

## A New Method of Non-Resonant X-ray Magnetic Scattering

Non-resonant X-ray magnetic scattering (NRXMS) is a very promising technique in studying the magnetic structure of small samples, such as microcrystals and/or thin films [1]. Since X-rays are an electromagnetic wave, they are sensitive to magnetic as well as to charge distributions in condensed matter. In addition, orbital angular momentum L and spin angular momentum S scatter X-rays differently, which provides a way of separating the L and S contributions to magnetization experimentally. Brilliant X-rays from a third-generation synchrotron radiation source make up quite a small cross section generated by NRXMS.

The X-ray susceptibility of matter, including magnetic effects, needs to be treated as a tensor [2]. Indeed, magnetic scattering can be discriminated from charge scattering by the polarization analysis of scattering processes. The polarization of X-rays is analyzed using a suitable analyzer crystal, which gives rise to scattering at 90°. A polarization analyzer eliminates polarization parallel to its own scattering plane; at the same time, the remaining polarization is considerably suppressed because the scattering factor and the Debye-Waller factor take a small value with the scattering at 90°. Then, the scattered intensity from a sample will be considerably reduced by the diffraction process on an analyzer crystal. In order to overcome the intensity loss by the analyzer crystal, we have developed a new polarization analysis technique without using an analyzer crystal [3].

Figure 1(a) shows a schematic view of the experimental setup with a conventional polarization analyzer. In this case, the linearly polarized synchrotron radiation and vertical scattering plane are fixed. The

polarization of the scattered X-rays from a sample is analyzed by rotating an analyzer crystal around the axis which is parallel to the scattered X-rays. On the other hand, in our new technique a variable scattering plane is employed instead of an analyzer crystal, where a sample crystal also plays the role of an analyzer crystal and the linearly polarized synchrotron radiation is kept fixed. A schematic of the inclined scattering plane geometry is seen in Fig. 1(b). In this figure,  $\vartheta$  represents the inclination angle of the scattering plane from the vertical plane.

The polarizations perpendicular and parallel to the scattering plane are defined as the  $\sigma$  and  $\pi$ polarizations, respectively. The amplitude of linearly polarized synchrotron radiation is parameterized as  $E(\vartheta) = E(\hat{e}_{\sigma} \cos \vartheta + \hat{e}_{\pi} \sin \vartheta)$ , where  $\hat{e}_{\sigma}$  and  $\hat{e}_{\pi}$  are the polarization vectors. One can consider the new technique to be the manipulation of an incident polarization. The inclination angle dependence of the charge scattering intensity is given by  $I_c(\vartheta) \propto \cos 2\vartheta +$  $[(1 + \cos 2\theta)/(1 - \cos 2\theta)]$ . Similarly, that of the magnetic scattering intensity is written as  $I_m(\vartheta) \propto \cos(2\vartheta + \alpha) + B$ where *B* is the baseline and  $\alpha$  the phase shift arising from the off-diagonal element of the X-ray susceptibility tensor. Figure 2 shows the inclination angle dependence of the charge and magnetic scattering intensities. It is apparent that the two scattering processes exhibit respective  $\vartheta$  -dependences. Therefore, the magnetic scattering can be discriminated from the charge scattering without using an analyzer crystal.



In order to demonstrate the validity of the new technique, we have conducted NRXMS experiments

Fig. 1. (a) Schematic view of the experimental setup with the conventional polarization analyzer.  $k^i$  is the incident wavevector and  $2\theta$  is the scattering angle. (b) A schematic view of the inclined scattering plane geometry.  $\vartheta$  is the inclination angle of the scattering plane from the vertical plane.



on the rare earth metal dysprosium, which has a helical magnetic structure in the temperature range between 89 K and 179 K. The single-crystal sample was disk-shaped, 5 mm in diameter and 1 mm thick and had its *c*-axis of a hexagonal lattice normal to the surface of the disk. The experiments were carried out at beamline **BL46XU** using a six-circle diffractometer. The incident photon energy was 20 keV and a NaI(TI) scintillation detector was used. The sample was mounted on the cold head of a closed-cycle helium refrigerator.

As the sample was cooled below the Néel temperature, magnetic reflections were observed as satellites of the fundamental reflections along the  $c^*$ direction. Figure 3 shows the inclination angle dependence of the intensity of the 008<sup>-</sup> satellite reflection normalized by the 008 fundamental reflection. The normalization by the close Bragg reflection was necessary because it was almost impossible to keep an identical sample volume illuminated by the incident X-ray beam, particularly through a blind shroud. Measurements were performed with a sample temperature of 120 K. The inclination angle dependence shown in the figure cannot be explained by the charge scattering that has an intensity maxima with  $\vartheta = 0^{\circ}$ . The solid curve in the figure, which represents the intensity variation expected for the simple basal plane spiral with L/S= 2, coincides with the experimental result very well.



Fig. 2. Inclination angle dependence of scattering intensities. The charge scattering will show a similar dependence to the dashed curve. The magnetic scattering is followed by the phase shift shown as a solid curve.

Consequently, it was confirmed, by the new technique, that the  $008^-$  satellite reflection is indeed the magnetic reflection.

In conclusion, we have developed a new polarization analysis technique without using an analyzer crystal. Thereby, the diffraction data of magnetic scattering can be collected in a short time. Our technique fully conserves the capability to separate the *S* and *L* contributions to magnetization and is applicable not only to NRXMS but also to resonant X-ray scattering.



Fig. 3. Inclination angle dependence of the intensity of the  $008^-$  satellite reflection normalized by the 008 fundamental reflection at T = 120 K. The solid curve represents the result of fitting with L(K)/S(K) = 2.

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## References

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