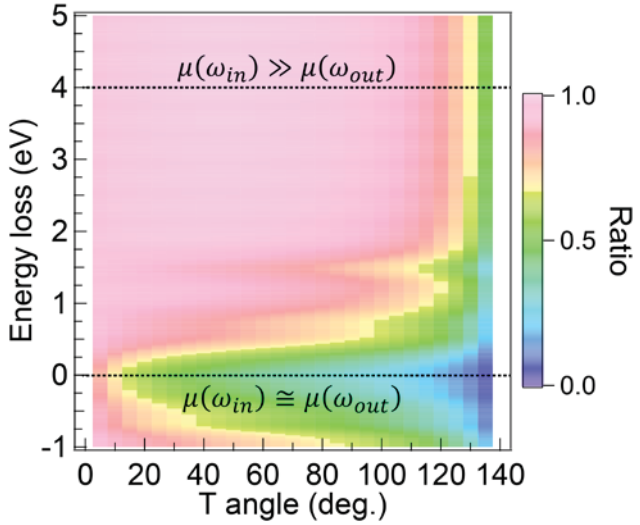

Figure: The correction coefficients as a function of rotation angle and emitted photon energy for low spin Co^{3+}.

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The angular dependence of orbital excitations in strained $LaCoO_3$ films

PT-55

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Bulk $LaCoO_3$ shows a diamagnetic to paramagnetic transition at ~100 K. It has been discussed as a spin state crossover from a low-spin (LS, S=0) state to a high-spin (HS, S=2) state or to an intermediate-spin (IS, S=1) state[1,2]. In the last decade, long-range ferromagnetic order has been observed in epitaxial $LaCoO_3$ thin films and described as the exchange interaction between LS and HS Co^{3+} ions[3]. Recently, Fumega and Pardo proposed that the ferromagnetic state is more stable in the structure with the oxygen vacancy order which implies Co^{2+} ions can be involved[4], where Co^{2+} ions are always found in a magnetically active HS state. However, a later article indicated that a large amount of oxygen vacancies (>10%) can strongly reduce the magnetism[5].

For Co^{3+}, the *2p3d* Resonant Inelastic X-ray Scattering (RIXS) allows us to distinguish the spin state manifolds[2,6] and to reveal the electronic structure of the strained $LaCoO_3$ films. We observed a low energy excitation at ~0.2 eV on the tensile-strained $LaCoO_3/SrTiO_3$ film, while it is not observed in neither bulk $LaCoO_3$ nor the compressive-strained $LaCoO_3/LaAlO_3$ film. Through the angular and polarization dependence, the 0.2 eV feature can be identified as an orbital excitation from the $^5B_{2g}(D_{4h})$ state to the $^5E_g(D_{4h})$ state. In comparison with the single cluster simulation, it suggests that the HS Co^{3+} plays an important role in long-range ferromagnetic order on the strained $LaCoO_3$ films.

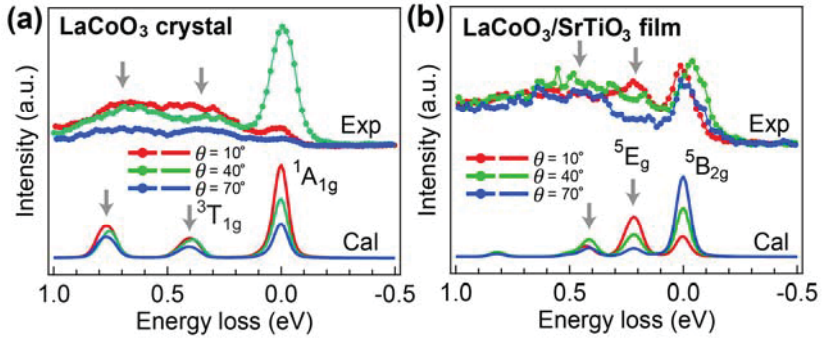

Figure: The angular dependent *2p3d* RIXS of $LaCoO_3$ crystal and the $LaCoO_3$ film on $SrTiO_3$ with respect to the rotation of sample.

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EDRIXS: An open source toolkit for simulating spectra of resonant inelastic x-ray scattering

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In this talk, we present an open source toolkit (dubbed EDRIXS) [1] to facilitate the simulations of RIXS spectra of strongly correlated materials based on exact diagonalization (ED) of certain model Hamiltonians. The model Hamiltonian can be from a single atom, small cluster or Anderson impurity model, with model parameters from density functional theory plus Wannier90 or dynamical mean-field theory calculations. The spectra of x-ray absorption spectroscopy (XAS) and RIXS are then calculated using Krylov subspace techniques. This toolkit contains highly efficient ED, XAS and RIXS solvers written in modern Fortran 90 language and a convenient Python library used to prepare inputs and set up calculations. We first give a short introduction to RIXS spectroscopy, and then we discuss the implementation details of this toolkit. Finally, we show several examples [2] to demonstrate its usage.

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Inelastic X-ray Scattering under High Pressure at 16 ID-D of HPCAT

PT-57

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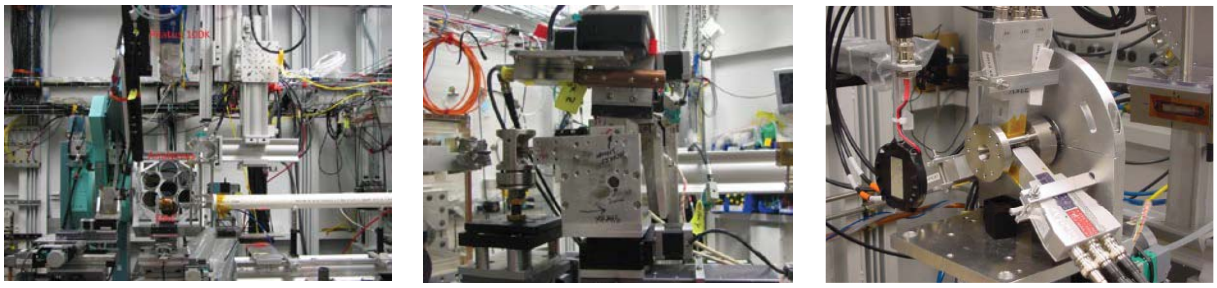
The structural, electronic and magnetic properties of materials under high pressure are of fundamental interest in physics, chemistry, materials science, and earth sciences. The 16 ID-D beamline of the High Pressure Collaborative Access Team (HPCAT) at the Advanced Photon Source (APS) is dedicated to high pressure research using X-ray spectroscopic techniques typically integrated with diamond anvil cells. The beamline provides X-rays of 4.5-37 keV, and current available techniques include X-ray emission spectroscopy, inelastic X-ray scattering and nuclear resonant scattering [1,2].

Firstly, we discuss on the particular requirements and instrumentation for X-ray spectroscopic techniques under high pressure and give a general description of 16 ID-D beamline. We then present several examples to illustrate the recent progress in high pressure inelastic X-ray scattering studies at HPCAT, followed by a discussion on recent developments include XRS spectrometer with a polycapillary full lens [3], cryostats for low temperature and high pressure XRS and NRIXS. With the newly installed polycapillary full lens as post sample collimation slit, XRS spectra above megabar pressure was measured at the first time [4]. An outlook toward future development in high pressure nuclear resonant and inelastic X-ray scattering with new diffraction-limited storage rings (APS-U etc.) is also discussed.



(a) (b) (c)

Figure (a) XES spectrometer with 7 analyzers, (b) XRS instrumentation with polycapillary full lens as post sample collimation slit, (c) high pressure NRIXS measurements with three APDs.

This work is performed at HPCAT (Sector 16), Advanced Photon Source (APS), Argonne National Laboratory. HPCAT operations are supported by DOE-NNSA's Office of Experimental Sciences. The Advanced Photon Source is a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

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Feasibility study of elemental mapping using ratio of elastic and inelastic back-scattered X-rays

PT-58

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Elemental kinds, composition ratios, and spatial distributions are the most basic information on materials and determine the physical and chemical properties of materials. However, the conventional X-ray fluorescent analysis cannot detect such elemental information in deeper area of samples because of the small escape depth of fluorescent X-rays. We have developed a novel elemental imaging method using back-scattered X-rays. The intensity ratio of the back-scattered elastic (Rayleigh) and inelastic (Compton) X-rays (RC ratio) depends on the Zeff (effective atomic number) [1, 2], and the energy of back-scattered X-rays depends on that of incident X-rays, and it can be set as desired. Therefore, elemental maps can be obtained by combination of the spectrum analysis of the back-scattered high-energy X-rays and the X-ray scanning method.

We demonstrated the feasibility of observing a phantom that consisted of a polyethylene ring and aluminum and copper blocks covered by an aluminum plate with a 1-mm thickness by using synchrotron radiation (SR) X-ray at the beamline 07 (BL07) of SAGA Light Source in Tosu, Japan. A pencil monochromated SR X-ray beam in the shape of a 0.5-mm square (25-keV) was formed by using the X-ray beam slit and scanned on the phantom by using the sample positioner. The X-rays scattered at each point on the sample were detected by a silicon drift detector (SDD) with a 1-sec counting time at a scattering angle of 135 degrees, and the sample was set at an incident angle of 45 degrees. The scanning step and number of both x- and z-directions were 1 mm and 81 points, respectively, and, therefore, the scanned area was an 8-mm square.

A fine elastic, inelastic, and RC ratio images were obtained. Cross-sections of elastic scattering are positively correlated with the atomic number, and, therefore, the copper area was visualized with bright contrast. On the other hand, cross-sections of inelastic scattering are negatively correlated with the atomic number, and, therefore, the polyethylene area was visualized with bright contrast. The RC ratio was found to be positively correlated with the atomic number, and each material was successfully visualized in the order of atomic number, that is, copper, aluminum, and polyethylene. Namely, an elemental map of the inner area was successfully obtained by using back-scattered X-rays in front-side observation. The approximated quadratic equation used to calculate the ratio between the RC ratio and Zeff was given, and Zeff of each region in the phantom can be calculated with the accuracy of $< \pm 1$. In addition, cluster analysis of a map of elastic and inelastic X-ray intensities was applied, and fine segmented image of the phantom was obtained.

These results show that the newly developed method provides a way to obtain elemental information in deep area of samples, so observing and analyzing the elements of various samples, such as magnets, electronic devices, and biomedical tissues, are expected to be possible. In addition, fine observation with a nm-order spatial resolution is also expected to be possible with focused X-ray micro beams and coherent X-ray diffraction.

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