The Ultrahigh Resolution IXS Beamline of NSLS-II: Recent Advances and Scientific Opportunities

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The Ultrahigh Resolution IXS Beamline of NSLS-II: Recent Advances and Scientific Opportunities

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Abstract. The ultrahigh resolution IXS beamline of NSLS-II is designed to probe a region of dynamic response that requires an ultrahigh energy and momentum resolution of up to 0.1 meV and < 0.1 nm^{-1}, respectively, which is currently still beyond the reach of existing low and high frequency inelastic scattering probes. Recent advances at NSLS-II in developing the required x-ray optics and instrumentation based on the use of extremely asymmetric Bragg back reflections of Si have allowed us to achieve sub-meV energy resolution with sharp tails and high efficiency at a medium energy of around 9.1 keV, thereby validating the optical design of the beamline for the baseline scope and paving the way for further development towards the ultimate goal of 0.1 meV. The IXS beamline is expected to provide a broad range of scientific opportunities, particularly in areas of liquid, disordered and bio-molecular systems.

1. Introduction

Momentum-resolved Inelastic X-ray Scattering (IXS) with meV energy resolution provides a powerful technique for studying vibrational dynamics and excitations in condensed matter systems [1]. The scientific objective of the IXS beamline of NSLS-II is focused on very high-resolution (1 meV ~ 0.1 meV) IXS experiments. The ultimate goal is to achieve an energy resolution of 0.1 meV, which represents an order of magnitude improvement over the best currently operating instruments in the world. With the 0.1 meV energy resolution, we anticipate improved understanding of the low-energy excitations in liquid, disordered and bio-molecular systems, where atomic motions due to the naturally occurring inhomogeneity and/or density fluctuations dominate the scattering at low momentum transfer (Q), for which very high Q resolution (< 0.1 nm^{-1}) would also be required. Achieving these goals would allow the NSLS-II IXS beamline to bridge, at least partially, the dynamic gap (0.1 nm^{-1} < Q < 2 nm^{-1} and 0.1 meV < E < 5 meV) between various existing inelastic scattering probes [1]. This is an area where potential new science may be expected. Study of relaxation dynamics, sound propagation and transport properties, phonons in single crystals, surfaces, thin films, confined liquids, systems under extreme pressure, as well as phonons in excited states (pump and probe experiments),

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and exotic excitations in strongly correlated materials, for which an energy resolution of 1 meV would be sufficient for most cases, is also expected to make up a large bulk of the scientific research at this beamline.

2. Beamline and Spectrometer Design

To meet these objectives, the optical design of the beamline and the spectrometer employs the so-called CDW or CDDW monochromator designs (that comprise a Collimator, one or two Dispersing crystals, and a Wavelength selector) based on the angular dispersion effect in asymmetric Bragg reflection [2]. The dispersion effect makes it possible to achieve sub-meV resolution at a moderate energy of ~10 keV. These new monochromator designs differ fundamentally from the symmetric Bragg back reflection optics – the spherical backscattering crystal analyzers – employed in all existing IXS instruments, which must work with high index Bragg reflections at higher energies (> 20 keV) in order to achieve meV resolution. The D crystal of these monochromators is implemented at the Si(800) back reflection at a medium energy of 9.13 keV, where the NSLS-II undulator sources have superior performance in both brightness and spectral flux [3]. To realize the analyzer optics of the spectrometer, the optical design takes advantage of the unusually large angular acceptance of the CDW/CDDW monochromators by combining a Montel multilayer collimating mirror with an angular acceptance of 5-10 mrad in both the horizontal and vertical scattering directions, making it comparable to the angular acceptance of the spherical crystal analyzers currently in use in existing IXS spectrometers [4]. Latest independent prototype test results (see Ref [5,6] and Section 3) have verified the working principle of the CDW/CDDW monochromators and demonstrated sub-meV energy resolution with much sharper tails compared to those of the symmetric Bragg back reflection optics. This is a major advantage of the new scheme compared to the spherical backscattering crystal analyzers, which will help to eliminate spurious background and to improve contrast.

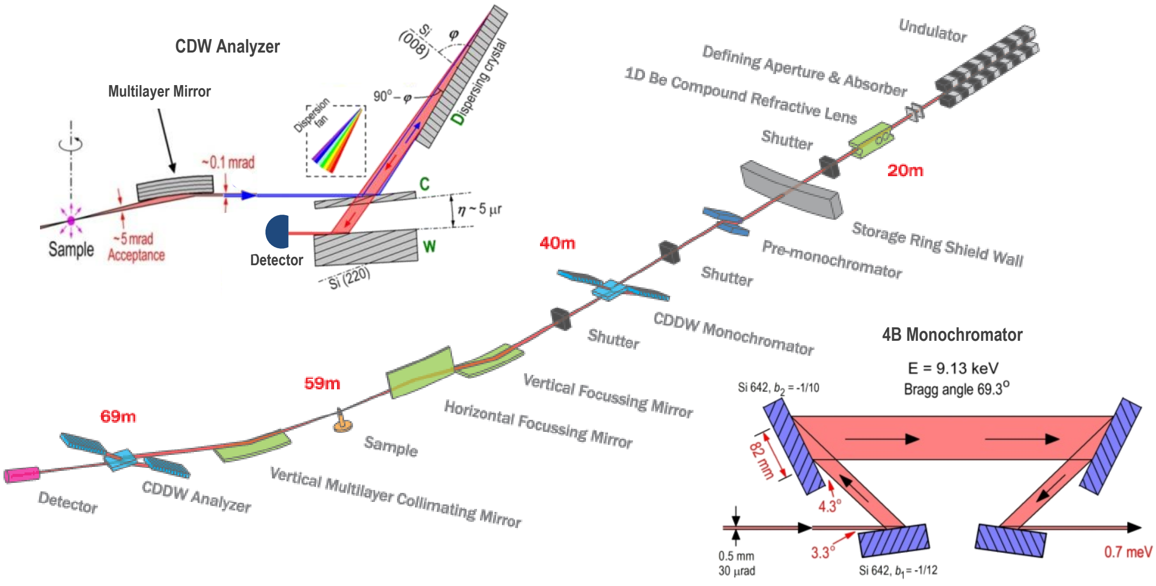

Figure 1. A schematic view of the optical layout of the IXS beamline and spectrometer. The insets are the 4-bounce monochromator (lower right) and the CDW analyzer (upper left) chosen for operation at 1 meV resolution.

Figure 1 illustrates schematically the optical layout of the IXS beamline and spectrometer. The insertion device is a room-temperature 22-mm period in-vacuum undulator with a total length of 6 m installed in a high-β straight section and delivers 1.6×10^{15} photons/sec/0.1%bw at 9.13 keV [3]. Since asymmetric dispersive crystal optics introduces severe source size broadening proportional to the distance of the optics from an achromatic source [7], a set of 1D Be compound refractive lenses (Be-CRLs) is used to pre-focus the beam vertically at a secondary source aperture in front of the high-resolution monochromator to minimize the source size broadening. The focusing of the Be-CRLs is close to 1:1 and maintains the natural divergence of the undulator beam to ensure full acceptance by the high heatload pre-monochromator. After the high-resolution monochromator, the beam is focused onto the sample by a set of KB (Kirkpatrick-Baez) mirrors. Shadow ray-tracing studies of the entire optical scheme, including slope errors of the mirrors, heatload effects on the pre-monochromator and dispersion broadening by the high-resolution monochromator, indicate that a focus of ≤ 5 µm (V) × 7 µm (H) can be obtained on the sample.

The current optical design of the beamline has been optimized to maximize the flux delivered to the sample by ensuring full acceptance of the beam through all optical components, limited only by the spectral efficiency of the high-resolution monochromator. The CDDW monochromator offers a spectral efficiency of 22% with an energy width of 0.4 meV by design [5]. We have also studied a 4-bounce (4B) monochromator (Figure 1) designed for 0.7 meV resolution with an angular acceptance of 30 µrad, which improves the spectral efficiency to ~35%. The 4B design provides a large energy scan range by scanning the Bragg angle of the crystals, but its energy sensitivity to angular change (3.45 meV per 1 µrad) is substantially higher than that of the CDDW (0.25 meV per 1 µrad) and therefore demands mechanical stages with greater angular resolution and stability. Development effort at NSLS-II in this area has been very successful [8]. For the CDDW, energy scan beyond the dispersion width (the intrinsic width of the (800) reflection) would require simultaneously scanning the temperature of the two D crystals. This 4B monochromator has been chosen for operation at 1 meV resolution and is expected to deliver up to 3.7×10^{10} photons/sec at the sample.

The analyzer optics will be implemented by combining a laterally graded Montel multilayer mirror with the CDW optics to achieve a 5-10 mrad angular acceptance. At an optimized distance of 200 mm from the sample, the collected scattered x-rays will be collimated to less than 100 µrad in both the vertical and horizontal direction, provided that the beam size is less than 25×25 µm^2 [4]. The true acceptance of the analyzer optics is however limited by the finite length of the D crystal, which defines the vertical size of the beam accepted from the Montel mirror. At an asymmetry angle of 88.5° (for 0.7 meV), for instance, a 1-m long D crystal accepts only ~1.2 mm beam or x-rays scattered from the sample within a 5 mrad angular spread. This presents the greatest challenge of the new analyzer scheme due to the requirements on the perfection of the D crystal in terms of the lattice ($\Delta d/d < 10^{-8}$) and temperature ($\Delta T < 4$ mK) homogeneity, angular stability (< 2 µrad) and surface quality (roughness < 2 nm; slope error < 10 µrad) to achieve the 0.1 meV resolution. Mounting and controlling such a long crystal without introducing strain and deformation, and at the same time achieving the required angular precision and stability also present great challenges. To mitigate these challenges, the D crystal is divided into shorter 200-mm segments, each independently controlled (see Figure 2). This reduces greatly the technical difficulties of the scheme, making it feasible for 1-meV resolution operation as demonstrated by our prototype test results (see Ref. [6] and Section 3).

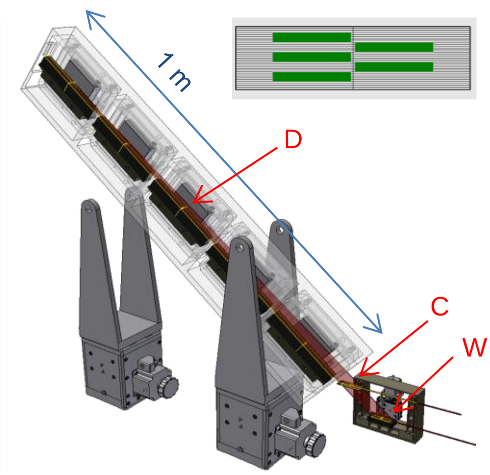

Figure 2. The CDW analyzer assembly comprising five 200 mm D crystals. Inset shows the beam images on a pseudo-2D strip detector.

A custom designed pseudo-2D strip detector will be used to separate x-rays reflected from each segment (see Figure 2). Variation in the photon energy due to differences in the lattice, temperature and angular position of the segments can be captured individually and corrected, further lessening the uniformity requirements as discussed above across the entire length of the D crystal. Furthermore, a small azimuthal angle de-tuning will be introduced in each D segment in order to remove the reflectivity loss caused by the multiple beam diffraction (MBD) in the CDW scheme by using the 45° rotated azimuth orientation of the D crystal as discussed in detail in Ref. [9], thereby improving the efficiency by up to ~30%. The required de-tuning is within a few mrad, which is enough, when applied in opposite directions for adjacent D segments, to separate their signals to prevent cross talk, as illustrated in Figure 2.

3. Status of High-Resolution Crystal Optics

To verify the performance of the CDW and 4B monochromators, we have carried out prototype test experiments at the Taiwan Beamline (BL12XU) at SPring-8, and at the P01 Beamline at PETRA III. The test system implemented includes configurations for testing the CDW individually or using one CDW as the analyzer to test another CDW (Figure 3a) or the 4B as the monochromator (Figure 3b). The configuration includes also testing of the Montel multilayer mirror both individually and as part of the entire optical scheme of the IXS beamline chosen for operation at the 1 meV resolution.

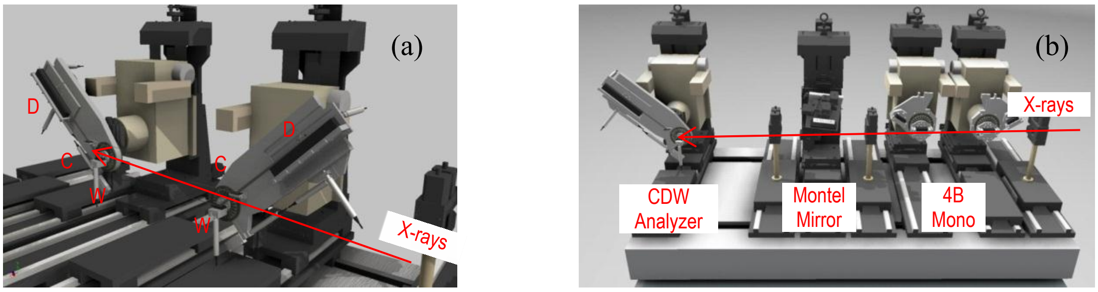

Figure 3. The prototype test system. (a) CDW-CDW configuration; (b) 4B-CDW configuration with the Montel multilayer mirror.

Figure 4 shows a selection of intensity profiles measured in the tests. Figure 4(a) is a D crystal rocking curve of the CDW monochromator measured with the incident beam from the beamline. Two reflections are visible: the sharp peak of 4.6 μrad width is the symmetric (800) reflection from the front edge of the D crystal; the broad peak of ~105 μrad width is the asymmetric (800) reflection from the diffraction surface of the D crystal showing the dispersion effect. Both of the width and the separation of the two peaks agree well with the theory for an asymmetry angle φ of 88.5° for the D crystal [2]. Combining two CDW units using one as the analyzer, the rocking curve width of the analyzer D crystal against the monochromatized beam from the first CDW provides pertinent information on the resolution of the CDW monochromator. An example is shown in Figure 4(b) with a rocking curve width of 3.65 μrad. The corresponding energy width can be estimated to be 0.87 meV based on the angular dispersion relation: $\Delta E/E = \Delta\theta/\tan(\varphi)$. Assuming the two units are identical, the energy resolution of one CDW is estimated to be at 0.7 meV [6]. The resolution of the 4B monochromator can be measured similarly using one of the CDW as analyzer. This is shown in Figure 4(c) with an angular width of 0.24 μrad. The corresponding energy width of 0.83 meV is given by the Bragg dispersion: $\Delta E/E = \Delta\theta/\tan(\theta_B)$. In both the CDW and 4B scans, the rocking curves are sharper than a Lorentzian of the same width as is shown in Figure 4(b) and 4(c), but the tails are still longer than the ones given by simulations based on dynamical diffraction theory, indicating room for improvement. The tails are likely to be caused by diffused scattering from the finite surface roughness of the dispersion crystals. Detailed analysis of the data including the dynamical diffraction theory simulations will be published elsewhere.

Another key performance factor important for the beamline operation is the energy scanning and repeatability of the 4B monochromator. This was tested in combination with the CDW analyzer by stepping the 4B crystal Bragg angle while scanning the D crystal to match it. This resulted in a series of evenly separated rocking curves with their intensity following closely the dispersion peak profile of the D crystal (Figure 5). Repeatability tests of the 4B energy scan with the hutch temperature stabilized to ±15 mK over 3 hours period yielded an energy shift of ~0.2 meV, which is quite acceptable for operation at 1 meV resolution. Further improvement is expected with a better control of the temperature environment and the scanning mechanism.

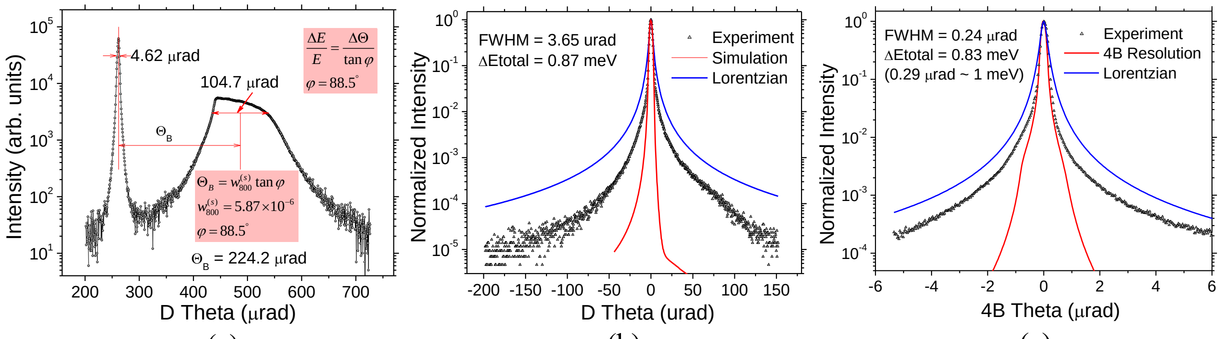

Figure 4. D crystal rocking curves of (a) the CDW monochromator and (b) the CDW analyzer. (c) The angular scan profile of the 4B monochromator measured through the CDW analyzer.

4. Expected Performance

The spectral efficiency of the high-resolution optics plays an important role in determining the overall performance of the IXS beamline. This was studied also in the tests. The spectral efficiency or transmissivity per energy width of the CDW was determined to be ~20% compared to a theoretical value of 38%. The loss was mostly induced by the reduced anomalous transmission of the thin C crystal, which is known to be sensitive to strain. The spectral efficiency of the 4B was found to be ~30% compared to the theoretical value of 35%. With this experimental result and based on Shadow ray-tracing, the performance of the IXS beamline at 1 meV resolution using the 4B monochromator is expected to be among the best in the world in terms of the energy resolution and incident flux (see Table 1). The flux density in particular will be more than one order of magnitude higher than those of other existing facilities, making it ideal for studies of small sample systems.

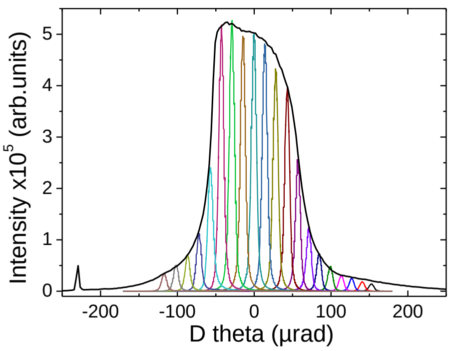

Figure 5. 4B-CDW scans with varying 4B crystal Bragg angle. 4B energy increases from left to right.

Table 1. Comparison of key performance parameters with existing IXS facilities in the world using spherical backscattering analyzers and the expected performance of the NSLS-II IXS beamline.

Facility (Beamline)	ΔE (meV)	ΔQ (nm^{-1})	E (keV)	Intensity (phs/sec)	Beam Size (V×H μm^2)	Flux Density (phs/sec/μm^2)	Sharp tails
ESRF (ID28)	1.6	0.3	21.7	6×10^9	7×12	7.1×10^7	--
APS (30ID)	1.5	0.6	23.8	2×10^9	15×35	3.8×10^6	--
SPring-8 (43LXU)	1.5	0.3	21.7	$\sim5\times10^{10}$	20×35	7.1×10^7	--
NSLS-II (IXS)	1.0	0.2	9.1	$\sim3.2\times10^{10}$	5×7	$\sim1\times10^9$	Yes

The efficiency of the analyzer depends on the spectral efficiency of the CDW optics and the Montel mirror and is limited by the angular acceptance of the entire optics. From our experimental results [4,6], the combined spectral efficiency would be ~10%, although further improvement is possible. This is comparable to that of the spherical backscattering analyzer operating at similar resolutions at energies greater than 21 keV. The detection efficiency can be improved by having multiple analyzers as implemented in all existing IXS facilities, which multiply the collection solid angle by the number of analyzers used. A similar approach will be implemented for the spectrometer for NSLS-II.

A major difference between the NSLS-II IXS beamline and other existing IXS facilities is the lower incident energy used. For the same solid angle of the analyzer, the lower energy yields a better Q resolution. Estimates of the count rate on typical sample systems studied by IXS indicate that the NSLS-II IXS spectrometer performs better in general for lower-Z materials. In particular, for sample systems where the sample volume is constrained by their sample environments such as the high-pressure diamond anvil cells, the available sample volume matches better to the smaller absorption length. This suggests that the scientific strength of the beamline will be on low-Z materials and systems under extreme high pressure conditions.

5. Discussion and Conclusion

The ultimate success of the NSLS-II IXS beamline depends on the success of the 0.1 meV crystal optics development effort. Based upon a recent study of the CDDW monochromator in which an energy resolution of 0.46 meV with 16% spectral efficiency was reported [5], achieving the 0.1 meV resolution using the CDDW design for the beamline monochromator appears to be a realistic goal. One could also explore other multi-bounce monochromator designs for which energy widths close to 0.1 meV have been shown [10,11]. However, a common drawback of these high-resolution monochromators is that they all use extremely asymmetric crystals and thus their length becomes prohibitively long as the beam size increases. Their use as analyzers as suggested in [12] would make it prohibitive to achieve the required performance. In fact, the large angular acceptance needed for analyzers would imply an mm sized beam, thus the length of the crystals required would be in meters. This appears to be the greatest limitation of this new scheme for practical implementation at the 0.1 meV resolution level. For 1 meV resolution, our approach appears feasible based on all the test results obtained so far. This novel design, soon to be implemented in a user instrument, provides a unique opportunity for NSLS-II to build a fundamentally new IXS spectrometer with unprecedented performance.

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