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# High-efficiency high-energy-resolution spectrometer for inelastic X-ray scattering

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**Abstract**

A nine-element analyzer system for inelastic X-ray scattering has been designed and constructed. Each individual analyzer crystal is carefully aligned with an inverse joystick goniometer. For the analyzers silicon wafers with 100 mm diameter are spherically bent to 1 or 0.85 m radius, respectively. Additionally, an analyzer with an extra small radius of 0.182 m and diameter of 100 mm was constructed for X-ray absorption spectroscopy in fluorescence mode. All analyzer crystals with large radius have highly uniform focusing property. The total energy resolution is approximately 0.5 eV at backscattering for the 1 m radius Si(440) analyzer array and approximately 4 eV for the 0.182 m radius Si(440) analyzer at 6493 eV.


## 1. Background and spectrometer design

Over the last 20 years, high-energy resolution inelastic X-ray scattering and valence band resonant emission spectroscopy have gained interest in the scientific community. The high flux from synchrotron radiation sources has significantly benefited this research area. Significant experimental work was carried out in a large variety of fields, such as in physics, chemistry, material science and biology [1–6].

Due to the low cross-section in inelastic scattering, one needs to consider increasing the incidence flux or the efficiency of the spectrometer. Since the flux from most synchrotron radiation sources is close to its theoretical limit, the only alternative remains to improve the spectrometer to maximize detection efficiency.

We chose to increase the solid angle covered by the analyzer system to collect more scattered photons. A few systems have already been built and are in use [7,8] for high- and low-resolution inelastic X-ray scattering. These systems use linear arrays of analyzer crystals, and either separate detectors for each analyzer to measure the signal at several momentum transfer values at the same time [7], or just to cover a larger solid angle by using one detector [8].

In our case, the multiple crystal analyzers are mounted in a two-dimensional compact array. Each analyzer is mounted on its individual "inverted-joystick" goniometer [9] for perfect alignment of all nine analyzer crystals. Furthermore, translation of the analyzer system and of the detector keeps sample, analyzer crystals and detector on the Rowland circle, which improves the resolution. Finally, the system is modular and hence it is possible to upgrade the system with a specially designed nine-element detector, and to measure, by slightly detuning each analyzer crystal, the spectrum at nine different momentum transfer values at the same time.

The spectrometer is based on a two-circle goniometer. Its accuracy is better than 25 arcsec and its repeatability is less than 4 arcsec. Analyzer energy-scans are simple $\theta$–$2\theta$ scans. Fig. 1 shows a schematic drawing of our nine-crystal analyzer array spectrometer. The crystal analyzers are made from flat crystal wafers, which are bent to a spherically curved shape to fit on curved substrate mirror blank. Several aspects such as energy resolution, solid angle covered by the analyzer, and the manufacturing process of the analyzers influence each other. The bending radius influences the energy resolution, and it is limited by the strain tensor that causes the crystal to break if the bending radius is too small compared to the thickness of the crystal. The bending radius also determines the distance of the analyzer crystal to the sample, and thus the covered solid angle. The strain of bending the crystal broadens the rocking curve, thus increasing the band-width and decreasing the peak-reflectivity.

We chose to use a bending radius of 1 m with a diameter of 100 mm and a thickness of 300 μm. The silicon wafers (FZ type with resistivity larger than 1000 Ω cm, TTV < 5 um, both surfaces polished) are glued onto glass substrates, which gives reasonable energy resolution of about 0.5 eV for Si(440) close

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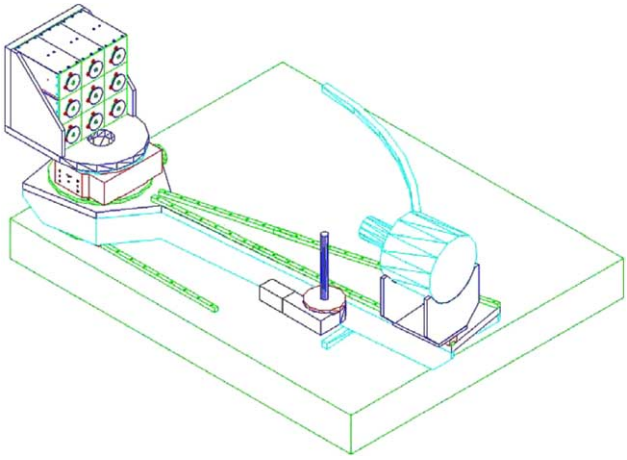

Fig. 1. Schematic drawing of the nine-crystal array IXS spectrometer.

to backscattering. The analyzer array with nine analyzers covers a solid angle of approximately 1.1%.

In addition to the multi-analyzer array, an analyzer with an extra short radius of 0.182 m, based on wafers of 100 mm diameter and thickness of 100 μm was also developed. This single analyzer covers 2% of the full solid angle. Although the energy resolution cannot compete with larger radius analyzers, it is still very useful in some experiments, such as in fluorescence mode X-ray absorption spectroscopy (XAS) and spin polarized X-ray absorption spectroscopy (SP-XAS). With the resolution of 4 eV it attains a high signal to noise ratio suitable for XAS and extended X-ray absorption fine structure (EXAFS) measurements.

## 2. Experiment results

We performed several experiments with the new spectrometer at two beamlines at the NSLS in Brookhaven National Laboratory, which include non-resonant Raman scattering (NRRS), X-ray emission spectroscopy (XES), and SP-XAS.

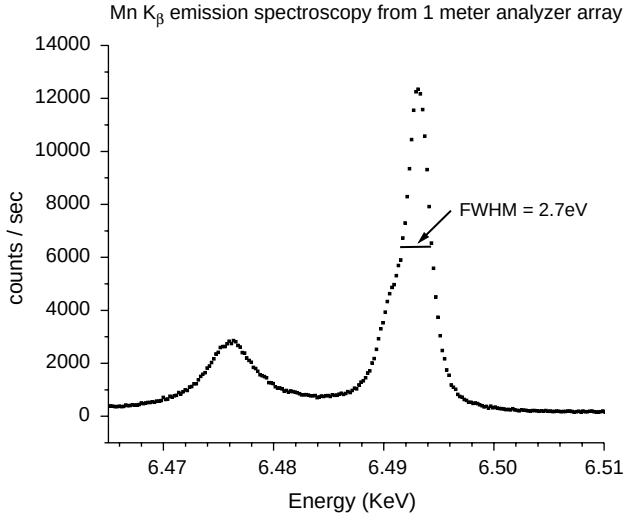


Fig. 2. Mn $K_\beta$ emission measured with 1 m radius analyzer array at X21 at NSLS.

The experiments were carried out at beamline X21 with the analyzer array (nine Si(440) crystals with bending radius 1 m). The incidence flux at X21 is $2\times10^{10}$ photons/s in a bandwidth of 0.2 eV, and the beam is focused to a spot size of 0.5 mm × 1 mm. At beamline X23B, the single 0.182 m short radius Si(440) analyzer was used; at beamline X23B the incident flux is $10^9$ photons/s with 1 mm × 1 mm spot size.

The following results are measured at beamline X21 with the nine-analyzer array: Fig. 2 shows the $MnF_2$ X-ray emission spectrum with high-energy resolution. The two peaks result from different spin states: The main peak at 6.493 keV originates from Mn 3p spin down hole final state, while the satellite peak at 6.476 keV is from Mn 3p spin up hole final state [4]. That splitting gives the opportunity to measure locally spin resolved XAS or EXAFS.

Fig. 3 displays measurements of NRRS on the carbon K-edge of graphite. Our graphite sample was a thin foil of exfoliated graphite, which still preserves a high orientation of the graphite planes. The width of the distribution function is of the order of 30°. In the experiment, the scattering vector was aligned in the direction of the *c*-axis, so that we probe the $p_z$-orbitals, however, a small projection of the $sp^2$ orbitals in the plane still enters the spectrum.

The following data were taken at beamline X23B (a standard bending magnet line) with a single Si(440) crystal analyzer with the extra short radius of 0. 182 mm. In this case, the crystal analyzer is mounted on one single "inverted-joystick" goniometer. The detector position was fixed, and had an 18 × 18 $mm^2$ entry aperture.

Fig. 4 shows Mn $K_\beta$ emission spectrum of an $MnF_2$ powder sample. The count rate is quite high compared to the nine-crystal array due to the large solid angle (∼2% of 4π). Although energy resolution is fair (around 4 eV), it has an advantage where count rate is considered. It is obvious that two spin channels are clearly separated, which will enable one to

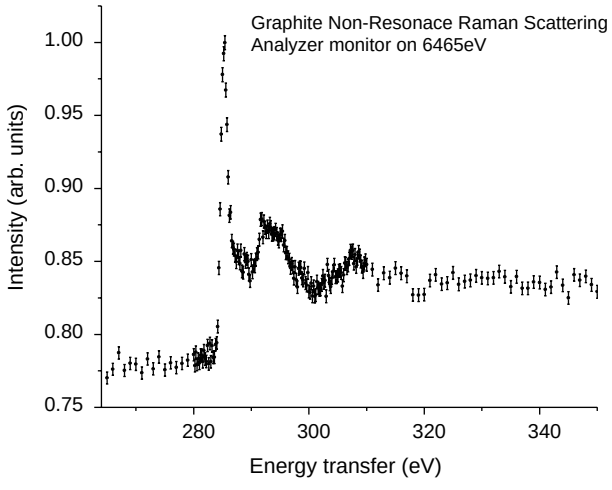


Fig. 3. Carbon K-edge non-resonant Raman scattering spectrum of graphite foil measured with the momentum transfer parallel to the *c*-axis. The narrow peak at 287 eV is caused by transitions into the $p_z$-orbitals, while the broader structure at 290 eV results from transitions into the $sp^2$-band. These data differ from other published data of perfect powder samples, since this sample is not a powder.

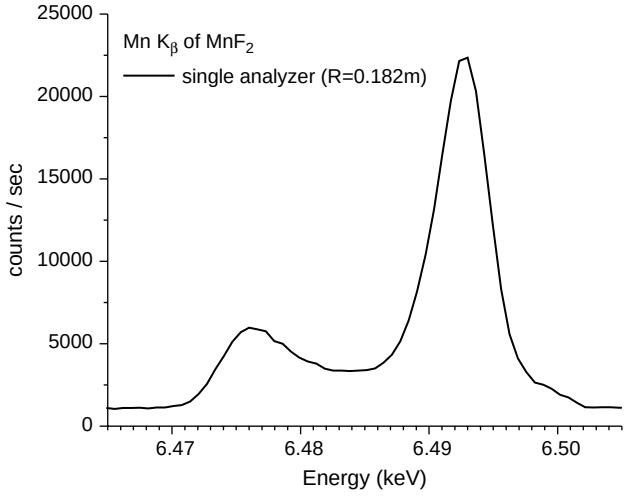


Fig. 4. $MnF_2$ $K_\beta$ emission measured with single crystal analyzer with radius $R=182$ mm and 100 mm diameter.

carry out SP-XAS. Note that the flux of this beam line flux is just $10^9$ photons/s.

Fig. 5 shows $MnF_2$ SP-XAS with spin down and up measured with the single short radius bent analyzer. Each data point has a 2 s accumulation time.

Fig. 6 shows SP-XAS of $Nd_{0.5}Sr_{0.5}MnO_3$ (solid line). In this case, the Nd L-edge signal around 6730 eV is not observed (position marked with an arrow, a little dip is due to concentrated Nd partly absorbing incident X-ray, which can be treated by detailed post-collection analysis methods). In conventional EXAFS measured with an ion chamber (dotted line) the Nd LII-edge is clearly observed. Since the Mn $K_\alpha$(5899 eV) and Nd $L_{\beta 2}$(6089 eV) fluorescence are quite close (190 eV), conventional detector systems cannot separate these fluorescence lines clearly, even with a Ge detector (150 eV resolution) since the high resolution is attained only at low count rates. With our analyzer system, with an energy

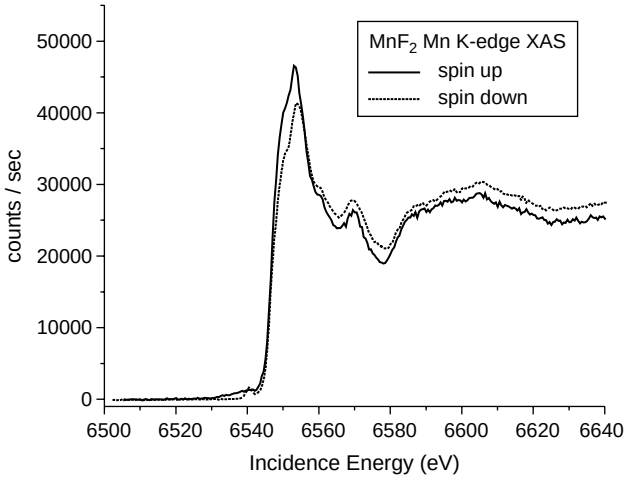


Fig. 5. Mn K-edge SP-XAS of $MnF_2$. Black solid line is spin up XAS, in which the analyzer monitored the Mn $K_\beta$ satellite line at 6476 eV, while the dotted line is spin down XAS signal in which the analyzer monitored the Mn $K_\beta$ main peak at 6493eV.

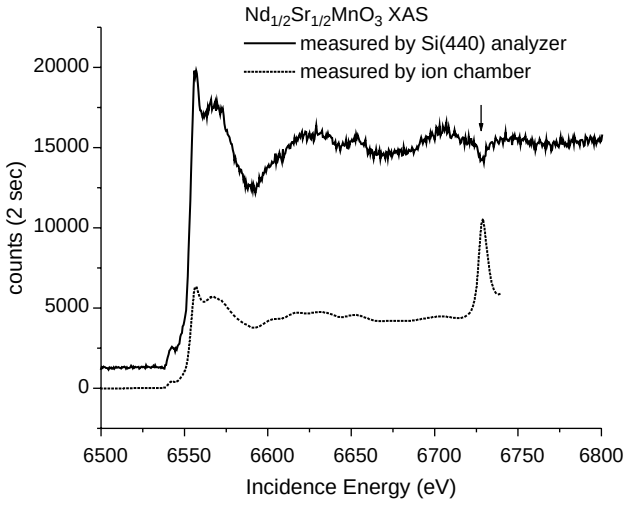


Fig. 6. Solid line is SP-XAS of $Nd_{0.5}Sr_{0.5}MnO_3$, measured with $K_\beta$ fluorescence mode. The dotted line gives conventional EXAFS spectra measured in transmission mode with ion chambers.

resolution below 4 eV, these two fluorescence lines can be easily separated and a high efficiency detector can be used.

Fig. 7 shows SP-XAS from a single crystal film of $La_{0.8}MnO_3$ on $LaAlO_3$ with thickness 2000 Å. No diffraction peaks are observed due to the high-energy resolution. The analyzer system can clearly measure the XAS or EXAFS by monitoring fluorescence lines. Here, we chose the Mn $K_\beta$ line, since the Si(440) is not suitable for the detection of the $K_\alpha$ line.

## 3. Summary

A high-energy resolution IXS spectrometer was built with a nine-crystal analyzer array. With a bending radius of 1 m and diameter of 100 mm, Si(440) analyzer crystals result in an energy resolution about 0.5 eV at backscattering. An extra short 182 mm radius 100 mm diameter analyzer was also developed; its energy resolution is about 4 eV at Mn $K_\beta$ line with 2% of the full solid angle collected. High-energy resolution measurements were carried out in NRRS, XES, and SP-XAS.

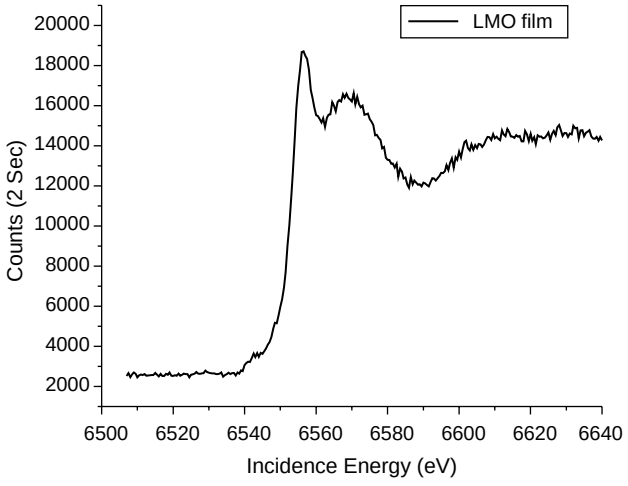


Fig. 7. SP-XAS of a single crystal film of $La_{0.8}MnO_3$ on $LaAlO_3$ with thickness 2000 Å. The analyzer monitored the Mn $K_\beta$ main peak.

## Acknowledgements

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