

<b>Inelastic scattering II</b>	
Current designated sector:	Facility goes to:
ID28	ID28

### 1.1 ID CARD

ID28 investigates the vibrational properties of condensed matter. The submitted proposals cover mainly hard condensed matter (crystals and ordered systems) and disordered systems and liquids. Typical scientific questions relate to the role of phonons in superconducting materials and strongly correlated electron systems, and the determination of elastic and thermodynamic properties of minerals and advanced materials. Further themes address fundamental questions in glass forming systems and liquids, including surface and confinement effects.

### 1.2 SCIENTIFIC CASE

ID28 is fully dedicated to the study of the vibrational properties of condensed matter with meV energy resolution. Key characteristics of the beamline are (i) an energy resolution of 1.5, 3 and 6 meV at 21747, 17794, and 15816 eV, respectively, (ii) a momentum transfer,  $Q$ , range from 1 – 100  $\text{nm}^{-1}$ , (iii) a focal spot size on the sample of 30 x 60  $\mu\text{m}^2$  (horizontal x vertical, FWHM), and (iv) a nine-analyser spectrometer which allows for parallel data acquisition within the horizontal scattering plane, spanning a  $Q$ -range of 8 to 12  $\text{nm}^{-1}$ .

The proposed evolution contains the following elements:

- Improvement of the focusing optics with the target being a focal spot size of 15 x 5  $\mu\text{m}^2$ , thus enhancing the flux density on the sample by more than a factor 20.
- Equipment of the spectrometer with thirty analysers. The thus enlarged, in parallel covered,  $Q$ -range of 24 to 36  $\text{nm}^{-1}$  will reduce the data collection time for disordered and polycrystalline systems by a factor three, and will allow efficient dispersion mapping for single crystals.
- An end-station for (thermal) diffuse scattering (TDS), located in the proposed ID27/ID28 extension, at ~110 m with micro-focusing capabilities, and equipped with a state-of-the-art large area pixel detector as a unique complement to conventional inelastic X-ray scattering (IXS) phonon spectroscopy (Option I).

- A small angle IXS spectrometer with sub-meV resolution for the study of liquid and disordered materials (Option II, should be considered if the meV IXS programme on ID16 is phased out).

The outlined development not only signifies a substantial increase in the performance of the existing beamline, but offers a worldwide unique facility combining IXS and TDS in the study of lattice dynamics. Besides strengthening the current research programmes (Krisch & Sette, 2007; Bosak et al, 2009) new opportunities will emerge. Examples include:

- Strongly correlated electron systems such as superconductors and actinides: study of the interplay of structural, electronic and lattice degrees of freedom.
- High pressure science (diamond anvil cell techniques and laser heating): experimental determination of sound velocities, elastic moduli and thermodynamic properties in materials relevant for Earth and Planetary Sciences.
- Material science: Insight into the structure/hardness relation and determination of *intrinsic* elastic properties of textured polycrystalline materials.
- Surface science and nanotechnology: vibrational properties of quantum dots, thin organic and inorganic films, and surfaces, using grazing incidence techniques.
- Biological and radiation sensitive materials for which a more efficient spectrometer will minimise radiation damage.
- Stroboscopic- or single shot phonon spectroscopy with time resolution down to milliseconds for the study of phase transformations, non-equilibrium dynamics, and dynamics in extreme conditions of temperature and pressure, as well as high electric and magnetic fields.

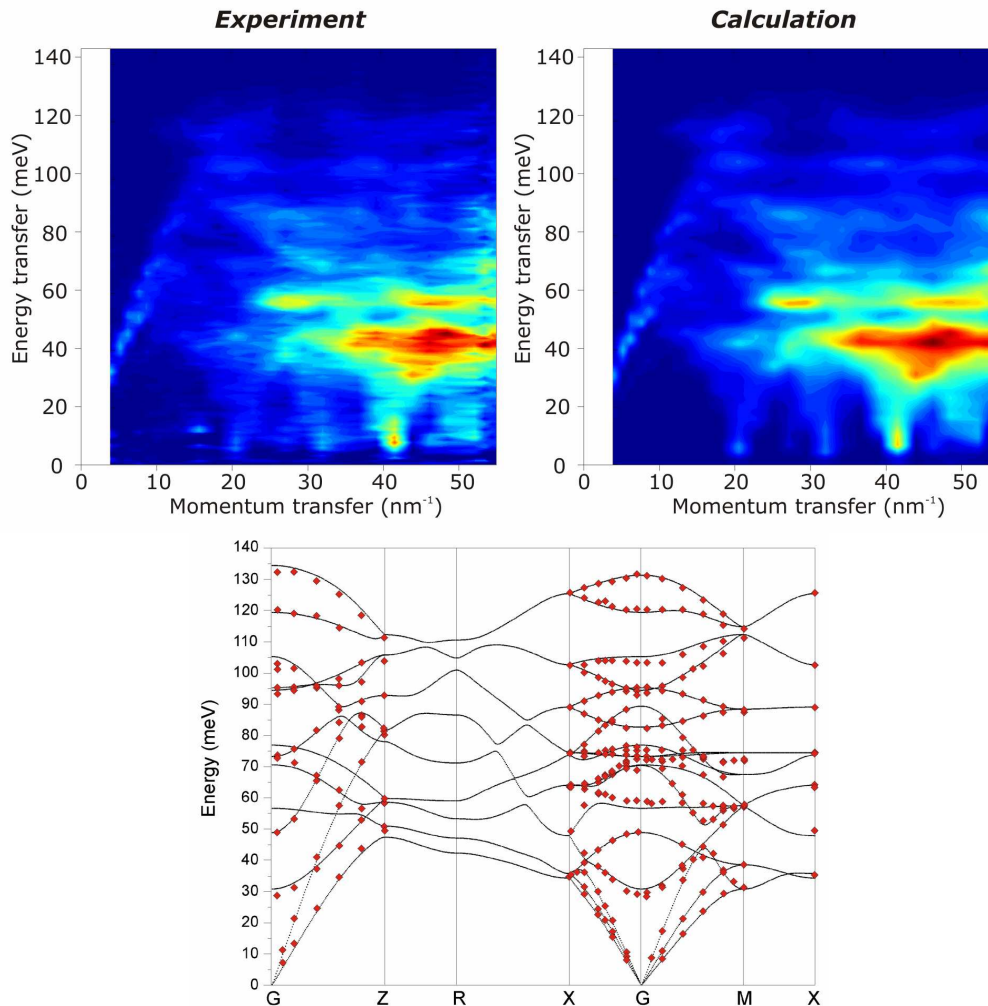
Several of the necessary methodologies have already been implemented, and give a flavour of the tremendous potential. Selected examples are presented below.

### **Determination of phonon dispersion in polycrystalline materials**

IXS studies on polycrystalline materials have until recently only provided orientation-averaged properties such as the average longitudinal sound velocity, the aggregate compressional modulus (from low momentum-transfer (Q) spectra), or the vibrational density-of-states (from high-Q spectra). On the other hand, IXS spectra collected over the whole accessible Q-range from typically  $2 - 70 \text{ nm}^{-1}$ , still display a marked Q-dependence due to - though relaxed - selection rules, and therefore contain information on the full lattice dynamics. In principle, a least squares refinement of model calculations versus the experimental IXS spectra, or a direct comparison with *ab initio* computations, allows access to the single crystal properties. Both methodologies have already been successfully applied in the case of beryllium, textured graphite (Fischer et al, 2009), and stishovite, a high-density polymorph of  $\text{SiO}_2$  (Bosak et al, 2009).

The left panel of Figure 1 shows the experimental E-Q-Intensity map for polycrystalline stishovite, derived from approximately sixty individual IXS spectra. The right panel displays the corresponding theoretical map obtained from *ab initio*

lattice dynamics calculations, using the CASTEP code. One notices the excellent match between the two maps, which is achieved simply by applying a single scaling factor of 1.05 to the energy scale of all IXS spectra. Applying the same scaling factor to the single crystal phonon dispersion gives almost perfect match with recently collected experimental single crystal data. It can therefore be concluded that the criterion of match between experimental and theoretical polycrystalline IXS spectra constitutes the discriminating test for the validity of the calculation, which in turn can be used to derive elastic and thermodynamic properties with a high level of confidence.



**Figure 1. caption: intensity map (w/o separation). Top panels: Experimental (left) and theoretical (right) intensity map of the polycrystal. Bottom panel: Experimental (red points) and theoretical (solid lines) phonon dispersions of the single-crystal. A single scaling parameter of 1.05 was utilised for the energy scale.**

### Combined inelastic X-ray and (thermal) diffuse scattering studies

Diffuse scattering contains contributions from static disorder (elastic scattering) and from phonons (inelastic scattering, commonly called thermal diffuse scattering (TDS)). These two contributions cannot be disentangled in a diffuse scattering experiment since the detectors used do not provide the required meV energy

resolution. The combination of diffuse scattering (DS) and inelastic X-ray scattering (IXS) offers the unique possibility of rapidly identifying the region of interest in reciprocal space, and, in a subsequent focused study, to disentangle static and dynamic (phonon) contributions (Hoesch et al, 2009).

Figure 2 shows the results of such a combined study on  $\text{Li}_x(\text{K}_{0.5}\text{Na}_{0.5})_{1-x}\text{NbO}_3$ , one of the most promising candidates for environmental friendly lead-free piezoelectric materials (Vakhrushev et al, 2009). These materials display a morphotropic phase boundary, i.e. a nearly vertical phase boundary in the composition-temperature phase diagram, but the origin of this “easy” phase boundary remains unclear. A 3D study of the diffuse scattering in the  $\text{Li}_{0.02}(\text{K}_{0.5}\text{Na}_{0.5})_{0.98}\text{NbO}_3$  as a function of temperature, following the cubic-tetragonal-orthorhombic phase sequence, revealed that in the close vicinity of the tetragonal to orthorhombic transition the crystal is effectively cubic again, and its symmetry can easily be altered by weak external action. Subsequent IXS measurements demonstrated that the phenomenon is dynamic in nature. This type of combined IXS-DS experiments largely benefits from both high-efficiency pixel detectors and a multi-analyser IXS spectrometer.

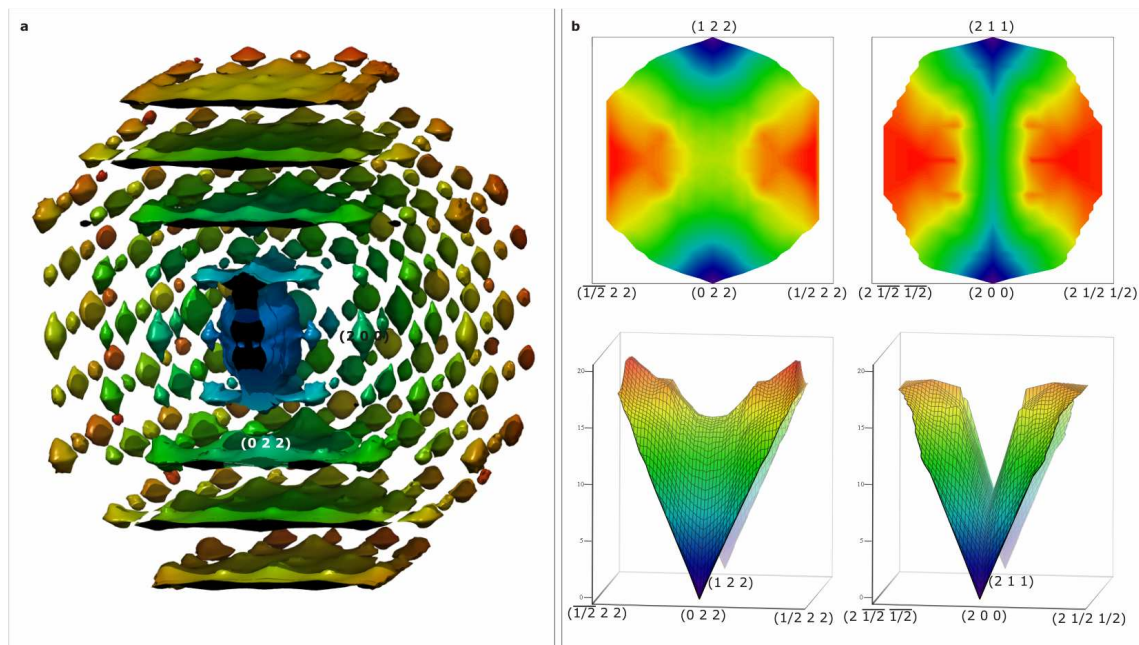


Figure 2. a) 3D constant intensity representation of thermal diffuse scattering in the orthorhombic phase of  $\text{Li}_{0.02}(\text{K}_{0.5}\text{Na}_{0.5})_{0.98}\text{NbO}_3$  (295 K), recorded with the Pilatus 6M detector on beamline X06SA/PXI at the SLS; b) Phonon dispersion sheets, reconstructed from IXS spectra, recorded with the 9-analyser spectrometer on ID28: Colour map and surface representations of the phonon energy evolution in  $(x\ 2+y\ 2+y)$  and  $(2+x\ y\ y)$  planes.

### 1.3 PROJECT HISTORY

The present CDR has evolved from the PHIXS CDR as published in the Purple Book. Most of the proposed new methodologies have already been implemented, or proof-of-principle experiments have been performed. This is particularly true for the thermal diffuse scattering studies.

#### 1.4 BASIC TECHNICAL CONSIDERATIONS

There are no principle technical challenges or risks. The proposed development can be implemented with existing technology and expertise.

X-ray source: With the installation of the third revolver undulator (32/17.6 mm) at the end of 2009, the optimum configuration within the limit of a 5 metre straight section will be obtained. As IXS is one of the most photon hungry techniques, it represents a strong case for a non-canted 7 m section. For the energy range 20 - 25 keV a length-optimised in-vacuum undulator provides a factor two increase in flux.

Detectors: Whilst for the proposed diffuse scattering station the most appropriate choice is the commercially available Pilatus detector, a customised, smaller area detector of the same type with high Z sensors would be the best choice for the IXS spectrometer.

#### 1.5 REFERENCES

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