Inelastic X-ray Scattering

Inelastic X-ray Scattering (IXS) is a powerful experimental technique that probes directly the dynamical behavior of a variety of condensed matter systems, and has been successfully applied to a wide range of problems, including phonon dispersion in solids and liquids, dynamics of disordered and biological systems, as well as electronic excitations in metals, semiconductors and insulators. IXS offers several unique strengths compared to other inelastic (such as neutron and electron) scattering techniques. For examples, the energy and momentum transfers ($\triangle E$, $\mathbf{q}$) are independently variable and cover the full range of dielectric response, with $\triangle E$ ranging from meV to keV and $\mathbf{q}$ spanning over typical Brillouin zone sizes. Furthermore, because X-rays couple directly with the electronic charge, the technique is particularly sensitive to all kinds of electronic excitations. The high energy of the hard X-rays also means that IXS is a truly bulk-sensitive probe and can be applied to systems under extreme environments of pressure, temperature, and external electric and magnetic fields. The energy tunability of synchrotron radiation sources allows element specific experiments. The small beam size offers furthermore the possibility to study small samples.

As part of the Taiwan X-ray facility at SPring-8, BL12XU is dedicated to the development and application of the IXS technique for the study of electronic excitations in correlated electron systems with variable energy resolution from 10-1000 meV. Major experimental capabilities have now been developed for performing both *non-resonant* and *resonant* IXS experiments. In the present article, we discuss some of the technical and experimental aspects of the IXS technique with illustrations using results from recent commissioning experiments performed on the beamline.

Beamline and IXS Spectrometer

Conceptually IXS is a simple experiment. As depicted in Fig.1, well-collimated and monochromatic X-ray photons with energy and momentum (E_1, $\mathbf{q_1}$) are used to bombard the sample. The scattered photons with energy and momentum (E_2, $\mathbf{q_2}$) are then detected over a solid angle $d\Omega$. The number of scattered photons within a given energy and momentum transfer space provides information on the intrinsic properties of the system. Due to the low inelastic scattering cross section, only a very small portion of the incident photons are scattered. IXS therefore requires a highly intense and bright incident X-ray beam and efficient ways to collect and perform the energy analysis of the scattered photons.

BL12XU takes full advantage of the highly brilliant hard X-rays generated from a SPring-8 standard 4.5-m long in-vacuum undulator. The optical system (see Fig. 2) consists of five elements, which include a high heat-load Si(111) double-crystal monochromator (DCM), a collimating mirror (CM), a high-resolution monochromator (HRM), a phase retarding plate (PRP) to generate circularly polarized light, and a focusing mirror (FM). The DCM requires no pre-collimation as the natural divergence of the SPring-8 standard undulator beam is smaller than the intrinsic angular acceptance of the Si(111) reflection over the energy range of the beamline: 5-35 keV. The collimating mirror collimates the beam further to increase the throughput of the HRM operating at higher-order reflections of Si that generally have small angular acceptance. It also functions as a higher-order light filter by having two stripes: Si and Pt for the energy range of 5-12 keV and 12-30 keV, respectively. To achieve variable energy resolutions of 10-1000 meV, the HRM can be configured using two high-precision co-axial goniometers to various combinations of 2-bounce or 4-bounce (inline or nested) channel-cut crystals. An in-line combination of two symmetric Si(333) channel-cut crystals, for example, gives an energy resolution of 50 meV at 9.886 keV (see Table 1). Additional HRM crystals can be designed for dif-

ferent energy resolutions. After the HRM and the PRP (to be implemented later), the beam is delivered to the focusing mirror and focused both horizontally and vertically to 120(H)×75(V) μm^2 at the sample position. Total flux obtained at the sample is 1.2×10^{13} phs/sec/1.4 eV at 9.886 keV scaled to 100-mA ring current with the DCM direct beam, which is reduced proportionally with the HRM bandwidth.

The IXS spectrometer (Fig. 3) was designed and built to accommodate a wide range of experimental requirements. The heavy-duty goniometer tower has a load capacity for a cryomagnet. A cryostat with custom-built carrier provides a sample environment down to 4 K. The 3-m analyzer arm can accommodate spherical crystal analyzers with bending radii of 1-3 m, for the detection and energy analysis of the scattered X-rays. Such analyzers are essentially X-ray diffraction optics made from spherically bent perfect crystal wafers, such as silicon. Bending the wafer into a sphere produces semi-focusing of the analyzed X-rays into the detector, collecting more photons at the expense of momentum resolution. However, the strain induced by the bending broadens the energy resolution and reduces the intrinsic reflectivity of the analyzer. To improve the energy resolution and reflectivity, the wafer is usually diced into individual blocks before being glued onto the spherical concave substrate. Various methods have been developed at the NSRRC for the fabrication of such crystal analyzers. The resultant resolution and efficiency of the analyzer depend on a number of factors, including the bending radius, the block size, the groove width and depth, and the order of the Bragg reflection used, etc. Table 1 lists some of the analyzers with measured energy resolutions ranging from 50-650 meV at the Si(555) near-backscattering energy of 9.886 keV, giving several optimal configurations for the operation of the beamline and the IXS spectrometer.

Experimental Capabilities

(1) Non-Resonant Inelastic X-ray Scattering (NRIXS)

NRIXS is the first major experimental capability implemented on BL12XU. Here the incident beam energy (i.e., E_1 in Fig. 1) is scanned near the backscattering energy 9.886 keV of a Si(555) crystal analyzer. The beamline configuration uses a pair of Si(333) channel-cut crystals as the HRM, delivering a beam of just 50 meV width at 9.886 keV. The flux of the delivered beam at the sample reaches 1.5×10^{11} photons/sec at 100 mA. Together with a highly efficient, diamond-saw diced 2-m radius Si(555) crystal analyzer, we have obtained a total energy resolution of 70 meV (Table 1).

NRIXS can be used to study various types of

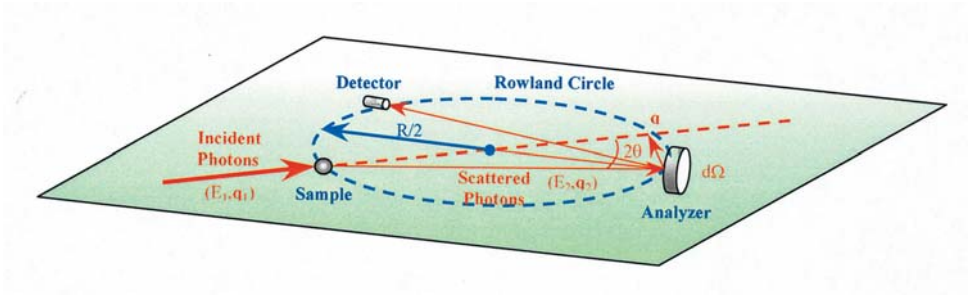

*Fig. 1: Schematic of an inelastic X-ray scattering experiment. The sample and the detector are placed on the Rowland circle of the bent crystal analyzer (bending radius R). For non-resonant IXS experiments, the energy transfer $\triangle E$ is given by $E_1 - E_2$ with E_2 fixed at the near-backscattering energy of the Bragg reflection of the analyzer crystal. For resonant IXS experiments, E_1 is tuned to the absorption edge of a 3d or 4f element, and $\triangle E$ is given by $E_2 - E_1$. The momentum transfer **q** in both cases is simply defined by the scattering angle (2θ) with a magnitude given by $q = |\mathbf{q}_1 - \mathbf{q}_2| = 4\pi Sin(2\theta/2)/\lambda$ and a resolution determined by the solid angle ($d\Omega$) of the analyzer. The energy resolution of an experiment depends on a number of factors and is dominated by the energy width ($\triangle E_1$) of the incident beam and the energy resolution ($\triangle E_2$) of the analyzer.*

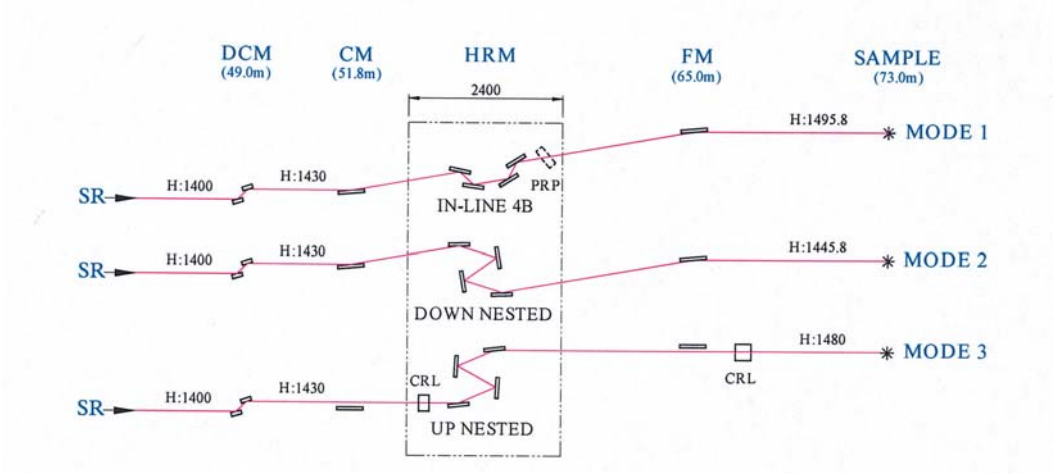

Fig. 2. Beamline optics configurations of BL12XU.

electronic excitations, including valence electron excitations, plasmon and exciton dispersion and other single-particle and collective excitations. It can be used also to study core-level excitations, as an alternative to absorption spectroscopy. Since the incident and scattered photons are high-energy X-rays, sample selections and environments are less restricted. The high energy resolution also affords studies on finer features which were not possible before, as demonstrated by our successful measurement of the dynamical structure of electrons in single crystal MgB_2.

Figure 4 shows the data taken from MgB_2 at room temperature with a total energy resolution of 65 meV and a momentum resolution of 0.06 A^{-1}. Figure 4(b) gives an overview of the energy loss spectrum at q = 0.58 A^{-1} showing the conventional plasmon feature at ~ 20eV and a smaller feature at ~ 4eV that displays substantial dispersion over the energy range of 2.5-4.5 eV (see Fig. 5 in *Report from Taiwan Beamline Office at SPring-8*). This feature may be identified with the theoretically predicted sharp coherent charge excitation for **q** along the c-axis [W. Ku et al., *Phys. Rev. Lett.* **88**, 057001 (2002)]. Previous experimental effort in confirming this collective mode has been hampered by the lack of large single crystals, which was now circumvented by the small focused and intense beam at BL12XU. The high energy resolution and sufficiently wide energy scanning range of the high-resolution NRIXS setup have also played a decisive role in obtaining the experimental data shown in Fig.4. Comparison with further calculations may shed new light on the nature of

Table 1. Beamline and IXS spectrometer configurations and performance for NRIXS experiments at the close-to-backscattering energy (9.886 keV) of Si(555) reflection. The flux for various HRM configurations is estimated relative to the Si(333) case. The energy resolution of the different analyzers are total energy resolutions measured using the 50-meV incident beam from the Si(333) HRM. Efficient combinations are those with the incident bandwidth closely matched to the energy resolution of the analyzer, such as the first combination highlighted here.

Beamline			IXS Spectrometer		
HRM Configuration	Flux ($\times 10^{11}$photons/sec)	Bandwidth (meV)	Si(555) Analyzer	Relative Efficiency (/meV)	Resolution (meV)
Si(333)	1.5	50	2-m Diamond-saw Diced	25%	70
Si(440)	2.3	80	2-m DRIE Diced		165
Si(400)	5.7	200	2-m Continuously Bent		230
Si(220)	13.5	480	1-m Continuously Bent		680

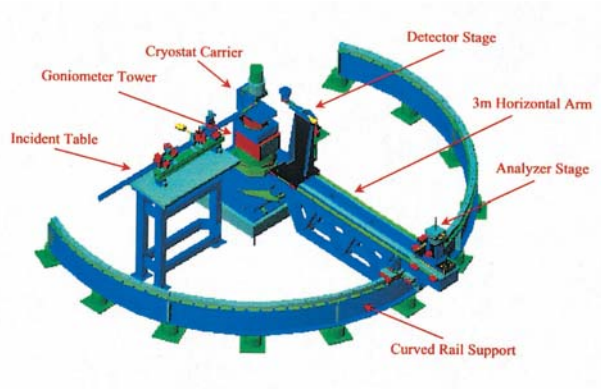

Fig. 3: Overview of the inelastic X-ray scattering spectrometer at BL12XU, custom designed and built in collaboration with the Advanced Design Consulting, Inc. NY, USA.

the dynamical response of electrons in this fascinating material. The results will be published elsewhere.

(2) Resonant Inelastic X-ray Scattering (RIXS)

RIXS is the second major experimental capability implemented on BL12XU. The initial setup uses 1-m radius spherically bent Ge(444) and Si(444) analyzers and the DCM direct beam, providing coverage of the *K* absorption edges of the late transition metals (Co, Ni and Cu) with a total energy resolution of ~1 eV and an incident flux of up to 1×10^{13} photons/sec at 100 mA. The RIXS energy (i.e., E_2 in Fig.1) can be scanned over 7.64-8.77 keV using the Ge(444) or 7.96-9.13 keV using the Si(444) analyzer by scanning the Bragg angle of the analyzer crystal and the position of the detector simultaneously while maintaining the Rowland circle geometry. The major difference of a RIXS experiment from NRIXS is that the incident photon energy is tuned to the absorption edge of the core electrons, which leads to a large enhancement of the inelastic scattering cross sections as previously demonstrated on NiO [C.C. Kao et al., *Phys. Rev. B* **54,** 16361 (1996)]. The RIXS data obtained from BL12XU on NiO are shown in Fig.5, which confirm essentially the earlier report, and demonstrate the readiness of the RIXS setup. The availability of resonant experiments at BL12XU opens up the possibility of studying secondary processes involving intermediate states as well as complex materials containing high-Z elements where sample absorption still poses a severe problem.

(3) Other Developments

In both types of experiments, the spherical crystal analyzer is an indispensable part of the IXS spectrometer. Several crystal analyzers with 1 or 2-m bending radii have been developed and tested, which lead to several possibilities in operating the beamline and the IXS spectrometer with different energy resolutions (see Table 1). The optimal energy resolution has to be balanced between the

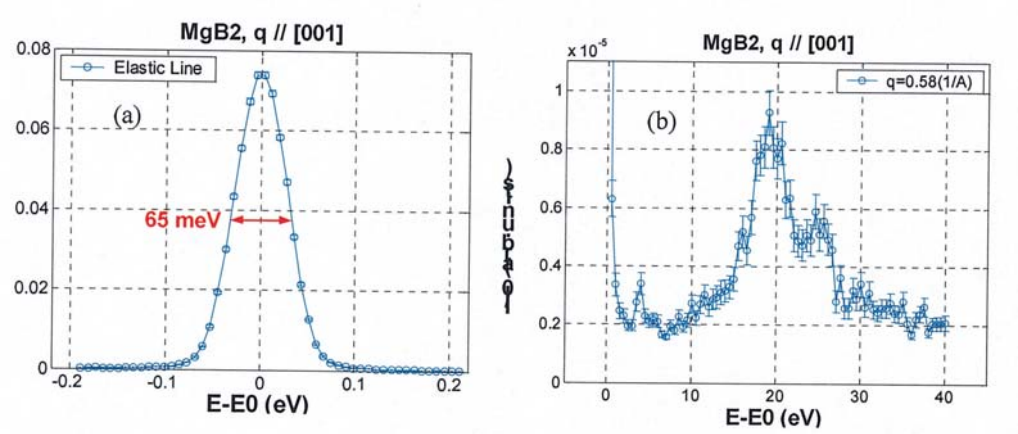

Fig. 4: NRIXS experimental data obtained from a free-standing single crystal of MgB_2 along the [001] direction perpendicular to the Mg and B planes at room temperature. The crystal measures 300 × 500 × (~20) μm^3 in size. The quasi-elastic line in (a) gives a FWHM of 65meV (i.e., the total energy resolution) for the experiment. The energy loss spectrum at q = 0.58 $Å^{-1}$ showing the conventional plasmon feature at ~ 20 eV and the new collective mode at ~ 4eV is displayed in (b). The momentum resolution is 0.06 $Å^{-1}$.

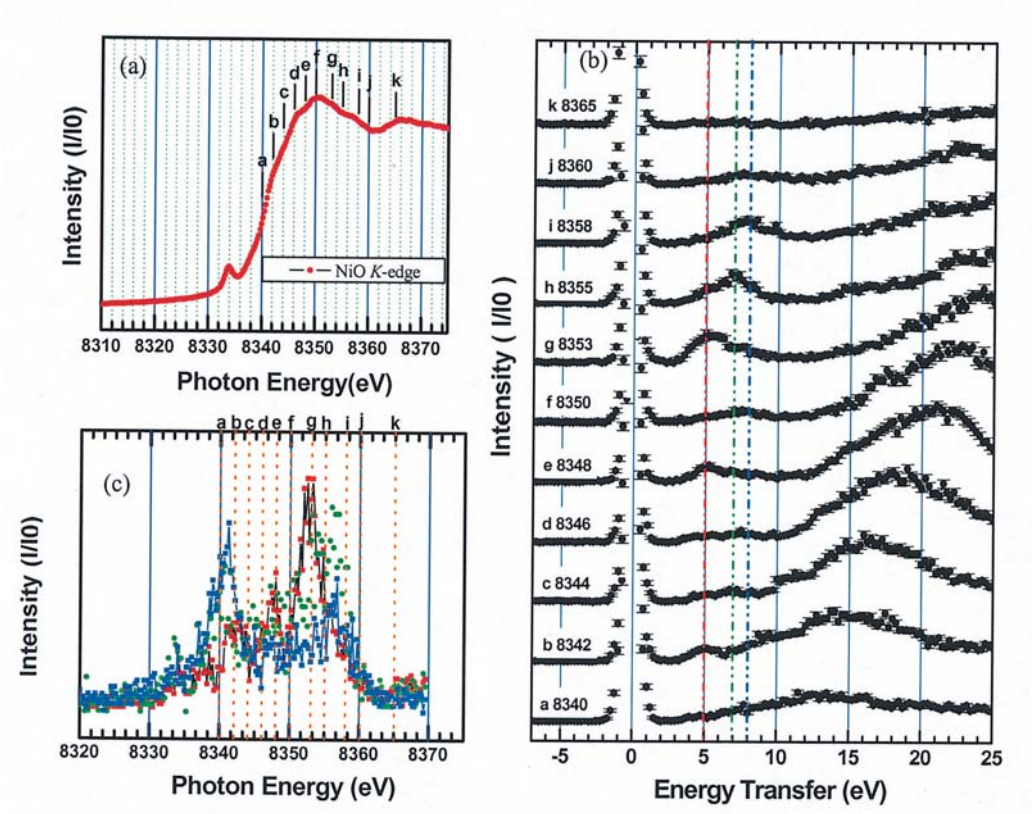

*Fig. 5: RIXS experimental data obtained from a NiO single crystal with **q** pointing along the [001] direction at 2θ = 30°. The K absorption edges are determined by taking the total fluorescent yield from the sample as shown in (a). The alphabets indicate the incident photon energies used to acquire the RIXS spectra displayed in (b), where the charge transfer excitations between 5 - 8eV can be seen between incident photon energies of 8342 - 8360eV. Constant final state spectra, in which the resonant enhancement of the charge transfer features can be seen more clearly, are shown in (c). They are taken by scanning simultaneously the incident photon energy E_1 and the RIXS energy E_2, maintaining a constant offset. The offset energies are 5eV (red), 7eV (green), and 8eV (blue). These spectra are equivalent to making a cut through the RIXS spectra in (b) at the corresponding energy transfers.*

desire to resolve finer features with better energy resolution but less incident flux, and a higher counting efficiency with wider bandwidth and more incident photons. The appropriate energy resolution depends on the specific experiment. For studies on single-particle and collective excitations in metals, semiconductors and insulators, the optimal energy resolution would be in the range 10-100 meV. For near edge scattering from light elements, and measurements of the dynamical structure factor in intermediate q-regime of metals, one needs energy resolutions from about 100meV up to 1eV. In cases where one does not require good momentum resolution but higher energy resolution is desirable (e.g., X-ray emission spectroscopy and near edge scattering), one may use multiple analyzers to enhance the counting efficiency. A new mechanical system, which provides the possibility to mount a 3 × 5 array of crystal analyzers, is now being developed and will be installed to the IXS spectrometer at the end of 2003. For RIXS experiments, improving the total energy resolution to 0.2 eV or better using 2-m radius analyzers will allow studies of the low-energy excitations in strongly correlated electron systems to be conducted on the beamline.

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