Magnetoelectric X-ray Scattering in GaFeO$_3$ and Fe$_3$O$_4$

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Introduction
The linear magnetoelectric (ME) effect can be observed in magnetically ordered crystals whose magnetic group does not contain the space inversion operation, i.e., where time reversal $R$ and space inversion $I$ operations are broken simultaneously. In optical or x-ray region, the novel nonreciprocal magneto-optical phenomena, termed magnetochiral or optical ME effect, are expected to show up in such a crystal as characterized by a different optical response (absorption or refraction) depending on light propagation direction $k$. Spectroscopy of this nonreciprocal effect is important to clarify the microscopic mechanism of the ME effect which is attracting renewed interest also from the viewpoint of the application to spin-electronics.

In this study, we have investigated resonant ME x-ray scattering (MEXS) in a polar ferrimagnet GaFeO$_3$ and a spinel-type ferrimagnet Fe$_3$O$_4$. Here, we propose that present element-specific resonant MEXS would be a unique probe of the magnetic state with broken centrosymmetry.

Experimental
The samples used in this work were GaFeO$_3$ and Fe$_3$O$_4$ single crystals grown by a floating-zone method. The measurements of magnetic x-ray scattering were carried out at BL-1A and 16A2. An incident beam was injected on the (010) and (111) surface for GaFeO$_3$ and Fe$_3$O$_4$, respectively, and a modulated magnetic field was applied perpendicular to the incident beam, namely in Voigt configuration. The modulated magnetic and/or ME scattering intensity $\Delta I(f)$ was recorded as the ratio of spin-dependent to spin-independent scattering intensity $\Delta I(f)/I$ with a lock-in amplifier.

Results and Discussions
First, we describe the results in a polar ferrimagnet GaFeO$_3$. For the (020) and (040) Bragg reflections, we have observed a change in the scattering intensity with reversal of the magnetization direction near the Fe $K$ pre-edge [1]. In Fig. 1, we show the magnetic-field modulation spectra $\Delta I(f)/I$ for the (020) and (040) Bragg reflections, respectively. One can see a large energy-independent component for (020) and resonant peaks for both reflections. The energy-independent component is assigned to the well-known conventional magnetic x-ray scattering (MXS) from the viewpoint of the spin structure of GaFeO$_3$. On the other hand, the resonant signal near the pre-edge is possibly related to MEXS, which is sensitive not only to the magnetism but also to the breaking of the inversion symmetry [2]. Actually, we certainly observed, that the MEXS signal is reversed in sign when the sample (spontaneous polarization) was reversed, which evidences our expectation (Fig. 1).

Figure 1: Magnetic field modulation spectra of Bragg reflections at 50 K for (020) (open circles), (040) (solid circles), and (040) (open squares). The absorption spectrum around the Fe $K$-absorption edge of GaFeO$_3$ is also shown by a solid line.

Next, we describe the results of a spinel-type ferrimagnet Fe$_3$O$_4$. Although $I$ symmetry is globally restored, it is locally broken at the tetrahedral A site. It is possible to detect the local ME scattering at noncentrosymmetric site as the nonreciprocal effect, due to the possibility of adjusting phase factors, even in systems where the $I$ symmetry is globally restored. At the (222) Bragg reflection with a specific structure factor, in fact, resonant magnetic component was observed around the pre-edge region of Fe $K$-absorption. As compared with the (333) and (444) Bragg reflections, we deduced that the resonant structure observed at the (222) Bragg reflection is due to MEXS at the spinel A site with locally broken inversion symmetry ($T_d$).

References

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