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MERIX—Next generation medium energy resolution inelastic X-ray scattering instrument at the APS

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ABSTRACT

MERIX (Medium Energy Resolution Inelastic X-ray Scattering) is an instrument for Resonant Inelastic X-ray Scattering (RIXS) studies, in the hard X-ray regime, designed to work with photons in the 5–12 keV range, spanning atomic resonances near the K-edges of 3d elements, and the L-edges of 4f and 5d elements. The energy analysis of inelastically scattered photons is performed with segmented spherical crystal analyzers in close to Bragg backscattering geometry. For each resonance (edge) a specially designed analyzer is used, fabricated from Ge, Si, or $LiNbO_3$ crystals. MERIX uses a position sensitive (micro-strip) detector to take snapshots of IXS spectra which are dispersed in space and over an energy range of a few eV with $\simeq$20–40 meV energy resolution. The spectral resolution of the MERIX spectrometer depends on the analyzer and varies from $\simeq$45 meV to $\simeq$170 meV, while the momentum transfer resolution is $\simeq$1–4 nm^{-1}. Samples are illuminated by micro-focused beams of size $\simeq 10\,\mu m \times 45\,\mu m$, allowing for studies at high-pressure and other extreme conditions. Polarization selectivity is ensured by vertical or horizontal momentum transfer scans. MERIX features $\simeq$100 times higher count-rates compared to previously built RIXS instruments.

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1. Introduction

New spectrometers have often sacrificed count-rate and cost for better spectral resolution. It is a rare breakthrough that improves all three, as became possible with an inelastic X-ray scattering spectrometer MERIX.

Momentum-resolved Inelastic X-ray Scattering (IXS) probes collective excitations in condensed matter. There are two major objects of studies: vibrational excitations and electronic excitations. Vibrational dynamics requires IXS spectrometers with a high energy resolution of about 1 meV. Studies of electronic excitations require in most cases spectrometers with a medium energy resolution typically in the range of 100 meV.[1]

In the last decade Resonant IXS (RIXS) has become one of mainstream spectroscopic techniques to study electronic excitations, due to the resonant enhancement of the inelastic scattering cross-section [1]. RIXS is a photon-in photon-out spectroscopy. No charged particles enter or leave the sample. Photon energy losses (e.g., at the Cu K-edge) are due to a "shake-up" process of the valence 3d-electrons, in between the creation and annihilation of the 1s–4p core exciton, as shown schematically in Fig. 1.

RIXS captures the physics of charge dynamics through the observation of momentum-dependent excitations. It probes excited electronic states: charge-transfer, Mott-Hubbard, $d-d$ excitations, etc. Unlike X-ray absorption spectroscopy there is no lifetime broadening due to deep core hole effects. RIXS provides momentum-dependent information, is bulk sensitive, element specific, and allows studies under extreme conditions (high pressure, magnetic fields, extreme temperatures, etc.). For a overview of the field see recent reviews [2,3].

A next generation IXS instrument in the hard X-ray regime for studies of electronic excitations, with targeted energy resolution about 100 meV, has been designed, built, and in operation since 2007 at the Advanced Photon Source (APS), Argonne National Laboratory, beamline 30ID. Known as MERIX, for Medium Energy Resolution Inelastic X-ray Scattering, the instrument comprises

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[1] It is certainly a compromise, as 1 meV resolution would also be ideally suited to electronic excitation studies, however, low count rates at such energy resolution, make the studies almost impossible with the existing light sources.

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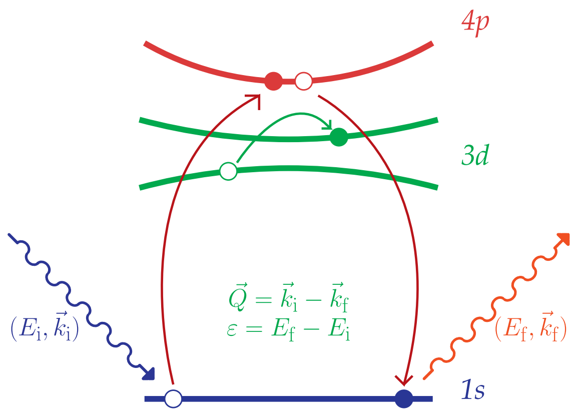

Fig. 1. Schematic presentation of a possible 3d electronic excitation with energy $\varepsilon = E_i - E_f$, and momentum $Q = k_i - k_f$, following an indirect K-edge RIXS process with incident and scattered photons having energies E_i, E_f, and momenta k_i, k_f, respectively.

an undulator X-ray source, a high-heat-load monochromator, a medium-resolution monochromator, a focusing KB mirror system, and as the main component, the MERIX spectrometer. It is used for studies of collective valence electron excitations, primarily in 3d and 4f correlated electron systems. A basic feature of the inelastic cross section in the 5–12 keV range is the presence of resonances at transition metal (and other) absorption edges. As a result of these resonances, there are certain key energies at which MERIX works (a principal one being the Cu K-edge), using RIXS. This technique is extremely photon hungry and even small gains in intensity can be extremely important. This fact led to several key design decisions.

Design of the MERIX spectrometer was influenced by the previous experience with RIXS spectrometers built at NSLS [4], USA, at the ESRF [5,6], France, BL11XU and BL12XU at SPring-8, Japan, and at the APS 9ID [7], USA. In particular, the latter spectrometer served as a prototype for MERIX. Several significant improvements have been added, allowing MERIX to achieve better energy resolution with simultaneously up to 100 times higher count-rates, primarily due to implementing the advanced technique of dispersive spectral analysis first proposed in [5,6]. MERIX offers its users a broad set of RIXS analyzers covering almost all K-edges of 3d elements and some L-edges of 4f and 5d elements.

Understanding the polarization dependence of the scattering at resonance is crucial. This is addressed by the ability of the spectrometer to scatter both vertically and horizontally. The MERIX instrument is equipped with focusing mirrors, providing micro-focused beam on the sample with a spot size of ≃10 μm (V) × 45 μm (H) – enabling studies of small samples and samples under high pressure. The MERIX monochromator delivers X-ray photons to the sample with a 70-meV or 120-meV energy bandwidths. The flux in the micro-focused beam of 9-keV photons on the sample is 1.2×10^{12} ph/s (with 70-meV bandwidth) or 2×10^{12} ph/s (with 120-meV bandwidth). The total energy resolution of the instrument at the Cu K-edge resonance is 85 meV. The main MERIX parameters are summarized in Table 1.

MERIX instrument components are described in more detail in the following sections, with the emphasis on the MERIX spectrometer.

2. Layout of MERIX instrument

MERIX has been built at the 30ID undulator beamline of the Advanced Photon Source, a 7 GeV and 100 mA machine in operation at Argonne National Laboratory since 1996. MERIX occupies two stations A and B of the beamline. The optical scheme of the MERIX instrument is shown in Fig. 2.

Table 1
Typical parameters of the MERIX instrument, shown for 9 keV X-rays, when it is appropriate.

Incident photon energy range	5–12 keV
Energy transfer range	$\varepsilon \lesssim 20$ eV
Spectrometer energy resolution	$\gtrsim$45 meV
Monochromator energy resolution	$\simeq$70 meV
Instrument energy resolution	$\gtrsim$85 meV
Momentum transfer range	$Q \lesssim 0$–$80\ \mathrm{nm}^{-1}$
Momentum transfer resolution	$\Delta Q \simeq 1$–$4\ \mathrm{nm}^{-1}$
Beam size on sample	10 (V) μm × 45 (H) μm
Flux on sample	1.2×10^{12} ph/s/70 meV

The beamline front-end is of an enhanced design that allows up to three permanent-magnet planar undulators to be used together. Currently, there are two undulators installed with a period of 3.0 cm and 2.4 m length operated in tandem (total length 4.8 m, 160 periods). The two insertion devices were chosen on the basis of their tunability over the required energy range, and for maximum flux per unit power generated.

Station A is a white beam station. It houses white-beam-defining components, a high-heat-load monochromator (HHLM), and a monochromatic photon shutter. The beam-defining components are water-cooled double Be window (total thickness of 0.5 mm), the white beam slit system to define the beam prior to incidence on the monochromator, and in addition, the bremsstrahlung stop.

The high-heat-load monochromator is a double-diamond-crystal water-cooled, with constant 25-mm vertical off-set, tunable between 5 and 30 keV. The (1 1 1) Bragg reflection from diamond crystals is used. The diamond crystals are of type IIa, manufactured by Sumitomo Electric Industries, Ltd. of Japan [8]. The mechanical assembly is built by Kohzu-Seiki Co. of Japan. Typical spectral flux density of 9 keV photons measured after the HHLM through a 2 mm × 0.5 mm opening in the white beam slit is $\simeq 4.5 \times 10^{13}$ photons/s/eV. The angular acceptance for 9 keV X-rays is 25 μrad, while spectral bandwidth of the monochromator is 0.65 eV, in agreement with theoretical expectations. To exclude temperature drifts under variable heat load conditions an active hardware feedback has been implemented [9].

Station B is a monochromatic station and houses the high-resolution MERIX monochromators followed by a focusing KB mirror system. The MERIX spectrometer is placed at the end of this station.

3. MERIX monochromator

Achieving monochromatization of hard X-rays with a bandwidth ≃50–100 meV, is not difficult. It can be done by employing the double crystal monochromator scheme in the same non-dispersive (+ −) configuration as for the HHLM, with properly chosen Bragg reflections. However, this would not allow constant spectral bandwidth over the whole 5–12 keV energy range. In addition, one cannot achieve the highest throughput, as the angular acceptance is not necessarily very well matched to the angular divergence of the incident beam. A four crystal monochromator scheme in the (+ − − +) configuration [10] with asymmetrically cut crystals [11] is used for MERIX, to achieve a higher throughput and a more or less constant bandpass over the 5–12 keV range [12], as shown schematically in Fig. 2. All four crystals use the same Si(0 0 4) Bragg reflection with the asymmetry angle $\eta = 7.5^{\circ}$ to crystal surface. The monochromator bandpass ΔE_M is, in the first approximation [11,13]

$$\frac{\Delta E_M}{E} = \frac{\epsilon_H}{2} b\sqrt{b}, \quad b = \frac{\sin(\theta - \eta)}{\sin(\theta + \eta)}, \quad \sin\theta = \frac{E_H}{E}, \tag{1}$$