



Soft x-ray resonant diffraction study of magnetic structure in magnetoelectric Y-type hexaferrite

H. Ueda^{a,*}, Y. Tanaka^b, Y. Wakabayashi^a, T. Kimura^c

^a Division of Materials Physics, Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan

^b RIKEN SPring-8 Center, Sayo, Hyogo 679-5148, Japan

^c Department of Advanced Materials Science, University of Tokyo, Kashiwa, Chiba 277-8561, Japan

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ABSTRACT

The effect of magnetic field on the magnetic structure associated with magnetoelectric properties in a Y-type hexaferrite, $\text{Ba}_{1.3}\text{Sr}_{0.7}\text{CoZnFe}_{11}\text{AlO}_{22}$, was investigated by utilizing the soft x-ray resonant diffraction technique. In this hexaferrite, the so-called alternating longitudinal conical phase is stabilized at room temperature and zero magnetic field. Below room temperature, however, this phase is transformed into the so-called transverse conical phase by applying an in-plane magnetic field (≈ 0.3 T). The transverse conical phase persists even after removing the magnetic field. The magnetoelectricity, which is magnetically-induced electric polarization, observed in the hexaferrite is discussed in terms of the temperature-dependent magnetic structure at zero field.

1. Introduction

Magnetoelectric (ME) multiferroics, in which ferroelectric and magnetic orders develop simultaneously and closely couple to each other, have been extensively explored since the discovery of ferroelectricity induced by a cycloidal magnetic order in TbMnO_3 [1]. Because they enable us to mutually control their dielectricity and magnetism via the ME coupling, they are one of the promising functional material groups for practical applications such as magnetic storage devices with low energy consumption. However, most of them operate only far below room temperature due to magnetic frustration: one of the key factors for the realization of non-trivial magnetic orders which give rise to the close ME coupling. Among various ME multiferroics studied to date, hexaferrites, which are classified into several types by their chemical formulae and crystal structures [2–4], are one of the rare ME material families operating at room temperature. Therefore, a lot of studies on the ME hexaferrites have been carried out up-to-date both from scientific and technological points of view [5–13].

The magnetoelectricity in this family was firstly reported in the Y-type hexaferrite, $\text{Ba}_{0.5}\text{Sr}_{1.5}\text{Zn}_2\text{Fe}_{12}\text{O}_{22}$ [6]. Y-type hexaferrites possess $A_2\text{Me}_2\text{Fe}_{12}\text{O}_{22}$ chemical formula and belong to $R\bar{3}m$ space group. Here, A and Me denote an alkaline earth metal ion and a divalent metal ion, respectively. Their crystal structures consist of alternately stacked large (L) and small (S) spin blocks along the crystallographic c -axis in a hexagonal setting as shown in Fig. 1(a). In this paper, we employ a conventional hexagonal setting and represent magnetic structures

assuming that one block has one magnetic moment as a whole. Owing to the inter-block magnetic frustration, which can be tuned by an appropriate chemical substitution [14], some of the Y-type hexaferrites show helimagnetically ordered phase with the incommensurate propagation vector $\mathbf{k}_1 = (0, 0, \delta)$ at zero magnetic field [15,16]. Here, δ is a non-integer. Their magnetoelectricity is ascribed to a metamagnetic transition into the so-called transverse conical (TC) phase with the commensurate propagation vector, e. g., $\mathbf{k}_2 = (0, 0, 1.5)$, as illustrated in Fig. 1(c) [17]. Finite electric polarization induced by the TC structure, which is composed of collinear ferrimagnetic and non-collinear cycloidal components, is well explained by the so-called spin-current mechanism [18]. Thus, in the TC phase, macroscopic magnetization (M) and electric polarization (P) lie within the basal plane but are perpendicular to each other.

Recently, we reported that a Y-type hexaferrite, $\text{Ba}_{1.3}\text{Sr}_{0.7}\text{CoZnFe}_{11}\text{AlO}_{22}$, shows the so-called alternating longitudinal conical (ALC) structure at room-temperature and zero-field [19]. This structure consists of a helical component lying in the basal plane and a collinear $\uparrow\uparrow\downarrow\downarrow$ type antiferromagnetic component as shown in Fig. 1(b). The latter component can induce ferroelectric polarization along the c -axis not through the inverse Dzyaloshinskii-Moriya interaction as in the TC phase but through the magnetostriction as reported in a similar compound, $\text{Ba}_{0.3}\text{Sr}_{1.7}\text{Co}_2\text{Fe}_{11}\text{AlO}_{22}$ [20]. In this study, we compare the zero-field magnetic structures at 300 K and 150 K in the Y-type hexaferrite and reveal that the difference is essential for ME characteristics at the respective temperatures. To clarify the magnetic structures,

* Corresponding author.

E-mail address: ueda@crystal.mp.es.osaka-u.ac.jp (H. Ueda).

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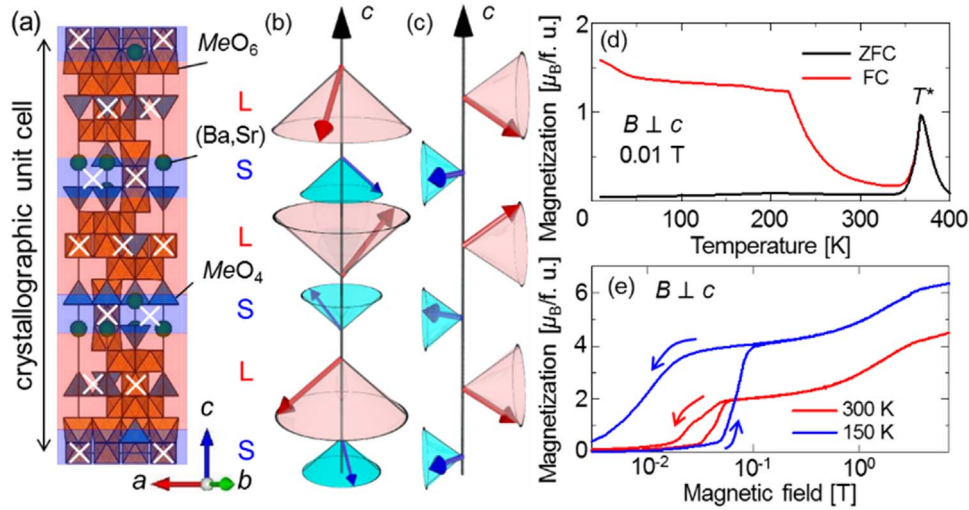


Fig. 1. (a) Crystallographic structure of $\text{Ba}_{1.3}\text{Sr}_{0.7}\text{CoZnFe}_{11}\text{AlO}_{22}$, composed of alternate stacks of large (L) and small (S) spin blocks along the hexagonal c -axis. Here, $\text{Me} = \text{Fe}, \text{Al}, \text{Co}$, and Zn . White crosses indicate the inversion center. This illustration was drawn by VESTA [25]. (b) Alternating longitudinal conical and (c) transverse conical magnetic structures. Red and blue arrows represent net magnetic moments of the L and S blocks, respectively. (d) Temperature dependence of in-plane magnetization taken in heating process and 0.01 T for a single crystal of $\text{Ba}_{1.3}\text{Sr}_{0.7}\text{CoZnFe}_{11}\text{AlO}_{22}$. ZFC (FC) denotes the data obtained after zero-field cooling (field cooling at 0.1 T) from 400 K to 10 K. (e) Isothermal in-plane magnetization curves obtained at 300 K and 150 K after ZFC. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

we utilized the resonant soft x-ray diffraction technique with highly-focused x-ray beam ($\approx 30 \times 15 \mu\text{m}^2$ in area). Our study revealed that the change of the magnetic structure affects the ME properties of this compound.

2. Experimental procedure

Soft x-ray resonant diffraction measurements were carried out at the beamline 17SU, SPring-8, Japan. The single crystal of $\text{Ba}_{1.3}\text{Sr}_{0.7}\text{CoZnFe}_{11}\text{AlO}_{22}$ and the experimental setup used for the measurements are the same with those of Ref. 19. The photon energy of the incident soft x-rays is fixed at Fe L_3 edge (= 710 eV). The penetration depth ξ of the incident x-rays into the specimen was estimated to be ~ 60 nm at Fe L_3 edge from the full width at half maximum ΔQ [nm^{-1}] of the (0 0 3) Bragg reflection by using the relation $\xi = 2\pi/\Delta Q$. Note that all the diffraction data shown in this paper were taken in the absence of a magnetic field (B). Single crystals used for dielectric measurements were annealed under 100 atm diluted O_2 atmosphere (20% $\text{O}_2 + \text{Ar}$) by using a hot isostatic pressing device (KOBELCO, O_2 -Dr. HIP) to reduce electrical conductivity of the crystals. Measurements of M were carried out in B perpendicular to the c -axis by using a superconducting quantum interference device magnetometer (Quantum Design, MPMS3). Before each measurement of M , zero-field-cooling (ZFC) or field-cooling (FC) with 0.1 T procedure was carried out from 400 K to target temperature: 400 K is higher enough than the magnetic transition temperature (≈ 366 K) as shown in the later section. Dielectric constant (ϵ) was measured with a frequency of 10 kHz by using an LCR meter (Agilent E4980A), and P was obtained by integrating ME currents measured by an electrometer (Keithley, 6517A). Silver electrodes were vacuum deposited onto the largest faces perpendicular to the crystallographic [100] direction. An in-plane B parallel to the electrodes was applied for the dielectric measurements by a superconducting magnet (Quantum Design, PPMS). Prior to ME current measurements, the specimen was poled by the following procedure at a constant temperature. At first, an in-plane magnetic field ($|B| = 7$ T) was applied to get into the paraelectric collinear-ferrimagnetic phase. Then, an electric field ($|E| = 500$ kV/m) was applied perpendicular to both B and the c -axis, and B was swept to the ferroelectric TC phase. After that, E was switched off. Due to the low resistivity around room temperature, all the data of ME properties above 200 K shown in this paper were obtained after the above-

mentioned ME poling procedure at 200 K where the resistivity is high enough to align the ferroelectric domains.

3. Results and discussion

Fig. 1(d) displays the temperature (T) dependence of in-plane M in $B = 0.01$ T. There is an anomaly with a sharp peak at $T^* \approx 366$ K, suggesting a magnetic transition at T^* . In the data obtained after the FC procedure, M clearly develops toward lower temperatures and shows a cusp singularity, indicating another phase transition, approximately at 220 K. A similar behavior was reported in $\text{Ba}_{0.3}\text{Sr}_{1.7}\text{Co}_2\text{Fe}_{11}\text{AlO}_{22}$ and ascribed to the phase transition from the ALC phase to the TC one while the ALC–TC mixed phase exists in the temperature region just above the cusp singularity [20]. Thus, it is plausible that the FC procedure stabilizes the TC phase at the lower T region ($\leq \sim 220$ K). In contrast, there is no anomaly except at T^* in the data taken after the ZFC procedure, suggesting that the ground-state magnetic structure after ZFC is different from that after FC. This implies that the ALC structure stabilized at room-temperature and zero-field persists as a metastable state even at 10 K after the ZFC procedure [21]. Fig. 1(e) shows the isothermal in-plane M curves obtained at 300 K and 150 K after ZFC. There are a noticeable hysteresis and remanent magnetization at 150 K although the virgin M curve shows metamagnetic transitions similar to the data taken at 300 K where the ALC structure is stabilized at zero field. Thus, we expect that the low- T ALC structure stabilized by ZFC is transformed into a different magnetic structure with spontaneous M by applying B , that is possibly the TC structure, which sustains even after B is eliminated.

Fig. 2 shows the results of the resonant soft x-ray diffraction measurements. The reflection condition of the $R\bar{3}m$ space group along the c -axis is (0 0 $3n$) where n is an integer. As seen in Fig. 2(a), there are two types of magnetic Bragg reflections at 300 K, the incommensurate (0 0 $3 \pm \delta$) with $\delta \approx 0.9$ and the commensurate (0 0 3 ± 1.5), in addition to the (0 0 3) fundamental Bragg reflection. This result is consistent with the existence of the ALC structure characterized by two propagation vectors, $\mathbf{k}_1 = (0, 0, \delta)$ and $\mathbf{k}_2 = (0, 0, 1.5)$. With decreasing T down to 150 K without any B , δ slightly shifts to ~ 0.7 and the intensities of the magnetic reflections increase. The enlarged views of each reflection obtained by both right- and left-handed circularly polarized (RCP and LCP) incident x-rays at 150 K are displayed in

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