BL35XU
Inelastic and Nuclear Resonant Scattering

1. Introduction

Since 2021A, BL35XU has been operated for research using two techniques: inelastic X-ray scattering (IXS) and nuclear resonant scattering (NRS). The techniques are both flux limited and strongly benefit from the short (20 mm)-period insertion device at BL35XU. Both methods also use a high-energy resolution of the meV scale or better. In FY2023, several tests and developments were conducted to improve performance and expand research interests, and these are described below.

2. Non-resonant High-energy-resolution Inelastic X-ray Scattering

The IXS station at BL35XU uses high-resolution IXS to investigate atomic motion–phonon dispersion in crystals and excitations in liquids. This station has a 3×4 analyzer array that enables the measurement of IXS spectra at 12 Q positions at one arm position. IXS is particularly notable as it allows access to small (micron-scale) samples, including thin films, which is not possible with other methods (e.g., inelastic neutron scattering). The IXS energy resolution is typically 1.5 or 3 meV.

The first goal of IXS measurements is to generate plots and data files of the scattered intensity as a function of energy transfer for each analyzer for each arm position where a measurement is made. This typically requires summing data from several scans, including the conversion from the temperature of the monochromator to energy, normalization, and the application of several calibrations/corrections that are determined at the start of the experiment. Then one must plot the spectra so the results are accessible for the 12-analyzer array. A series of procedures requires the editing of two text files for summing and plotting, which were created manually for each arm position, and the transfer of the (HKL) information from the diffraction code to the plotting code. Although such manual procedures of file editing allow great flexibility, it was complicated, especially for beginners, and took time.

Therefore, to standardize and simplify this procedure, a new Python 3-based wrapper software uses one “list” file that contains information for all the spectra. This is a good summary for users conducting their experiments. The Python code then creates the input files, transfers data around as needed, and runs the earlier processing software.

The Python-3-based SIXCIRCLE code [1] is now used at the IXS stations at SPring-8 in place of the FOURC program in SPEC. The SIXCIRCLE code numerically solves the diffraction conditions for the six-circle angles 2Θ, θ, χ, φ, μ, and γ and the surface angles α and β, with the option to hold a subset of them (or ω) constant. It also calculates the momentum transfers and momentum transfer resolution for all the analyzers in the array. The output has a high affinity for the wrapper software for data processing and plotting.

The above programs allow the data from an

Fig 1. Integrated data processing for IXS combining several steps into a smooth and automatically updating system. "list.csv" with one line of input per measurement combines several processes, as discussed in the text, and allows easy creation of graphics for investigating inelastic response.

entire experiment to be easily processed, including a fair number of sophisticated plotting options that have been found to be useful for IXS, including overlay with the "sarf" (deconvolved resolution function, see [2]). Examples are given in Fig. 1.

The stability of the incident beam position is crucial in time-consuming measurements such as IXS. However, it is known that the incident beam position drifts during energy scan by more than a few tens of microns (black dots in Fig. 2). This is because the direction of the backscattering crystal (BX) changes in the energy scan where the temperature of the BX is changed. The positional change was monitored as the intensity of the incident X-rays passing through a slit just before the sample. The beam position is tuned by adjusting the scanning angles of the BX. One set of these scans takes a few minutes and disturbs the energy scan. Moreover, this method is not sensitive enough to detect small (± 10 μm) deviations. This can be problematic for tiny high-pressure samples or thin films in a grazing-incidence geometry.

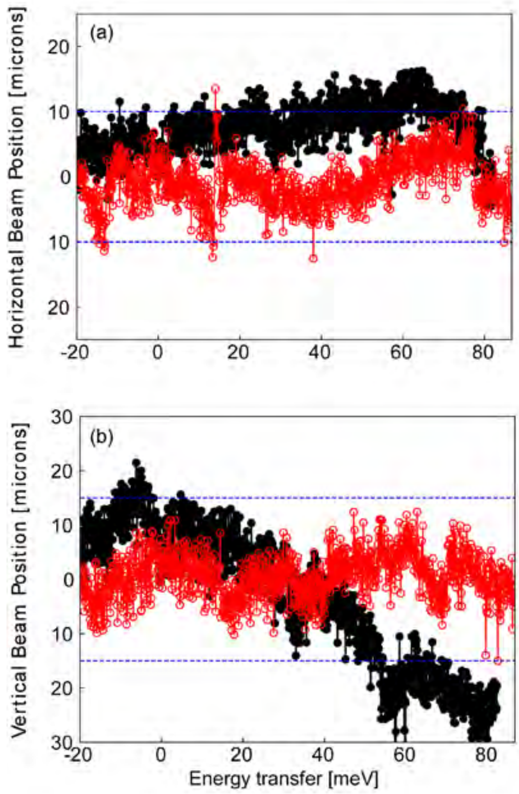

Fig. 2. Energy dependence of the incident beam position at XBPM in the (a) horizontal and (b) vertical directions. Red and black dots are data with and without feedback control, respectively. Blue horizontal lines indicate displacements where feedback was triggered.

The X-ray beam position monitor (XBPM) can detect a 10 μm displacement at the sample position in the usual focusing setup. By making a transfer matrix between the position on XBPM and the BX angle motors, the beam position can be corrected in both directions without disturbing the energy scan. In the case of Fig. 2, the beam position was corrected when the deviation reached 10 μm in the horizontal or 15 μm in the vertical direction (red points). This feedback is effective in maintaining the beam position within a period of more than 12 h.

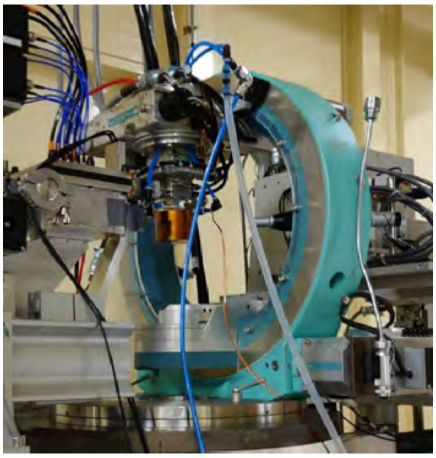

Fig. 3. Furnace for making IXS measurements of thin films at grazing incidence, mounted in the Eulerian Cradle.

There has been an ongoing project at BL35XU to investigate thin-film samples. This allows access to structured/layered materials, which are often interesting to control thermal transport, and also to interface-stabilized materials that are simply not available in bulk. However, investigating thin-film samples requires some specialized setups, as experiments must usually be in a grazing-incidence geometry to increase the X-ray path length in the film. This means there must be precise control over the sample position and grazing angle, as well as good calculations since the grazing angle is comparable to the angular deviations of the incident beam out of the horizontal spectrometer scattering plane caused by the focusing optics (the bent cylindrical mirror or the KB optics). As the footprint size on the sample is limited, precise control of the incident beam position is required. A wrong footprint position causes the reduction of the sample signal. To make matters worse, it sometimes causes an increase in the background signal, which makes it difficult to detect whether the beam is missing on the basis of the scattering intensity. The SIXCIRCLE code and XBPM also meet these requirements.

Together with the SIXCIRCLE code and XBPM the IXS station has a home-made furnace for a thin-film sample (Fig. 3), which can be mounted on the Eulerian cradle. This has been tested up to 550 K and used in several user experiments.

3. Nuclear Resonant Scattering

NRS provides a variety of different techniques to probe the dynamics at an atomic or molecular scale or local electronic states of specific nuclei, such as valence state, magnetism, symmetry, and relaxation, through hyperfine interactions. Energy resolutions are typically in the meV–neV range depending on the technique and the target nucleus. Resolutions in the μeV–neV range are attributed to the narrow bandwidth of nuclei, while the meV resolution comes from the resolution of the monochromator. Most NRS methods require timing-mode operation of the storage ring. However, a “Synchrotron Mössbauer Source” (SMS) setup [3] can be used independently of the storage ring filling pattern/timing mode owing to a pure nuclear reflection in a highly perfect crystal. Two setups with SMS have been developed at BL35XU this fiscal year: one is for Mössbauer spectroscopy and the other is for quasi-elastic scattering.

The construction of an SMS system to perform hyperfine spectroscopy at BL35XU is in progress. This will use a slightly different geometry than the usual one such as installed in BL11XU [4] and ESRF [5]. Bragg scattering [6] and, in particular, backscattering in a sapphire crystal will be employed to avoid having to move the small and relatively fragile borate crystal. A precise air bearing stage with a furnace for the sapphire crystal has been installed for moving the sapphire crystal with minimal deviation (small pitch and yaw). The furnace has achieved <1 mK rms temperature stability. The setup is shown in Fig. 4.

There has been an ongoing collaboration between Tohoku University, RIKEN, and PSD staff, among others, to create a setup for energy-domain quasi-elastic scattering with sub-μeV resolution. This is essentially a synchrotron version of an older technique called "Rayleigh scattering of Mössbauer radiation" [7] but leverages both the more intense beams of the SR source and the advantages of the

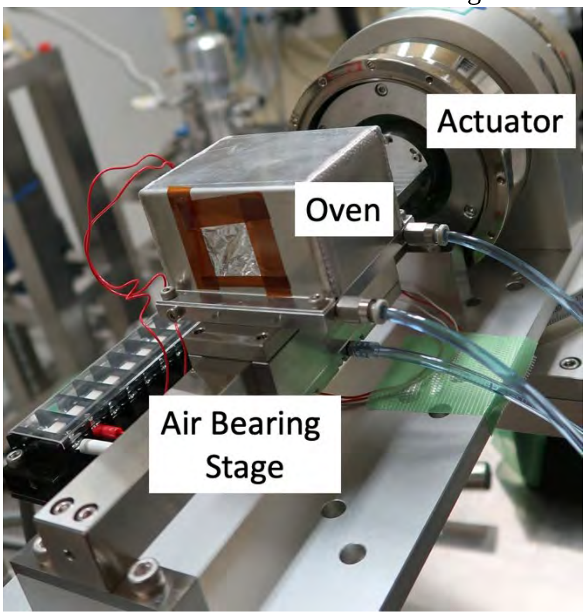

Fig. 4. Part of the SMS setup showing the oven or sapphire backscattering crystal on an air bearing stage with a "Mössbauer" velocity drive actuator. The tilt in the picture is purely an optical effect of the photograph and the motion is in the horizontal plane.

CITIUS area detector to allow, potentially, powerful measurements over a large range of momentum transfers and in any bunch mode. First measurements have recently been published, [8] and the setup should provide a much-needed window into investigations of glass-forming liquids and polymers.

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