ESRF meV-IXS Program Overview

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ID28 ESRF beamline is in user operation for meV-IXS studies since about twenty years. The main operational resolution is 3 meV; 1.5 meV is available on request. Single crystals are the main class of studied objects, while soft condensed matter retains some contribution.

The diffraction/diffuse scattering side station can operate in parallel with the spectrometer at the same wavelength; user experiments can be allocated for the use of single branch or both of them since 2016. Diffuse scattering provides the roadmaps for the inelastic scattering studies, thus significantly extending the throughput of main station.

The planned upgrade of optics, accompanied by beam position stabilisation, will reduce the conventional focal spot size down to 5-10 μm for both stations. Also, significant count rate gain is envisaged with the new generation of undulators.

Diffuse Scattering in Strongly Corelated Systems

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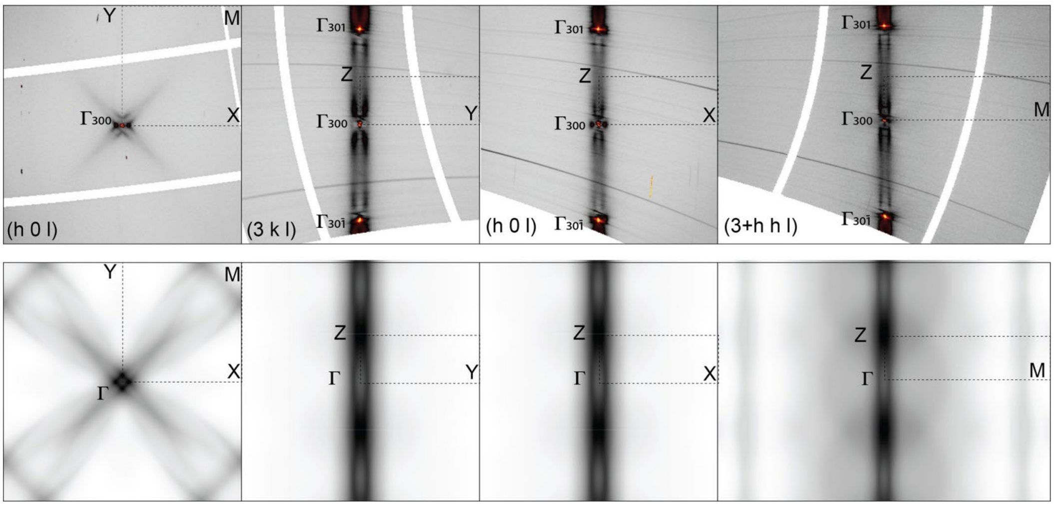
Measuring the phonon dispersions can be complemented and sometimes even replaced by the measurement of thermal diffuse scattering. In the latter case energy-integrated intensity maps can be collected very fast in large regions of reciprocal space. The newly constructed diffraction/diffuse scattering station of the ID28 ESRF beamline [1], in conjunction with the existing inelastic X-ray scattering spectrometer, provides a unique opportunity of combined studies.

Quasi-2D correlated systems appear to be particularly favorable for the mapping of nesting-related anomalies by TDS. The obtained information corresponds to the bulk properties, which, for instance, can bring new insights to the interpretation of results of surface-sensitive photoelectron spectroscopy.

Among the successful applications we can cite the following systems:

- $LaAgSb_2$ – soft phonon instability with extremely large real space periodicity [2]
- $ErTe_3$ – phonon anomalies, giving rise to two families of incommensurate modulation

In both cases the momentum transfer resolution of the diffuse scattering measurements was critical for the analysis, otherwise a full tracing of anomalies with inelastic scattering alone would become extremely difficult if even possible.



Comparison of DS maps of $LaAgSb_2$ with the corresponding calculated maps of intraband electronic susceptinility

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Disorder-Phonon Coupling in the Correlated Disorder System γ-($U_{1-x}Mo_x$)

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Understanding the role of disorder, and the correlations that exist within it, is one of the defining challenges in contemporary materials science. However, there are few material systems, devoid of other complex interactions, that can be used to systematically study the effects of correlated disorder arising from crystallographic conflict. The pseudo-*bcc* uranium molybdenum system is however an exemplar case study and as such we fabricated thin (~ 300 nm) epitaxial films of γ-($U_{1-x}Mo_x$) alloys in the range $0.17 < x < 0.31$ to be studied that the ID28 beamline (ESRF, France).

We established, via extensive diffuse x-ray scattering studies [1], that the intrinsic symmetry conflict; where uranium, which prefers a locally anisotropic environment, is forced into an isotropic *bcc* global symmetry, produces a new form of intrinsically tuneable correlated disorder where every atom is displaced to form a short-range superstructure with correlations existing over nanometre sized regions. This serves to modulate the local crystallographic periodicity, thus giving rise to the possibility of a form of disorder-phonon coupling. To investigate this possibility one alloy composition was measured with grazing incidence inelastic x-ray scattering (GI-IXS) and compared with extensive *ab-initio* modelling [2].

We discovered strong disorder-phonon coupling that relaxes degeneracy conditions at the **P** position, hardens the LA-2/3$\langle 111 \rangle_p$ mode ubiquitous to monotonic *bcc* crystals and produces significant phonon linewidth broadening at almost all positions in the 1st Brillion zone [1]. This broadening is almost entirely due to the presence of correlations, and results in a strong reduction of the phononic contribution to thermal conductivity. Similar effects are expected in any system able to host comparable levels of crystallographic conflict and may be exploited when designing future functional materials. As the first IXS study, to our knowledge, on a thin film heterostructure composed of multiple single crystal layers this work also highlights the power of GI-IXS and may serve as a blueprint for future studies of thin film structures or devices with similar or greater complexity.

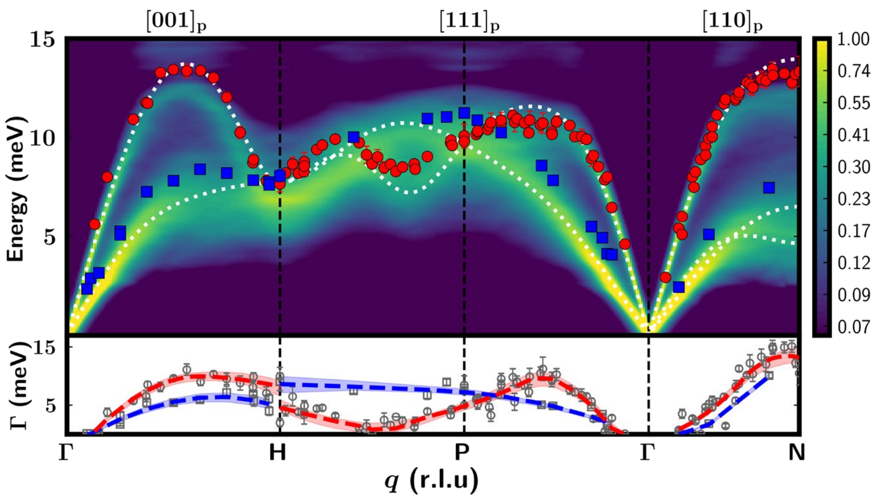

Figure 1: (Top Panel) Experimental (x = 0.23) and theoretical (x = 0.25) phonon dispersion curves. Transverse (longitudinal) acoustic modes are shown as blue squares (red circles) and theoretical results for two separate theoretical approaches are shown as dashed white lines and as a colour map, respectively. All directions are within the bcc BZ. (Bottom Panel) Raw linewidths are shown as grey squares (TA) and circles (LA) with deconvoluted linewidths shown by dashed blue (TA) and red (LA) trendlines with errors shown as confidence bands.

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IXS for ferroic transitions: archetypal antipolar soft mode in francisite $Cu_3Bi(SeO_3)_2O_2Cl$

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Inelastic scattering techniques at large have been for decades instrumental in investigations of ferroic transitions and their structural instabilities. When it comes to instabilities at the zone boundary, inelastic x-ray (IXS) or neutron scattering are typically required. Here, we apply IXS to studies of antiferroelectricity.

Antiferroelectric materials (AFE) undergo a structural phase transition from a high-symmetry to a low-symmetry phase where antiparallel arrays of electric dipoles emerge or order [1, 2]. In a direct analogy with antiferrodistortive transitions, one might in principle conceive an ideal AFE phase transition as being driven by an "antipolar" soft-mode, i.e. a soft phonon mode at the zone boundary involving antiparallel ion displacements and connected via the phonon branch to a polar mode at the zone center with parallel displacements of the same ions. However, in practice, this simple scenario has never been demonstrated. Instead, known AFE transitions are dominantly of the order-disorder type or display complex behaviours where antiparallel cation displacements are only part of the story, as in the model $PbZrO_3$.

Here, we investigate the orthorhombic francisite $Cu_3Bi(SeO_3)_2O_2Cl$. At 115 K this compound undergoes a structural phase transition characterized by antiparallel displacements of the Cl and Cu atoms along the *a* axis, leading to a doubling of the unit cell along the *c* axis [3]. The small but clear dielectric anomaly and the nearly degenerate energies of the non-polar and polar polymorphs [4] tend to confirm the AFE character of the transition, even though the switching under electric field has not been reported yet.

We found evidence for a soft mode both below and above T_c by means of, respectively, Raman spectroscopy and a combination of Inelastic X-ray scattering (IXS) and thermal diffuse scattering (TDS). In both cases, the soft-mode could be fitted with a damped harmonic oscillator and its energy squared was found to follow a linear behaviour as a function of temperature in a large temperature range around T_c (Fig. 1). Altogether, this confirms the scenario of a simple displacive antipolar transition [5].

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Tuning of charge density waves in correlated metals – New results and insights

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External control of electronic phases in correlated-electron materials is a long-standing challenge of condensed-matter research. In the recent years it has been realized that the underlying crystal lattice was more than a mere spectator and could be used as an insightful tuning knob. In the first part of this talk, I will show how the combination of pressure (hydrostatic or uniaxial) tuning and inelastic x-ray spectroscopy has been used in the course of the last decade to gain fresh insights on the properties of CDW in high temperature superconducting cuprates [1-3].

The second part of the talk, I will focus on $BaNi_2As_2$, a non-magnetic analogue of the parent compound of Fe-based superconductors. In this material incommensurate and commensurate CDW have been recently reported and yield a new form of electronic nematicity which strongly couples to the crystal lattice.

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Anisotropic phonon line-broadening in UO_2 below room temperature

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The interaction between the lattice and magnetic excitations in UO_2 was first observed in 1966 [1]. Many neutron studies have been reported over the years, with the latest in 2011 [2]. In 2014 [3] there was a report from bulk property measurements showing that the thermal conductivity of UO_2 below 300 K is anisotropic, which is formally forbidden in a cubic system. The interactions in UO_2 are extremely complex [4], but to understand the thermal conductivity we need a close examination of the phonon linewidths at low temperature.

We have undertaken X-ray inelastic scattering using ID28 to measure the phonons in UO_2 from a single crystal. X-rays have the advantage that they are insensitive to magnetic and quadrupole effects (providing the energy is far removed from resonances) and are sensitive only to the vibrational spectra, and any broadening of them caused by their interactions with the electronic system.

The experiments show large line-broadening effects in the transverse acoustic phonons in the [100] direction due to the electronic-lattice interactions, and no measurable effects in the [110] direction. This anisotropy is consistent with the lower thermal conductivity measured in the [100] direction [3]. The effects extend to at least 200 K, consistent with short-range magnetic correlations that extend to at least this temperature [5]. These measurements also help to understand the large differences between the thermal conductivity of ThO_2 and UO_2. However, by 300 K any line-broadening is negligible, consistent with the recent work of Pang *et al*. [6].

In addition, we have observed a quasi-elastic signal (i.e. at zero energy transfer) that appears to peak at ~ 40 K (*above* T_N = 31 K), and, in contrast with the phonon line-broadening, appears isotropic. The origin of this quasi-elastic is currently uncertain, but may be a signal of polaron formation due to the local oxygen displacements and the quadrupoles coupling to the lattice. An initial effort to observe possible diffuse scattering at low temperature did not succeed.

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High-resolution inelastic X-ray scattering at LCLS-II-HE: preliminary plans and outlook

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LCLS-II-HE upgrade will deliver two to three orders of magnitude higher average spectral flux (photons/s/meV) compared to storage-ring-based X-ray sources. This development can potentially extend the reach of inelastic X-ray scattering (IXS) to unexplored territories like measurements with signals that are currently too weak to detect. However, a more pronounced advantage of an IXS spectrometer at an X-ray FEL is likely related to the ultrashort pulses that are unique to LINAC-based sources. This new advancement will enable interrogation of collective modes associated with short-lived transient phases of materials that are controlled by various stimuli, including ultrafast mid-IR and THz pulses. In order to exploit this capability, we embarked on an extensive effort to build an IXS spectrometer as part of the Dynamic X-ray Scattering (DXS) Instrument. While this outlook is surely exciting, leveraging high average spectral flux for time-resolved IXS measurements appears to be an arduous task. The conventional spectrometer designs, honed at synchrotron facilities, can mostly be adopted. However, we realized that a renewed approach to monochromator designs is needed to preserve the FEL beam qualities, particularly the pulse duration.