Cycloidal Spin Order in (Gd,Tb)MnO3 with Electric Polarization along the a-Axis

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In a orthorhombically distorted perovskite $TbMnO_3$, ferroelectric polarization appears along the *c* axis below $T_C \sim 27$ K. The polarization flops by 90 degrees to the *a* direction by the application of a magnetic field along the *b* or *a* axis. This magnetically-induced polarization flop should be regarded as a magnetic phase transition accompanied by the ferroelectric transition.

The zero-field magnetic state below T_C was pinned down to a cycloid with the spin spiral plane perpendicular to the *a* axis. The cycloid should induce electric polarization through the inverse effect of Dzyaloshinski-Moriya (DM) interaction. In contrast, the magnetic structure of the high-field phase was not settled yet. From the theoretical point of view, there are at least two possible spin arrangements which can induce electric polarization along the *a* axis. One is of the cycloid type with the spiral plane perpendicular to the *c* axis, and the other is of the commensurate collinear type with the propagation vector (0 1/4 1). A synchrotron x-ray diffraction measurement in high magnetic fields revealed that the propagation vector changes from incommensurate (0 ≈ 0.27 1) to commensurate (0 1/4 1). Judging from the phase diagram of the $Gd_{1-x}Tb_xMnO_3$, the origin of the ferroelectric polarization along the *a* axis should be common to the high-field phase of $TbMnO_3$ and the zero-field phase of $Gd_{1-x}Tb_xMnO_3$ with $x \sim 0.3$. As a full analysis of the magnetic structure in a high magnetic field is not easy, we investigated the magnetic structure of $Gd_{1-x}Tb_xMnO_3$.

A single crystal of $^{160}Gd_{0.7}Tb_{0.3}MnO_3$ was grown by a floating zone method. Intensity data of magnetic reflections were collected at 15 K by using a four-circle neutron diffractometer FONDER. The wavelength of 1.24Å was used for the measurement. The absorption of the neutron beam by the crystal was 5.80/cm. Spin polarized neutron diffraction measurements were performed with a triple-axis spectrometer PONTA. The intensity of a magnetic reflection (0 1/4 3) were investigated for the incident neutron spin parallel and antiparallel to the scattering vector. In this configuration, the *c* component of the spin vector chirality should cause a difference in the intensity.

Figure 1 shows the profiles of magnetic satellites at Q_s = (0 1/4 3) for the ferroelectric state at 20 K after cooling in an electric field. The intensity clearly depends on the neutron spin direction. This result evidently indicates the chiral spin structure in $Gd_{0.7}Tb_{0.3}MnO_3$. The magnetically induced polarization flop in $TbMnO_3$ is ascribed to the flop of spiral plane around the *b* axis.

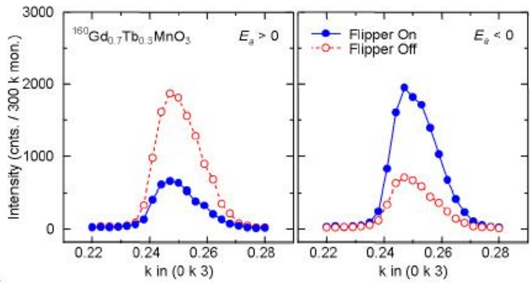

Fig. 1. Profiles of a magnetic reflection (0 1/4 3) in $^{160}Gd_{0.7}Tb_{0.3}MnO_3$ with the incident neutron spin polarized (open red circles) parallel and (solid blue circles) perpendicular to the scattering vector.