Magnetic-field control of electric polarization in a helimagnetic hexaferrite \( \text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22} \)

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Abstract. A Y-type hexaferrite \( \text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22} \) undergoes a transition to a proper-screw type helimagnetic structure with a propagation vector \( k_0 \parallel [001] \) at 195 K, below which the system shows field-induced successive transitions under magnetic fields up to 1 T. Magnetization measurements also indicate the presence of the conical spin structure at low magnetic fields (~10 mT) below about 50 K. We report on the magnetic control of the electric polarization in \( \text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22} \) with using the linearly oscillating fields up to \( \pm 1 \) T at 5 K. The polarization vector can be cyclically reversed by weak magnetic fields of \( \pm 30 \) mT. In addition, the polarization vector is repeatedly reversed without significant decay even by applying fields of \( \pm 1 \) T, which suggests that the sense of the spin helix is somehow conserved in the process of the field-induced phase transitions. We propose that the conical spin structure carries the polarization vector upon the reversal of magnetic field.

1. Introduction
A new class of multiferroic materials, characterized by a helimagnetic order giving rise to ferroelectricity, has been of great interest, since the discovery of a giant magnetoelectric effect in \( \text{TbMnO}_3 \) [1, 2, 3, 4, 5]. These materials often display a rich variety of magnetic phases under magnetic fields \( B \) up to several tesla, reflecting the presence of competing magnetic interactions, which enables a drastic change of the polarization. A Y-type hexaferrite \( \text{Ba}_{0.5}\text{Sr}_{1.5}\text{Zn}_2\text{Fe}_{12}\text{O}_{22} \) is also the one belonging to this class of materials [6]. While the ordering temperatures of such kinds of phases are typically confined within several tens of kelvin, the helimagnetic ordering temperature of \( \text{Ba}_{0.5}\text{Sr}_{1.5}\text{Zn}_2\text{Fe}_{12}\text{O}_{22} \) is exceptionally high as room temperature or even higher, though the ferroelectric polarization disappears below 0.25 T.

Recently we have found that the isostructural hexaferrite \( \text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22} \) shows magnetically induced ferroelectric polarization \( P \) at much lower \( B \) and succeeded in flexibly controlling the \( P \) vector by using rotating \( B \) of a few tens of mT [7, 8]. Both hexaferrites are helimagnetic with a propagation vector \( k_0 \) parallel to [001] at low temperatures [9, 10, 11]. When subjected to magnetic fields \( B \) perpendicular to \( k_0 \) in the helimagnetic states, they show field-induced successive transitions below 1 T. The phases at around 1 T in \( \text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22} \) as well as \( \text{Ba}_{0.5}\text{Sr}_{1.5}\text{Zn}_2\text{Fe}_{12}\text{O}_{22} \) have ferroelectric \( P \) perpendicular to both \( B \) and \( k_0 \), which can be rotated around the hexagonal axis [001] by the rotation of \( B \) around the same axis [6, 7, 8]. The points that differentiate \( \text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22} \) from \( \text{Ba}_{0.5}\text{Sr}_{1.5}\text{Zn}_2\text{Fe}_{12}\text{O}_{22} \) are the presence of a longitudinal...
conical spin state (Fig. 1(a)) below 50 K and the low-field-induced ferroelectric phases which have presumably transverse conical spin structures (Fig. 1(b)) as observed in a spinel CoCr$_2$O$_4$ [4, 7].

According to the spin-current model [12], the non-collinear arrangement of the adjacent spins can produce the local polarization, which reads as $p_{i,j} = A e_{i,j} \times (S_i \times S_j)$ ($A$ is a scalar determined by the exchange interaction and the spin-orbit interaction. The spins $S_i$ and $S_j$ reside on the adjacent sites along the unit vector $e_{i,j} (= k_0 / |k_0|)$). Thus, the longitudinal conical spin state can produce a local polarization $p_{i,j}$ but it is canceled out in a macroscopic scale. In order to produce the ferroelectric polarization predicted by this model, the cone axis (screw axis) should be deviated from $k_0$ such as the transverse conical spin structure mentioned above [4, 13, 14].

In this paper, we show a cyclic reversal of $P$ in Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$ with using the linearly oscillating fields up to $\pm 1$ T. The presence of conical spin structures at low temperatures as evidenced by the magnetization data plays a key role in carrying the ferroelectric polarization vector without significant decay.

2. Experimental

Single crystals of Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$ were grown from Na$_2$O-Fe$_2$O$_3$ flux in an ambient atmosphere as described in [11]. For measurements of the electric polarization ($P$) along [120], gold electrodes were deposited on the both faces ($2.7 \times 1.6$ mm$^2$ for the electrodes and 0.52 mm in thickness along the electric field direction) of the rectangularly shaped crystals. For the poling procedure to fix the sense of the spin helix, the electric field, $E = 200$ V/mm, was applied at 60 K, and an external magnetic field ($B$ ) was applied parallel to [100] (perpendicular to both $E$ and [001]) as 0 T $\rightarrow$ 3 T $\rightarrow$ 1 T so that the sample might traverse the paraelectric-ferroelectric phase boundary, followed by cooling down to 5 K at 1 T. Then, the poling $E$ was removed to measure the displacement current with changing $B$ at a rate of $1 \sim 8$ mT/s. The $P$ was deduced by integration of the displacement current as a function of time. DC magnetic susceptibility was measured with a MPMS SQUID magnetometer (Quantum Design) in an external magnetic field of 100 Oe.

3. Result and discussions

Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$ belongs to the family of Y-type hexaferrites having the centric space group $R3m$. As shown in the inset of Fig. 2, the single crystals have a hexagonal morphology, reflecting the crystallographic symmetry ($5 \text{ mm} \times 5 \text{ mm} \times 2.5 \text{ mm}$ in dimension, typically). Figure 2 shows temperature dependence of the magnetizations divided by the applied field ($M/H$) for [100] and
Figure 2. Temperature dependence of magnetizations divided by field of Ba$_2$Mg$_2$Fe$_{12}$O$_{22}$, measured on cooling with a magnetic field of 100 Oe along [100] and [001]. The single crystal with the corresponding crystallographic directions is shown as an inset.

Figure 3. Reversal of the ferroelectric polarization at 5 K (left axis) by linearly oscillating magnetic field $B$ (right axis) of $\pm 30$ mT, $\pm 0.1$ T, and $\pm 1$ T, plotted as a function of time.

[001]. Both of them show anomalies at around 195 K and 50 K. As reported previously [11], the anomaly at 195 K corresponds to a transition from a collinear ferrimagnetic to a proper-screw type helimagnetic phase with $k_0$ along [001]. Because spins are lying within the easy magnetization plane (001) in both phases, this transition should cause a significant drop for $M/H$ along [100]. On the other hand, the anomaly at 50 K, especially for the $M/H$ along [001] showing gradual increase by lowering temperature, can be interpreted as a change of the magnetocrystalline anisotropy, which causes spin reorientation to make the system a conical helimagnet. We call this spin structure a longitudinal conical spin structure with the cone axis
parallel to \( k_0 \) (see Fig. 1(a)).

Next, we show the magnetic control of \( P \) in \( \text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22} \) by the oscillating \( B \) applied along [100] at 5 K (see Fig. 3). Under the cyclically oscillating \( B \) of \( \pm 30 \) mT, \( P \) oscillates with changing its sign and keeping almost the same amplitude. Given that the transverse conical spin structure is realized under magnetic fields of \( \pm 30 \) mT, this behavior can be explained by the conservation of the spin helicity (defined as \( \Sigma k_0 \cdot (S_i \times S_j) \)) through the longitudinal conical state at \( B = 0 \). When \( B \) is cyclically reversed with increasing the amplitude as \( \pm 30 \) mT, \( \pm 0.1 \) T, and \( \pm 1 \) T, \( P \) coincidentally oscillates with increasing the amplitude. The conservation of \( P \) by a cyclic change of \( B \) between -1 T and 1 T suggests that the sense of the spin helix is somehow conserved in the process of the field-induced phase transitions. The \( P \) drastically increases as \( B \) traverses around 0.1 T, where the change of the propagation vector is likely to occur, and is saturated at about 0.4 T. Neutron diffraction studies to clarify the relation between \( P \) and the conical spin arrangements under magnetic fields are worth future consideration.

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4. References

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