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X-ray Directional Dichroism in $GaFeO_3$

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Introduction

Polar ferromagnets are expected to show non-reciprocal directional dichroism (NDD), when an electromagnetic wave propagates perpendicular to the spontaneous polarization and magnetization. The absorption of the electromagnetic wave can change with the sign reversal of the vector triple product of propagation **k**, spontaneous polarization $\mathbf{P_0}$, and spontaneous magnetization $\mathbf{M_0}$. The NDD originates from interference between electric dipole (E1) and electric quadrupole (E2) terms. The E2 term is enhanced in proportion to light wave number. This is the reason why resonant x-ray absorption is quite advantageous to detect the NDD. Furthermore, the advantage of using a ferromagnetic compound is obvious both for the observation and the possible application of the NDD: A weak external magnetic field can reverse the direction of $\mathbf{M_0}$ while keeping the same **k**-vector, and hence cause the dichroism that is equivalent to the reversal of x-ray **k**-vector while keeping the $\mathbf{M_0}$ direction.

We adopted $GaFeO_3$ as a good candidate of the ferromagnet that can show NDD. $GaFeO_3$ is a rare example of the compounds, in which piezoelectricity and ferromagnetism coexist [1]. Pyroelectric polarization is generated along the b axis, while a magnetic easy-axis is parallel to the c axis.

Experimental

A single crystal boule of $GaFeO_3$ was grown for the first time by a floating zone (FZ) method under 9 atm O_2 atmosphere. For the measurement of x-ray non-reciprocal directional dichroism (XNDD), a plate-like specimen was cut from the crystal rod and the both surfaces were polished. A synchrotron x-ray beam was monochromated by a pair of Si (111) crystals. The x-ray beam was injected on the (100) plane and ac magnetic field was applied along the c axis, i.e., in the Voigt configuration. The modulated signal component was detected as the XNDD signal with a lock-in amplifier.

Results and Discussions

A clear XNDD was observed around the pre-edge and around the main edge [2]. The magnitude of the XNDD around the pre-edge is much larger than that around the main-edge for the both x-ray polarizations E^{ω}//b and E^{ω}//c. The ratio of the XNDD to relevant x-ray absorption corrected by the subtraction of background irrelevant to a resonant absorption is as large as 1.2% around the pre-edge. This indicates that the quadrupole transition from Fe 1s to Fe 3d plays an important role in this novel phenomenon, which shows a clear contrast to the x-ray magnetic circular dichroism (XMCD) which is mainly due to the dipole transition.

The observed XNDD is clearly distinguished from conventional magneto-optical effects such as the Cotton-Mouton effect and XMCD. The Cotton-Mouton effect is proportional to the square of the magnetization. The 2f-modulation component was hardly observed in the present case. Therefore the possibility will be excluded that the present signal of the first-harmonic component would be caused by the Cotton-Mouton effect, even if possible asymmetry of an applied magnetic field is taken into account. As for XMCD, the signal should not be observed in the Voigt configuration of an external magnetic field, combined with a linearly-polarized x-ray beam. In order to deny completely the possibility that the f-modulation would result from XMCD, we also performed the modulation measurement under the condition that the direction of an applied magnetic field was tilted by 10 degree around the b axis. The XMCD signal should be reverted with inverting the Faraday component of the applied magnetic field. The observed XNDD was not reversed within the present experimental accuracy. This indicates that the observed signal should be derived from the genuine Voigt component of the magnetic field.

We also confirmed through the temperature dependence of the XNDD spectra at the pre-edge, that the reversal of $\mathbf{M_0}$ itself is responsible for the observed dichroism with respect to the field direction. The behavior of the temperature dependence of the XNDD is similar to that of the magnetization after a zero-field cooling. The details will be reported on Photon Factory News in near future.

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