Control of the polarization flop direction in multiferroic \( R \text{MnO}_3 \) (\( R = \text{Tb}, \text{Dy} \)) by a tilted magnetic field

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Abstract. In spin-driven ferroelectrics, the electric polarization is often modified by the application of a magnetic field. \( R \text{MnO}_3 \) (\( R = \text{Tb}, \text{Dy} \)) is a well known multiferroic with electric polarization along the \( c \)-axis induced by the cycloidal ordering of Mn spin moments in the \( bc \) plane, which can be explained by the inverse effect of Dzyaloshinskii-Moriya interaction. \( R \text{MnO}_3 \) exhibits an electric-polarization flop from \( P//c \) to \( P//a \) originating from a rotation of the cycloidal plane from the \( bc \) to \( ab \) plane by a magnetic field applied along the \( b \)-axis. Here we show that the polarization flop direction in \( R \text{MnO}_3 \) of +90 or -90 degrees can be selected by changing the tilting direction of the applied magnetic field from the \( b \)-axis.

1. Introduction

Since a 90-degree rotation of electric polarization (\( P \)) by the application of a magnetic field (\( H \)) in \( \text{ThMnO}_3 \) was discovered by Kimura et al.[1], research on magnetoelectric multiferroics has become active in both physics and technology. Ferroelectricity which is driven by the cycloidal ordering of magnetic moments is explained by the inverse effect of the Dzyaloshinsky-Moriya interaction[2,3,4,5] expressed by \( P = A e_{ij} \times C_{ij} \), where \( C_{ij} = S_i \times S_j \) is often referred to as vector spin chirality, because \( C_{ij} \) can have a uniform component \( C \) in cycloidal magnets. Such cycloidal magnetic ordering arises from the competition between the nearest-neighbor ferromagnetic interaction and next nearest neighbor antiferromagnetic interactions. The application of a relatively weak magnetic field can hence modify the magnetic alignment. In \( \text{ThMnO}_3 \) and \( \text{DyMnO}_3 \), the electric polarization flops from the \( c \)-axis to the \( a \)-axis by the application of a magnetic field along the \( b \)-axis[1,6]. The mechanism of the magnetic-field induced \( P \) flop is represented by a 90-degree rotation of cycloidal ordering of Mn spin moments[7,8]. Upon the \( P \) flop transition, \( C \) may rotate by either +90 or -90 degrees. As a consequence, multi-domain state would appear. However, \( \text{ThMnO}_3 \) shows a selective rotation of \( P \) by rotating the magnetic field direction around \( ab \)-plane[9]. In other words, the rotation direction of \( C \) is determined by the magnetic field direction without the formation of multi-domain state. To investigate more intensively the selective rotation of \( P \), and hence \( C \), in \( \text{ThMnO}_3 \), we have performed a systematic measurement of \( P \) with rotating the magnetic-field direction in the multiferroic \( R \text{MnO}_3 \) system with \( R = \text{Gd}, \text{Tb} \) and \( \text{Dy} \). We have also investigated the rotating direction of \( P \) in the vicinity of \( P \) flop transition in \( \text{ThMnO}_3 \) in a magnetic field tilted from the \( b \)-axis.
2. Experimental

Single crystals of orthorhombic $RMnO_3$ ($R =$ Gd, Tb, Dy) were grown by a floating-zone method. To monitor the $P$ flop transition, thin-plate samples were prepared. The widest surface of the samples was perpendicular to the $a$-axis or to a normal vector forming an angle of 45 degrees both from the $a$- and $c$-axes in the $ac$ plane. In the latter configuration, the measured value of $P$ corresponds to $P_n = (P_a + P_c)/2$. Gold electrodes were sputtered onto the opposite faces of each sample to measure the electric polarization $P$. We obtained the $P$ value from the integration of the pyroelectric current, which was measured with an electrometer (KEITHLEY 6517A). Prior to each $P$ measurement, the sample was cooled to a ferroelectric phase in a poling electric field (typically 300 kV/m) to form a single-domain state. The direction of a magnetic field was varied by rotating the sample in a cryostat equipped with a 15-tesla superconducting magnet. The measurements of $P$ in a magnetic field were performed at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University, Japan.
3. Results and discussions

To uncover the role of the canted magnetic field (direction), we measured $P_a$ with rotating the magnetic field direction in $R\text{MnO}_3$ ($R=\text{Gd, Tb, Dy}$), as shown in Fig. 1. In the cases of $\text{TbMnO}_3$ and $\text{DyMnO}_3$, the sign of $P_a$ is reversed with retaining the single domain state after a 180-degree rotation of the magnetic-field direction. On the other hand, electric polarization is almost vanished after the magnetic field rotation in $\text{GdMnO}_3$, likely attributed to multi-domain ferroelectric state. These results indicate that the $P//a$-to-$P//c$ transition has an essential role for the electric-polarization reversal in a magnetic field rotation process in $R\text{MnO}_3$.

The magnetoelectric phase diagrams obtained by $P_a$ measurements with magnetic-field rotation are shown in Fig. 2. The $P//a$ phase appears for all the compounds in a relatively weak magnetic field when the field direction is close to the $b$-axis, while the detail shape of the area of the $P//a$ phase depends on the rare earth species. These phase diagrams imply that the induction of the $P//a$ phase is dominated by the interaction between rare earth 4f moments and Mn$^{3+}$ 3d moment. In $\text{TbMnO}_3$, for example, 4f moments of Tb$^{3+}$ behave as Ising spins along a direction canted by $+57$ or $-57$ degrees from the $b$-axis in the $ab$-plane[10]. The antiferromagnetic ordering is robust against a magnetic field applied perpendicular to one of the Ising axes. Rearrangement of Tb moments possibly requires a higher magnetic field for these directions.

Finally, we focus on the mechanism of the polarization reversal in $\text{TbMnO}_3$. Mostovoy predicts that the $C$ vector trends to orient along the magnetic field direction in typical cycloidal spin systems[11]. In the $\text{TbMnO}_3$ case, the $P$ flop transition occurs by the application of a magnetic field along the $b$-axis. However, the vector spin chiralities in both the $P//c$ phase and the $P//a$ phase are perpendicular to the magnetic-field direction. Because of the first-order nature, the $P//a$ phase and $P//c$ phase can coexist in the vicinity of the $P$ flop transition. Two types of domains are separated by domain walls. A magnetic field tilted from the $b$-axis as illustrated in Fig. 3(a) may lift the degeneracy in energies of domain walls of various types. When a magnetic field of 6T changes in direction from $\theta = 5^\circ$ to $\theta = 35^\circ$ with $\omega = 7^\circ$, the electric polarization flops from $+P_a$ to $+P_c$, as shown in Fig. 3 (b). On the other hand, when the $H$ direction is swept from $\theta = 5^\circ$ to $\theta = -35^\circ$, the electric polarization flops from $+P_a$ to $-P_c$. These selective rotations of $P$ cannot be explained by the uniform rotation of the cycloidal magnetic structure, but the selectivity of domain wall[12].

4. Conclusion

In conclusion, we measured $P_a$ with the rotating the magnetic field direction in $R\text{MnO}_3$ system. The selective rotation of electric polarization direction in the tilted magnetic field was discovered in $\text{TbMnO}_3$ and $\text{DyMnO}_3$ but not in $\text{GdMnO}_3$. The $P$ flop direction is dominated by the selectivity of domain wall in the magnetic field.

References

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Figure 3. Measurement of the \( P \) flop direction in the vicinity of the \( P \) flop transition in TbMnO\(_3\). (a) The tilted direction of magnetic field of 6T from the \( b \)-axis is expressed by \( \omega \) and \( \theta \). (b) and (c) Change of \( P_n \) from the \( P//a \) phase (\( \theta = +5 \) degrees) to \( P//c \) phase (\( \theta = +35 \) degrees or -35 degrees) with keeping \( \omega = +7 \) degrees.

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