BL35XU
Inelastic and Nuclear Resonant Scattering

1. Introduction

Since 2021A, BL35XU has been operated for the 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 FY2022, several tests and developments were conducted to improve performance, 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 i.e., phonon dispersion in crystals and excitations in liquids. IXS is particularly notable as it allows access to small (micron-scale) samples, including thin films, where other methods (e.g., inelastic neutron scattering) are not possible. The IXS energy resolution is typically 1.5 or 3 meV.

We continued to investigate the effect of X-ray irradiation on the first crystal of the high-heat-load monochromator (HHLM). In order to increase the heat load on the crystal, the ID gap was set to 6.974 mm, whereas that in FY2021 trials was 9.625 mm. The incident power was calculated using SPECTRA [1]. We measured the rocking curve width of the silicon (3 3 3) reflection at 76 keV of the first crystal 40 m downstream from the HHLM while changing the front-end slit size. The HHLM angle was 4.4 degrees in the present measurement, whereas it was 5.2 degrees in FY2021. A tantalum foil of 0.15 mm thickness was placed on the detector. The results are shown in Fig. 1 together with those in FY2021. A similar tendency was observed, that is, the rocking-curve width increases with the horizontal slit size at a constant vertical slit size. Note that the rocking-curve width was smaller at 4.4 degrees than that at 5.2 degrees with the same vertical slit size. With the largest vertical slit size, a maximum was observed at 0.6 kW incident on the HHLM.

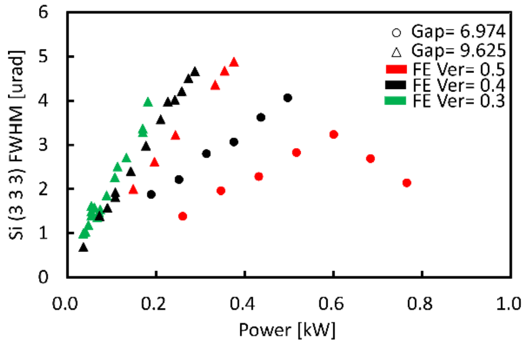

Fig. 1. Si (3 3 3) rocking curve width versus incident power on the HHLM. Circles and triangles correspond to 6.976 (present) and 9.625 mm (FY2021) ID gaps. Red, black, and green symbols indicate values with 0.5, 0.4, and 0.3 mm vertical slit sizes.

The backscattering monochromator is one of the key optics of the IXS station. With the monochromator currently used, an energy bandwidth narrow enough for IXS measurements is achieved only on a very limited region of the surface; the standard mechanochemical polishing

(MCP) used to fabricate the current crystal led to nonuniform and generally poor resolution. We then tried plasma etching (PE) to finish the surface of the backscattering crystal. The MCP of the new crystal already was better than the previous one. Then, the PE process further improved the energy bandwidth (Fig. 2). A depth of 2 microns was sufficient to remove the damaged layer on the surface. MCP followed by PE thus is clearly an improvement over only MCP, and more importantly, more reliable.

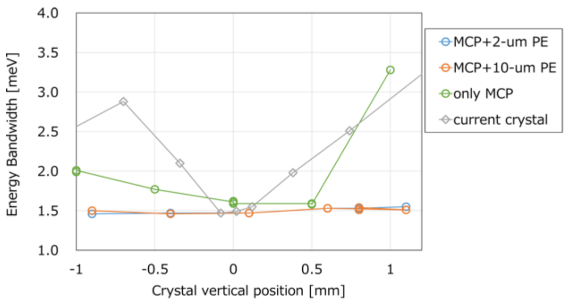

Fig. 2. Positional dependence of the energy bandwidth from the backscattering monochromator. Green: only MCP, blue: 2 μm PE on MCP, orange: 10 μm PE on MCP, gray: the current monochromator.

3. Nuclear resonant scattering

NRS uses a variety of different techniques to probe either atomic motion or the hyperfine environment of specific nuclei. Hyperfine spectroscopy gives users access to local information about fields (including magnetism), symmetry, relaxations, etc. Energy resolutions are typically in the meV–neV range where resolutions in the μeV–neV range are derived from the long lifetime of the excited states of nuclei.

We improved the electronics and data collection for synchrotron-radiation-based energy domain Mössbauer spectroscopy. In the original version of the technique [2], one collects the integrated delayed intensity as a function of the Doppler-shifted energy. However, we are now setting up a system that collects 2D histogram data providing binned data for both the velocity and the delay time of the photon. Users can then select an optimized or even multiple time regions for data processing to, for example, obtain the best S/N ratio or to otherwise match their interest. Eventually, we hope to be able to perform the full 2D (time and energy) analysis to understand the data.

In an earlier system, MCS6 purchased from FAST ComTec GmbH was used to obtain 2D spectra with a specially designed synchronization device. However, MCS6 has a limited counting rate, the response of the data acquisition software is slow, and there was no generally available data-processing software for obtaining the energy spectra.

We are now using a more modern MCS8 that is much faster and, in the last year, have started to develop advanced data-processing software. Figure 3(a) shows a 2D spectrum obtained using MCS8. The periodic peaks shown along the horizontal axis indicate the prompt signals. The delayed signals are clearly seen in the bunch intervals. MCS typically starts in time once per cycle of the ring, ~4.8 μs, to reduce the deadtime. After data are processed, users can obtain the 2D spectrum in terms of the decay process, as seen in Fig. 3(b). Users can eventually acquire the energy spectrum by integrating signals in any time window that they want, as shown in Fig. 3(c).

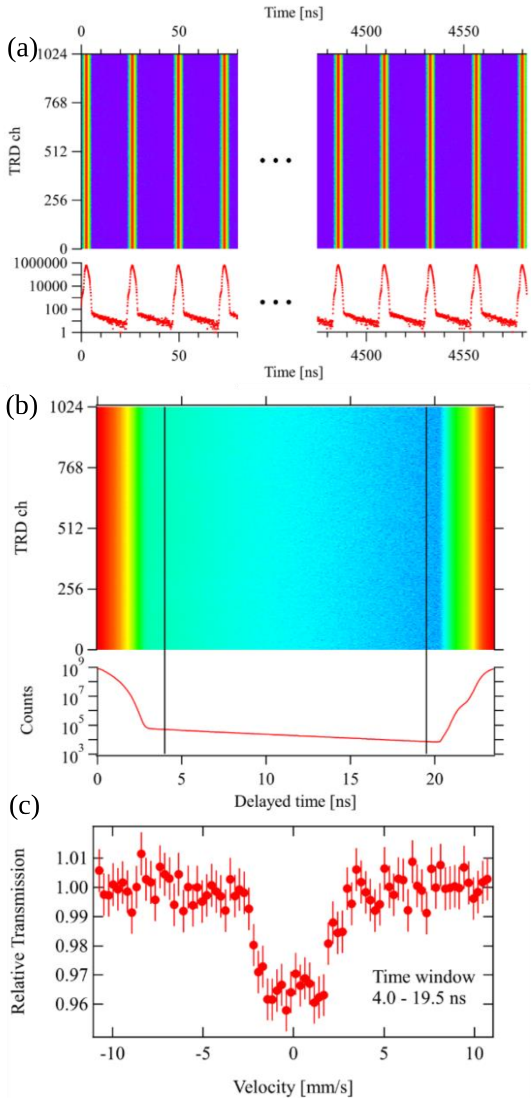

Fig. 3. Typical data in energy domain Mössbauer spectroscopy using MCS8, acquired over several hours in A-mode (bunch interval is 23.6 ns). (a) Data displayed in a 2D format. The horizontal and vertical axes show the relative time and velocity transducer (TRD) channel, respectively. The lower part shows the vertical integration over TRD channels. (b) Spectra as functions of the TRD channel and the delayed time calculated sums of the same delayed time along the horizontal axis in (a). Two vertical lines are guides for the eye for the time window to integration in the next step. (c) Spectrum integrating signals in the time window from 4.0 to 19.5 ns. The horizontal axis has been converted from the TRD channel into the Doppler velocity.

NRS methods use only a very small, <~meV, bandwidth of the X-ray beam. The remainder of the ~eV beam from the HHLM is then not used, and worse, it is generally a source of unwanted power that can adversely affect optics, leading to increased backgrounds, and can overload the NRS detectors.

We have optimized the experimental conditions to maximize the delayed signal rate in high-energy experiments. For high-energy NRS experiments, such as those with ^{174}Yb (76.5 keV) or ^{61}Ni (67.4 keV), in BL35XU, Si(333) reflection is used at the first optical element, that is, the HHLM. The HHLM should also lead to reflections of lower order, that is, (111). To suppress the X-rays with a higher order, a Si (HH0) channel-cut crystal is mounted downstream of the HHLM. Selecting a higher index H improves the energy resolution and decreases the number of photons, but may also reduce the number of desired resonant photons, as the angular acceptance of higher order reflections can be reduced. We have found the optimized front-end slit conditions for H = 4 and 8. Currently, BL35XU has two main options for high-energy optics. Since the condition with H = 8 has a resonant/total flux ratio of a factor of ~3 less than that with H = 4 even though this selection decreases the total flux to ~1/13, we recommend to use H = 8 when the samples have a sufficiently large cross section to cover the incident X-rays. Otherwise, H = 4 should be used when the flux at the detectors is sufficiently small so as not to reach the maximum count rate

(e.g., if users enclose the samples into a diamond-anvil cell.)

Fukui Hiroshi, Manjo Taishun, Nagasawa Nobumoto, Yoda Yoshitaka, and Baron Alfred
Precision Spectroscopy Division, JASRI

References:

[1] Tanaka, T. (2021). *J. Synchrotron Radiat.* **28**, 1267.
[2] Seto, M. *et al.* (2010). *J. Phys.: Conference Series* **217**, 012002.