Full control of magnetic moment in multiferroics Ba2CoGe2O7

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Electric control of magnetic moment is an intriguing topic in condensed mater physics since it can be applied to a novel device. Coexistence of magnetic order and electric polarization, multiferroicity, has a possibility to realize such a control. So far spin helicity was controlled in $TbMnO_3$ [1] and magnetic domain was controlled in Hexaferrite [2], but controlling local magnetic moment by applying electric field has not been demonstrated yet. $Ba_2MGe_2O_7$ (M = Co, Mn) are multiferroic compounds with spin-dependent $p-d$ hybridization mechanism [3]. The crystal structure is a tetragonal and the space group is P-421m. Below T_N = 6.7 K, the spin antiferromagnetically lies along the crystallographic a axis, and the electric polarization antiferroelectrically lies along the c axis. It is expected that the applying electric field along the c axis changes the antiferroelectric state to ferroelectric state with the magnetic moment rotating from [100] to [110]. Soda et al. demonstrated the rotation of the magnetic moment in the ab plane [4], but the moment did not make complete rotation by 45 degrees due to a large in-plane magnetic anisotropy. It was only 6 degrees at T = 1.5 K and E = 1.5 MV/m at best. Then we performed a similar experiment on $Ba_2MnGe_2O_7$ [5] having smaller anisotropy energy at PSI. The rotation of the magnetic moment was observed at 1.5 K, and the angle as large as 20 degrees was achieved by increasing the temperature. The perfect rotation of 45 degrees was, however, not achieved because of the short of the electric field. It should be noted that a non-linear behavior was observed in the field dependence of the rotation angle, and a non-trivial plateau was found at 2 MV/m $<$ E $<$ 5 MV/m, even though a simple linear behavior was expected [4]. Even though the anisotropy energy of $Ba_2MnGe_2O_7$ is smaller than that of $Ba_2CoGe_2O_7$, the electric polarization is also smaller, resulting in the smaller rotation angle at the same electric field at the base temperature: 1.5 degree for $Ba_2MnGe_2O_7$ and 6 degrees for $Ba_2CoGe_2O_7$ at E = 1.5 MV/m. Use of $Ba_2CoGe_2O_7$ is better choice. We have done the Polarized Neutron Diffraction to achieve the perfect rotation by applying stronger electric field on thinner $Ba_2CoGe_2O_7$ sample and by reducing the anisotropy energy at high temperature. Then we tried to detect non-trivial field dependence of the rotation angle of the magnetic moment to understand. A single crystal sample was grown by the floating-zone method. The single crystal was aligned so that the crystallographic ab plane was in the horizontal plane. For the single crystal with a thickness of 2.65 mm, aluminum electrodes were deposited onto the faces of (001) to apply the electric field. The sample was set in the Al sample can, and then sample space was vacuumed. Maximum voltage in the used equipment was 7.0 kV. Heusler alloy was used to obtain the polarized neutrons with the energy of 13.5 meV. Magnetic Bragg peaks are observed below T_N. From the analysis of the peaks, it is found that the easy-plane type antiferromagnetic structure having a magnetic propagation vector $\mathbf{k}_{mag} = (0, 0, 0)$ is realized, which is consistent with the previous report [6]. Since the sample has crystallographic domains, we performed the $\theta - 2\theta$ scan at maximum point of omega scan in order to estimated integrated intensity. (h, k, l)= (2, 1, 0) and its equivalent reflections were measured to investigate the electric field and temperature dependence. We assume that the magnetic moments continuously rotate from [100] (or [010]) to [110] directions when the antiferroelectric state becomes the ferroelectric state retaining an-

tiferromagnetic structure by applying the electric field along the c axis as same as the previous report [4]. Under this assumption the intensity variation at (2, 1, 0) and its equivalent position is represented by the sinusoidal curve as follows:

$$I \propto 1 + \sin 2\theta \sin 2\omega \qquad (1)$$

where I is the intensity of each magnetic reflections, and the ω is a rotation angle of the magnetic moment from [100] or [010] to [110]. The θ is the angle between the scattering vector and a axis. The rotation angle of the magnetic moment is evaluated from the amplitude of the sinusoidal curve. Figure 1 shows electric field dependences of the normalized magnetic intensities as a function of θ at 1.5 K. Normalized magnetic intensities were got from intensities divided by the average of intensities which were evaluated from magnetic intensities divided nuclear intensities at same Q position. The data show a sinusoidal curve at each electric field like equation (1). The amplitude increases with increase of the electric field, which means that the rotation angle of magnetic moment increases with. From fitting the data by equation (1), we obtained the rotation angle of magnetic moment against the electric field at 1.5 K, 6.0 K and 6.3 K. At 1.5 K data, the rotation angle is almost consistent with previous report. The rotation angle at 6.0 K and 6.3 K was reproduced by the theoretical calculation at 1.5 K in previous study [4], even though we apply higher electric field. This means that the controllability of magnetic moment is independence of temperature. Furthermore, non-trivial field dependence of the rotation angle of the magnetic moment was not observed.

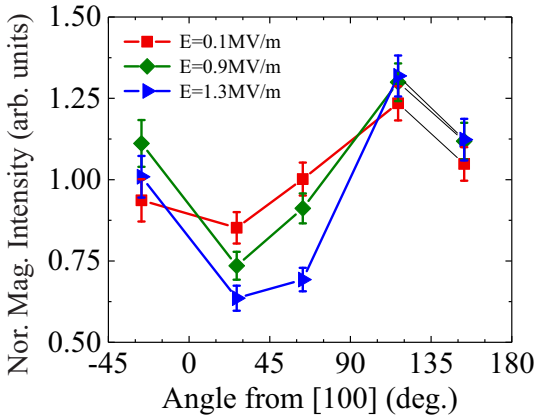

Fig. 1. Electric field dependences of normalized magnetic intensity at 1.5K.

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