

Magnetolectric domains and their switching mechanism in a *Y*-type hexaferriteF. P. Chmiel^{1,*}, D. Prabhakaran,¹ P. Steadman,² J. Chen,¹ R. Fan,² R. D. Johnson,¹ and P. G. Radaelli^{1,†}¹*Clarendon Laboratory, Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom*²*Diamond Light Source, Harwell Science and Innovation Campus, Didcot OX11 0DE, United Kingdom*

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By employing resonant x-ray microdiffraction, we image the magnetization and magnetic polarity domains of the *Y*-type hexaferrite $\text{Ba}_{0.5}\text{Sr}_{1.5}\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$. We show that the magnetic polarity domain structure can be controlled by both magnetic and electric fields and that full inversion of these domains can be achieved simply by reversal of an applied magnetic field in the absence of an electric field bias. Furthermore, we demonstrate that the diffraction intensity measured in different x-ray polarization channels cannot be reproduced by the accepted model for the polar magnetic structure, known as the 2-fan transverse conical (TC) model. We propose a modification to this model, which achieves good quantitative agreement with all of our data. We show that the deviations from the TC model are large and may be the result of an internal magnetic chirality, most likely inherited from the parent helical (nonpolar) phase.

DOI: [10.1103/PhysRevB.100.104411](https://doi.org/10.1103/PhysRevB.100.104411)**I. INTRODUCTION**

Electrical control of magnetic domain patterns is of current interest because of its technological importance in several applications [1]. Domain control has been demonstrated in the archetypal magnetolectric Cr_2O_3 , where the collinear antiferromagnetic ordering pattern can be set by the simultaneous application of magnetic and electric fields [2], but the technological impact of this material is limited by its small magnetolectric coefficient [3]. A viable alternative to Cr_2O_3 is represented by multiferroics with noncollinear magnetic order, where magnetically induced electric polarization induces strong coupling between the electric and magnetic orders and facilitates domain control. Among the candidate multiferroics, a class of materials known as the *Y*-type hexaferrites has emerged as one of the most promising, because of their high magnetic ordering temperatures, large magnetolectric coefficients, and the demonstrable coupling between the magnetic and polar orders [4–6].

The *Y*-type hexaferrites exhibit complex magnetic phase diagrams as a function of temperature and magnetic field [4]. In a specific range of fields and temperatures, a sizable electrical polarization is induced by the spin-current mechanism and is coupled with a large magnetolectric coefficient [7]. Notably, $\text{Ba}_{0.5}\text{Sr}_{1.5}\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ (BSMFO) possesses the largest known magnetolectric coefficient of any material: its sizable electric polarization can be switched, without any loss in magnitude, by small magnetic fields [5]. In other *Y*-type hexaferrites, magnetolectric effects have even been demonstrated at room temperature [8]. In spite of these desirable properties and after more than a decade of intense research, the precise nature of the magnetolectric domain switching mechanism, as well as the microscopic details of the polar magnetic structures, are yet to be determined.

In this paper, we present the results of a resonant x-ray diffraction (RXD) experiment designed to study the magnetolectric domain switching in BSMFO. Historically, resonant x-ray microdiffraction (spatially resolved diffraction) has been employed to determine the real-space distribution of chirality domains, such as those of the zero-field, proper-screw phases of the *Y*-type hexaferrites, which are not magnetolectric [9–11]. Here, we demonstrate that this technique is also sensitive to the magnetic polarity of the field-induced polar phases, even in the absence of net chirality. Using this technique, we spatially resolve the magnetolectric domain configuration of BSMFO. We show that a reversal of the applied magnetic field results in reversal of the magnetic polarity, which is directly coupled to the electrical polarization. We also show that the domain boundaries in the system are toroidal in nature, are stable upon magnetic field reversal, and can only be removed by simultaneous application of electric and magnetic fields. Polarization analysis of the scattered x rays with linear incident polarization, performed dynamically during the magnetolectric switching process, shows that the magnetolectric switching occurs by collective rotation of the magnetic structure in spin space around the high-symmetry axis. Moreover, the presence of an asymmetry between opposite directions of the magnetic field in the $\sigma\pi'$ and $\pi\sigma'$ channels incontrovertibly demonstrates that the magnetolectric phase cannot be a simple ‘transverse conical’ structure, as previously proposed [5], and that the deviation from this structure required to explain the data quantitatively must be large. We construct an explicit modification of the transverse conical structure, which fits all of our data well. Based on this model, we speculate that the magnetolectric phase could inherit internal chirality and a small ferrimagnetic *z*-axis component from the ‘parent’ nonpolar helical/longitudinal conical phase.

A. The magnetic structure of $\text{Ba}_{0.5}\text{Sr}_{1.5}\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$

$\text{Ba}_{0.5}\text{Sr}_{1.5}\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ crystallizes in space group $R\bar{3}m$ with lattice parameters $a = b \approx 5.8 \text{ \AA}$ and $c \approx 43 \text{ \AA}$, having

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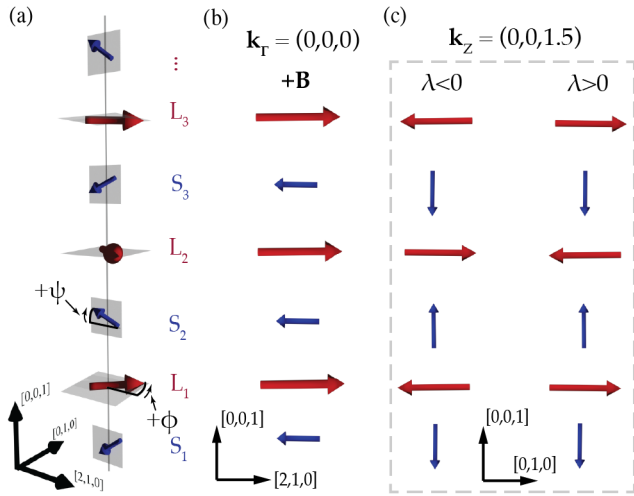


FIG. 1. (a) 2-fan transverse conical phase (TC) magnetic structure within the block model. Red and blue arrows represent L and S block moments, respectively, ϕ and ψ indicate the respective fan angles of the blocks. The applied magnetic field is parallel to the mean direction of the L blocks. (b),(c) Orthographic projection of the magnetic structure into two orthogonal planes, showing the TC structure can be considered as a linear combination of a ferrimagnetic component (b) and a commensurate cycloidal component (c), with respective propagation vectors of $\mathbf{k}_r = (0, 0, 0)$ and $\mathbf{k}_z = (0, 0, 1.5)$. Two possible magnetic polarity domains, arising because of the cycloidal component of the structure, are displayed in (c).

six symmetry-inequivalent magnetic Fe sites in the primitive unit cell. Most of BSMFO's magnetic phases are complex and noncollinear, and an unconstrained magnetic structural solution of all but the simplest structures has thus far proven elusive even for single-crystal neutron diffraction [12,13]. Therefore, magnetic structures are almost universally described by an approximate model, known as the *block model* [4]. In this approximation, the unit cell is divided into two magnetic blocks, L (large moment) and S (small moment), which are sequentially stacked along c . Within each magnetic block, Fe moments are assumed to be collinear and ferrimagnetic, by virtue of strong nearest-neighbor superexchange [14]. The magnetic structure is then described by a set of ‘superspins,’ located at the center of each block, each accounting for the net moment of the block. The net magnetic moments between neighboring blocks can be collinear, yielding a globally ferrimagnetic phase, or noncollinear, giving rise to a plethora of helical and conical phases [4]. In BSMFO, the net moments of the L and S blocks are $3m_{\text{Fe}}$ and $1m_{\text{Fe}}$, respectively [15], where m_{Fe} is the average moment magnitude of a single Fe site. In the first part of this paper, we will use the block model description of the magnetic structures (see Fig. 1, where L and S block moments are represented by red and blue arrows, respectively). Later, we will demonstrate that the ‘standard’ block model fails to describe some of our RXD data and discuss how the model needs to be modified on account of this.

At high temperatures, BSMFO is ferrimagnetically ordered, as discussed above, with antiparallel magnetizations between the L and S blocks. At ~ 390 K, the system undergoes

a transition to an incommensurate *planar* helical (proper screw) structure, in which neighboring L and S superspins are aligned approximately antiparallel to one another, and coherently rotate in the ab plane on traversing the c axis [16]. Upon further cooling, the blocks are reported to cant out of the basal plane to form what is known as the longitudinal conical (LC) structure, which was previously characterized in the Sr-free variant, $\text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ [17]. Application of small (~ 100 mT) in-plane magnetic fields in either of these noncollinear phases induces a series of ‘fan’ phases [12], in which the blocks are splayed in the ab plane around the direction of the net magnetization. In BSMFO and other hexaferrites, the periodic in-plane fan modulation is believed to be coupled with a modulation of the out-of-plane c component, forming the so-called 2-fan/transverse conical (TC) structure [4,7]. The TC phase is electrically polar, with its cycloidal component believed to be responsible for the induced electrical polarization through the spin current mechanism (see Sec. IB). The TC structure of relevance to this paper is stabilized below 100 K by application of a 150 mT magnetic field applied parallel to $[2,1,0]$ (a^*). It is described by two propagation vectors, $\mathbf{k}_r = (0, 0, 0)$ and $\mathbf{k}_z = (0, 0, 1.5)$ (see Appendix B 2 or Ref. [4] for details).

B. Magnetic polarity and electrical polarization

According to the spin-current mechanism, the electrical polarization of a noncollinear phase is given by [18]

$$\mathbf{P} \propto \hat{e}_{i,j} \times (\mathbf{S}_i \times \mathbf{S}_j), \quad (1)$$

where $S_{i,j}$ are neighboring spin blocks and $\hat{e}_{i,j}$ the unit vector joining them. In the TC phase the electric polarization is induced by the cycloidal component of the magnetic structure ($\mathbf{k}_z \parallel \mathbf{c}$). This results in an electrical polarization \mathbf{P} in the plane of rotation of the spins and perpendicular to both \mathbf{k}_z and \mathbf{B} [5], and hence to the magnetization \mathbf{M} . For the TC phase we can write the amplitude of \mathbf{M} as

$$m = m_L \cos \phi - m_S \cos \psi, \quad (2)$$

where m_i are the net moment magnitudes of the blocks and ϕ and ψ the block fan angles (see Fig. 1). Since \mathbf{P} and \mathbf{M} are orthogonal, the TC phase also possesses a *ferrotoroidal moment* \mathbf{T} , defined as:

$$\mathbf{T} = \mathbf{P} \times \mathbf{M}. \quad (3)$$

Here, the ferrotoroidal moment defines a specific type of magnetoelectric domain, which will be discussed in detail later.

A peculiar feature of BSMFO is that the sign of the electric polarization can be easily switched by reversing the sign of the magnetic field used to stabilize the TC phase [5,7]. From inspection of Eq. (1), one can see that reversal of the electric polarization could be accomplished by a global rotation of the magnetic structure in spin space around the axis parallel to $\hat{e}_{i,j}$. In the analysis that follows, it is useful to introduce a magnetic order parameter λ , known as the *magnetic polarity*, which provides a direct coupling between the magnetism and the electrical polarization of the form $\mathbf{P} = \chi \lambda$, where χ is a coupling constant [19]. λ is quadratic in the magnetic moments, parallel to \mathbf{P} , and in the TC phase its amplitude,

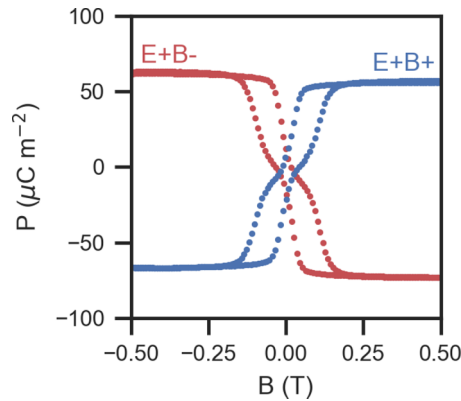


FIG. 2. Electric polarization of BSMFO as a function of applied magnetic field, following different magnetoelectric poling procedures. Measurements (color) were performed in the absence of an applied electric field at 10 K. Blue circles indicate measurement following a magnetoelectric poling with a E_+B_+ poling field product (see main text) and red circles with a E_+B_- field product. The magnitude of E_+ was 100 V mm^{-1} .

λ , can be written as

$$\lambda = m_L m_S \sin \phi \sin \psi. \quad (4)$$

Two possible magnetic polarity domains are displayed in Fig. 1(c). We have directly imaged the magnetic polarity of BSMFO by RXD and, by virtue of the linear relation between the magnetic polarity and electric polarization, also established the electric polarization domain configuration.

II. EXPERIMENTAL DETAILS

A. Crystal growth and characterization

Single crystals of $\text{Ba}_{0.5}\text{Sr}_{1.5}\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ were grown by the flux method [20]. Laboratory-based x-ray diffraction and magnetization measurements were performed (presented in the supplemental material [21]) to check the crystal quality used in our RXD experiment.

To investigate the polar properties of the sample, magnetocurrent measurements were performed using a custom probe inserted into a Quantum Design PPMS. The sample was mechanically polished into a cuboid and silver paste was used to make electrical contacts on the b faces, the $[2,1,0]$ direction (orthogonal to the b faces) was oriented parallel to the applied magnetic field. Samples were zero field cooled from room temperature to 10 K and then an electric field bias (of magnitude 100 V mm^{-1}) was applied to the sample. The magnetic field was increased to $+0.5 \text{ T}$ or -0.5 T , at which point the biasing electric field was removed. The magnetic field was then swept while measuring the displacement current, the integral of which yields the sample polarization as a function of the applied magnetic field.

By using this procedure, the electric polarization and its switching behavior were fully characterized at 10 K (Fig. 2). Initially, the sample was prepared into a single polar domain using a E_+B_+ ‘poling’ cross fields. After the removal of the electric field, the electric polarization (blue circles in Fig. 2) was measured as the magnetic field was swept from 0.5 T to -0.5 T and back. We observe an appreciable electric polar-

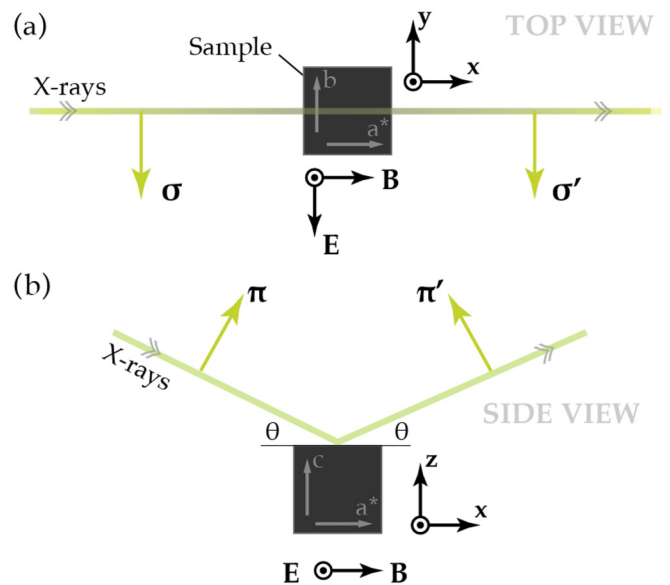


FIG. 3. Graphic demonstrating the geometry of the resonant x-ray diffraction experiment. (a) Orthographic projection down the crystallographic c axis. (b) Projection parallel to the b axis. The sample sits at the center of two pole pieces of an electromagnet, which can apply magnetic fields parallel to the scattering plane ($\parallel x$). Electric fields are applied orthogonal to the scattering plane ($\parallel y$). The basis ($\sigma - \pi$) of orthogonal linear polarization states are shown by solid yellow arrows and the Cartesian coordinate system used in our calculations is displayed by the inset axes. The crystallographic (hexagonal) basis is shown by the light gray arrows overlaid on the sample graphic.

ization [22] which, importantly, is coupled to the direction of the applied magnetic field. Consistent with previous studies on BMFO [7,23] we found the electric polarization to reverse in direction without any loss in magnitude, even in the absence of an electric field bias.

Secondly, we investigated the effect that different magnetoelectric poling fields had on our samples. The solid red points in Fig. 2 show the measured electric polarization after poling the sample with E_+B_- poling fields. In this situation, we see that the direction of the electric polarization is antiparallel to the E_+B_+ case, is of similar magnitude, and is coupled to the direction of the applied magnetic field in the same way. These observations confirm that our sample exhibits the magnetoelectric effects of interest.

B. Resonant x-ray diffraction: Experimental geometry

Resonant soft x-ray diffraction experiments were performed using the RASOR diffractometer at the I10 beamline located at the Diamond Light Source (Didcot, UK) [24]. The experimental geometry is shown in Fig. 3, which includes the Cartesian basis used throughout this paper. The sample was cleaved in ambient conditions, yielding smooth faces normal to c (z) and mounted such that c and a^* (x) were orientated in the scattering plane, with c in the specular direction. An electromagnet and a ^4He flow cryostat were used to apply magnetic fields between $\pm 300 \text{ mT}$ parallel to a^* at sample temperatures between 25 K and 300 K. Diffracted x rays were

measured with a photodiode and normalization of the signal was achieved by dividing by the incident beam intensity, recorded by a monitor upstream of the sample which measures the total electron yield from the focusing mirror. This corrects for variations in the incident intensity caused, for example, by the changes in the synchrotron ring current. The incident x-ray energy was set to 707.48 eV (the Fe L_3 absorption edge) for the duration of the experiment. At this energy, the x-ray attenuation length is approximately $0.5\mu\text{m}$ and the intensity of the incident x-ray beam rapidly decreases within the sample [25]. While this means our experiment is insensitive to a significant volume of the crystal, our calculations (displayed in the Supplemental Material [21]) show that we probe to a depth of tens of unit cells and our results, therefore, likely reflect the bulk properties of our crystal. When an *in situ* electric field was required, silver paste was used to make contacts on opposite faces of the sample, such that electric fields could be applied mutually perpendicular to the applied magnetic field and the c axis.

Diffraction measurements were performed along the $(0, 0, l)$ zone axis. In our experimental geometry, the Ewald sphere only contains the $(0,0,3)$ Bragg peak, with contributions from Thomson (nonmagnetic) scattering as well as magnetic scattering from the net magnetization of the sample, the two magnetic satellites $(0, 0, 3 \pm k_\Lambda)$ of the helical phase in zero field ($k_\Lambda \sim 0.6 - 0.8$), and the $(0,0,4.5)$ or $(0,0,1.5)$ purely magnetic peaks of the TC phase. The reciprocal space coordinates $(0, 0, 3 \pm 2k_\Lambda)$, which correspond to higher-order scattering processes, were also accessible (see below).

Two perpendicular, linear x-ray polarizations of the incident beam could be selected, denoted ϵ_σ and ϵ_π (Fig. 3), as well as circular positive (c^+) and circular left (c^-) polarizations. The linear polarization of the diffracted beam could be determined using a multilayer analyzer whose d spacing was selected to achieve scattering at Brewster's angle when using an incident x-ray energy of 707.48 eV. X-ray microdiffraction was achieved using a 50 mm diameter aperture placed 265 mm upstream of the sample.

III. RESONANT X-RAY SCATTERING CROSS SECTIONS

In this section, we summarize key equations for the magnetic scattering cross sections in the linear polarization channels and other important derived quantities such as circular dichroism. The equations are given in terms of the magnetic interaction vector, M_γ , and $F^{(0)}$ and $F^{(1)}$, the resonant scattering factors that are in general complex numbers [26]. We note that there exists an additional resonant scattering factor $F^{(2)}$, which for an incommensurate magnetic structure having propagation vector \mathbf{k} , gives rise to magnetic satellites at $Q = (003) \pm 2\mathbf{k}$, with an intensity proportional to $|F^{(2)}|^2$. For the helical phase of BSMFO, we measured an intensity ratio of $\leq 5\%$ between $2\mathbf{k}$ and \mathbf{k} satellites, indicating that the $|F^{(2)}|$ is small compared to $|F^{(0)}|$ and is at most $\sim 20\%$ of $|F^{(1)}|$ [26]. Nevertheless, for a commensurate structure, $F^{(2)}$ could provide a sizable contribution to the magnetostructural interference at the Γ point as well as $F^{(1)}-F^{(2)}$ interference, which we address in Sec. IV D. A complete summary of the theory of resonant magnetic scattering from which the

following equations are derived is provided in Appendix A, in which we only consider magnetic dipole contributions.

(i) The *circular dichroism* is defined as

$$\delta = \frac{I_+ - I_-}{I_+ + I_-} \quad (5)$$

where I_+ and I_- are the total scattered intensities with incident c^+ and c^- circular polarization and without any polarization analysis of the scattered beam.

(ii) The *circular asymmetry*, i.e., the asymmetry of the circular dichroism upon reversal of the magnetic field ($B+ \rightarrow B-$, assuming that the x and y components are reversed), defined as

$$\text{Asy}_\delta = \frac{\delta(B+) + \delta(B-)}{\delta(B+) - \delta(B-)} \quad (6)$$

(iii) The *linear asymmetries*, i.e., the asymmetry of the scattering in the linear channels upon reversal of the magnetic field, defined as:

$$\text{Asy}_{\epsilon\epsilon'} = \frac{I_{\epsilon\epsilon'}(B+) - I_{\epsilon\epsilon'}(B-)}{I_{\epsilon\epsilon'}(B+) + I_{\epsilon\epsilon'}(B-)} \quad (7)$$

As we shall see, the asymmetries are defined in such a way to be zero in the simplest versions of the magnetic structures, hence the opposite signs in Eqs. (6) and (7).

A. Structural-magnetic interference: Circular dichroism at the Γ point

For phases with a net magnetization, one would expect to observe circular dichroism at the Γ point [the (003) reflection in our case] since the Thomson and magnetic contributions interfere at this position [27]. For the simplest case in which the magnetic interaction vector is a real vector of amplitude m , directed along the x axis (this is always the case for the phases considered here), we have:

$$\delta^{(\Gamma)} = \frac{4m\Re(F^{(0)}\overline{F^{(1)}})\cos^3\theta}{|F^{(0)}|^2(\cos^2(2\theta) + 1) + 2|F^{(1)}|^2m^2\cos^2(\theta)} \quad (8)$$

where the overline indicates complex conjugation and θ is the Bragg angle. $\Re()$ and $\Im()$ denote real and imaginary parts, respectively. For this simple magnetic interaction vector, the dichroism changes sign upon reversal of the magnetization, and consequently $\text{Asy}_\delta^{(\Gamma)} = 0$.

B. Circular dichroism for purely magnetic peaks

The general form of the circular dichroism for a purely magnetic peak is

$$\delta = \frac{\Im(M_y(\overline{M_x}\cos\theta + \overline{M_z}\sin\theta))\sin(2\theta)}{|M_x|^2\cos^2\theta + |M_z|^2\sin^2\theta + |M_y|^2\sin^2(2\theta)} \quad (9)$$

A very important result can be obtained by inspecting Eq. (9): The circular dichroism of a purely magnetic Bragg peak is always zero for centrosymmetric magnetic phases. In these cases, the magnetic interaction vector is always a real vector multiplied by a global phase, and the numerator of Fig. 9 is identically zero. As we shall see, the circular dichroism is proportional to the magnetic chirality for the helical phase and to the magnetic polarity for the TC phase.

TABLE I. Magnetic interaction vectors for the phases and Bragg reflections considered in this paper. Helix/LC: Helical/longitudinal conical phase. TC: 2-fan transverse conical phase. m-TC: modified 2-fan transverse conical phase. The Bragg angles of the commensurate 2-fan phases at low temperatures and at the photon energy employed for the experiment ($\hbar\omega = 707.48$ eV, with a wavelength of 17.5250 Å) are also indicated. For the helical/LC phase, the superscripts refer to the direction of the magnetic moment parallel or perpendicular to the xy -plane (N.B. the perpendicular component is zero in the planar helical phase).

Reflection: $(0, 0, 3 - k)/(0,0,1.5)$, $\theta_{(0,0,1.5)} = 17.67^\circ$			
Phase	M_x	M_y	M_z
Helix/LC	$-\frac{1}{2}(m_L^\parallel + m_S^\parallel)$	$\frac{1}{2}i(m_L^\parallel - m_S^\parallel)$	0
TC	0	$im_L \sin \phi$	$m_S \sin \psi$
m-TC	$m_S \sin \psi \sin \psi_1$	$im_L \sin \phi \cos \phi_1$	$m_S \sin \psi \cos \psi_1$
Reflection: $(0, 0, 3 + k)/(0,0,4.5)$, $\theta_{(0,0,4.5)} = 66.30^\circ$			
Phase	M_x	M_y	M_z
Helix/LC	$-\frac{1}{2}(m_L^\parallel + m_S^\parallel)$	$-\frac{1}{2}i(m_L^\parallel - m_S^\parallel)$	0
TC	0	$-im_L \sin \phi$	$m_S \sin \psi$
m-TC	$m_S \sin \psi \sin \psi_1$	$-im_L \sin \phi \cos \phi_1$	$m_S \sin \psi \cos \psi_1$
Reflection: $(0,0,3)$, $\theta_{0,0,3} = 37.62^\circ$			
Phase	M_x	M_y	M_z
Helix/LC	0	0	$m_S^\perp - m_L^\perp$
TC	$m_L \cos \phi - m_S \cos \psi$	0	0
m-TC	$m_L \cos \phi \cos \phi_1 - m_S \cos \psi \cos \psi_1$	0	$m_S \cos \psi \sin \psi_1$

The circular dichroism averaged between the two directions of B is:

$$\begin{aligned} \delta_{\text{ave}} &= \frac{1}{2}(\delta_{B^+} - \delta_{B^-}) \\ &= \frac{\Im(M_y \overline{M_z}) \sin \theta \sin(2\theta)}{|M_x|^2 \cos^2 \theta + |M_z|^2 \sin^2 \theta + |M_y|^2 \sin^2(2\theta)}. \end{aligned} \quad (10)$$

The circular asymmetry is

$$A_{\text{sy}\delta} = \frac{\delta_{B^+} + \delta_{B^-}}{\delta_{B^+} - \delta_{B^-}} = \frac{\Im(M_y \overline{M_x}) \cot \theta}{\Im(M_y \overline{M_z})}. \quad (11)$$

1. General formulas for the linear polarization channels

The scattering intensities in the linear channels are, to within a multiplicative factor,

$$\begin{aligned} I_{\sigma\sigma'} &= |F^{(0)}|^2 \\ I_{\sigma\pi'} &= |F^{(1)}(M_x \cos \theta + M_z \sin \theta)|^2 \\ I_{\pi\sigma'} &= |F^{(1)}(M_x \cos \theta - M_z \sin \theta)|^2 \\ I_{\pi\pi'} &= |F^{(0)} \cos(2\theta) - iF^{(1)}M_y \sin(2\theta)|^2 \end{aligned} \quad (12)$$

where, for purely magnetic peaks, we can take $F^{(0)} = 0$. The linear asymmetries are:

$$\begin{aligned} A_{\text{sy}\sigma\sigma} &= 0 \\ A_{\text{sy}\sigma\pi'} &= \frac{\Re(M_x \overline{M_z}) \sin(2\theta)}{|M_x|^2 \cos^2 \theta + |M_z|^2 \sin^2 \theta} \\ A_{\text{sy}\pi\sigma'} &= -\frac{\Re(M_x \overline{M_z}) \sin(2\theta)}{|M_x|^2 \cos^2 \theta + |M_z|^2 \sin^2 \theta} \\ A_{\text{sy}\pi\pi'} &= \frac{\Im(F^{(1)} \overline{F^{(0)}} M_y) \sin(4\theta)}{|F^{(0)}|^2 \cos^2(2\theta) + |F^{(1)}|^2 |M_y|^2 \sin^2(2\theta)}. \end{aligned} \quad (13)$$

The last term is, once again, due to magnetic-structural interference; for purely magnetic peaks, we can set $F^{(0)} = 0$, so that also $A_{\text{sy}\pi\pi'} = 0$.

C. Magnetic interaction vector calculations

Using the block model approximation, the complex M_x , M_y , and M_z components of the magnetic interaction vectors for different Bragg peaks can be easily calculated (this is performed in detail in Appendix B). The results are summarized in Table I for all the relevant reflections, together with the Bragg angles at which these reflections occur at the photon energy of 707.48 eV. This information is sufficient to calculate all relevant intensities (to within a global scale factor), as well as the circular dichroism and asymmetries, using the formulas from the previous section.

IV. RESULTS AND DISCUSSION

A. Helical phase

We begin by discussing our characterization of the helical magnetic structure, stable in the absence of an applied field and for $T \leq 390$ K. For this phase, the expectation from the previous section is that the $(0,0,3)$ reflection should not be circular dichroic (there is no magnetic contribution to the scattering intensity at this position in this phase). Conversely, the two magnetic satellites, $(0,0,3 \pm k)$, should exhibit circular dichroism with magnitude equal to

$$\delta_{\text{helix}} = \frac{\pm 2\gamma \sin \theta}{1 + 2 \sin^2 \theta}, \quad (14)$$

where \pm denotes the measured satellite ($\pm \mathbf{k}$) and γ is the mean chirality of the probed region. Hence, the dichroism of the two satellites has opposite sign for domains of fixed chirality (γ).

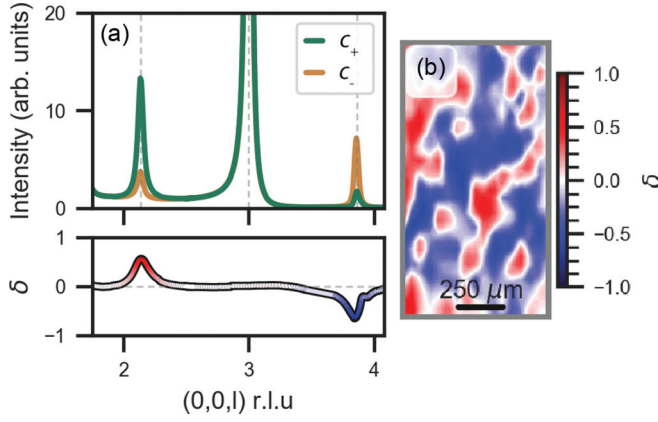


FIG. 4. Diffraction measurements of the helical phase. (a) $(0,0,l)$ scan measured with circular positive (solid green line) and circular negative (solid brown line) at 300 K in the absence of an applied magnetic field. The bottom panel displays the circular dichroism calculated from these measurements. Note the lack of appreciable dichroism for the $(0,0,3)$ reflection and the large dichroism, of opposite sign, for $(0,0,3\pm k)$. (b) Spatially resolved circular dichroism (color) of the $(0,0,3-k)$ reflection at 25 K in zero applied magnetic field.

Figure 4(a) shows a $(0,0,l)$ scan of the helical phase with circular incident x rays, which indeed demonstrates opposite circular dichroism for the two incommensurate magnetic satellites. Furthermore, we spatially resolved the circular dichroism of the $(0,0,3-k)$ reflection [Fig. 4(b)] which exhibits clear regions of contrast. These two regions correspond to the two possible chirality domains of the helical state. The domain morphology is similar to previous observations of chirality domains in other Y -type hexaferrites [9]. We also found that the measured helical domain configuration is invariant upon cooling to 25 K and to the application of small applied magnetic fields. It is reported that the helical phase transforms to a longitudinal conical state at low temperatures [5,17], which would imply that the $(0,0,3)$ reflection should develop a small amount of dichroism. However, no evidence of this transition was observed in our experiment for temperatures above 25 K. In the following we refer to this phase as the helical phase but remark that classification of this phase as a longitudinal state would not affect the presented results.

B. Magnetoelectric TC phase at low temperatures

Below approximately 100 K, the application of a 150 mT magnetic field in the basal plane of the helix transforms this phase to the TC state [21]. This transition was evident in our diffraction data by the disappearance of the two incommensurate magnetic satellites of the helical phase, and the emergence of two commensurate magnetic reflections at $(0,0,1.5)$ and $(0,0,4.5)$, both of which are characteristic of the TC phase [5].

1. Circular dichroism measurements

The intensities of the $(0,0,3)$ and $(0,0,4.5)$ reflections were measured with circularly polarised incident x rays of both polarities (c^\pm), as shown in Fig. 5. For these measurements,

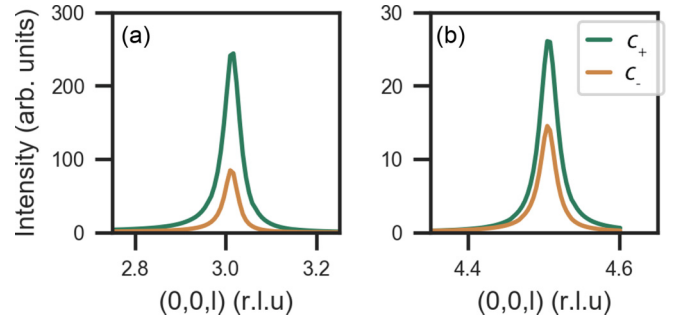


FIG. 5. Intensity of the $(0,0,3)$ and $(0,0,4.5)$ reflections [(a) and (b), respectively] measured with circularly positive (c_+) and circularly negative (c_-) polarized incident x rays. No polarization analysis of the scattered beam was performed. Measurements were performed at 40 K and with a 250 mT magnetic field applied parallel to $[2,1,0]$.

polarization analysis of the scattered beam was not performed. Clearly, both the $(0,0,3)$ and $(0,0,4.5)$ reflections exhibit a large difference in scattering intensity between c^+ and c^- polarized incident x rays and both, therefore, possess a large circular dichroism (Fig. 5). By inserting the appropriate magnetic interaction vectors and Bragg angles for the TC phase into Eqs. (8) and (9), we obtain the following expressions:

$$\begin{aligned} \delta_{(003)} &= \frac{4m\lambda(F^{(0)}\overline{F^{(1)}})\cos^3\theta}{|F^{(0)}|^2(\cos^2(2\theta)+1)+2|F^{(1)}|^2m^2\cos^2\theta} \\ \delta_{(001.5)} &= -\frac{2\lambda\cos\theta}{m_S^2\sin^2\psi+2m_L^2\sin^2\phi^2\cos^2\theta} \\ \delta_{(004.5)} &= \frac{2\lambda\cos\theta}{m_S^2\sin^2\psi+2m_L^2\sin^2\phi^2\cos^2\theta}. \end{aligned} \quad (15)$$

We conclude that, for this structural model, $\delta^{(003)}$ is to first order proportional to the amplitude of the net magnetization m , while $\delta^{(001.5)}$ and $\delta^{(004.5)}$ are proportional to the amplitude of the magnetic polarity λ [Eq. (4)]. We emphasise that the circular dichroism of the two magnetic peaks naturally arises due to the noncentrosymmetric nature of the TC phase [9], and there is therefore no need to invoke a hypothetical structural modulation with the same propagation vector [10]. The absence of any scattering intensity at the $(0,0,4.5)$ and $(0,0,1.5)$ positions for scans performed with linear incident polarization in the $\sigma\sigma'$ channel, which is only sensitive to the crystal structure, confirm that such a structural modulation is not present (see Sec. IV B 3).

To investigate how the circular dichroisms vary over the extent of the sample we restricted the diameter of the incident x-ray beam to $50\ \mu\text{m}$ and performed a raster scan in steps of $50\ \mu\text{m}$ to spatially resolve the circular dichroism of the $(0,0,3)$ and $(0,0,4.5)$ reflections. With a magnetic field of $-250\ \text{mT}$ applied to the sample, the $(0,0,3)$ reflection exhibits spatially homogeneous, positive circular dichroism over the extent of the sample [Fig. 6(a)], indicating that the sample is uniformly magnetized.

Unlike the $(0,0,3)$ reflection, the spatial map of the $(0,0,4.5)$ reflection exhibits two regions of opposite contrast [Fig. 6(c)]. Based on Eq. (15), the two regions of opposite contrast must be magnetic polarity domains with opposite λ . Importantly, since λ and \mathbf{P} are coupled, the two regions must also

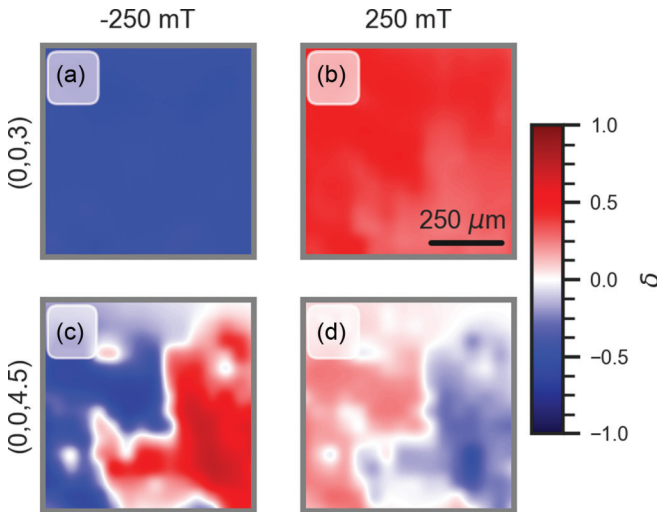


FIG. 6. Spatially resolved circular dichroism of the (0,0,3) (a) and (b) and the (0,0,4.5) (c) and (d) reflections at -250 mT (a) and (c) and after isothermal reversal of the magnetic field to $+250$ mT (b) and (d). The contrast of the (0,0,4.5) reflection reflects the magnetic and electric polarity domain configuration of our BSMFO single crystal. The color bar (far right) is common to all figures and denotes the measured circular dichroism which can take values between ± 1 . Figures are to the same scale, denoted in panel (b). In the bottom panels, the left domain [blue in (c) and red in (d)] has *positive* toroidal moment, while the right domain [red in (c) and blue in (d)] has *negative* toroidal moment (see text).

correspond to *polar* domains of the sample. Because no electrical bias was applied to the sample, we would expect equal populations of the two polarity domains, as we indeed observe. It is noteworthy that there is a complete lack of correlation between magnetic polarity domains at $T = 40$ K and $B = -250$ mT and the zero field helical domains we measured, which are considerably smaller (Fig. 4). Taking the (0,0,3) and (0,0,4.5) domain maps together, we can conclude that the magnetic polarity domains are also *ferrotoroidal* domains [Eq. (3)].

Next, we investigated the effect of reversing the direction of the applied magnetic field on the domain configuration. Figure 6(b) shows a map of the circular dichroism of the (0,0,3) reflection after field reversal to $+250$ mT. The sign of the circular dichroism has switched everywhere on the sample, consistent with a uniform reversal of the sample's magnetization. Remarkably, the sign of the (0,0,4.5) dichroism has also switched on a pixel-by-pixel basis [Fig. 6(d)], i.e., red regions turn blue and blue regions turn red, meaning that the magnetic polarity (and hence the electrical polarization) has switched sign within each of the domains. It is noteworthy that the boundary between the two domains has remained pinned throughout the switching process. This observation indicates that the ferrotoroidal domains are very stable, while the magnetization of each ferrotoroidal domain can be almost freely rotated, resulting in a simultaneous reversal of the electrical polarization. Finally, we note that the *magnitude* of the (0,0,4.5) dichroism is slightly weaker at $+250$ mT than at -250 mT (indicated by the fainter colors in the figure).

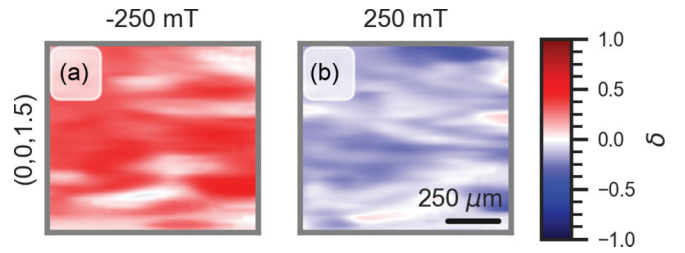


FIG. 7. Circular dichroism maps of the (0,0,1.5) reflection after an E_+B_+ magnetoelectric poling procedure. (a) for $B = -250$ mT and (b) for $B = +250$ mT.

There is therefore a small but noticeable *circular asymmetry* [Eq. (6)], which will be further discussed below.

2. Ferrotoroidal domain control

It is well known that ferrotoroidal domains can be manipulated by a combination of orthogonal electric and magnetic fields [28,29]. It is therefore natural to assume that electrical biasing of the sample under an applied magnetic field should result in the removal of the ferrotoroidal domain boundaries. To test this hypothesis, we used a second sample prepared with electrical contacts such that electric fields could be applied perpendicular to the applied magnetic field. In the absence of electric field bias, the circular asymmetry of the (0,0,1.5) reflection [the (0,0,4.5) reflection was not accessible in this second experiment] exhibited regions of multiple contrast (not shown). However, after performing a magnetoelectric poling procedure identical to the one as described in Sec. II A (Fig. 2), the raster scans of the circular dichroism for the (0,0,1.5) reflection [Fig. 7(a)] became uniform, indicating that the sample had acquired a uniform magnetic polarity. The magnetization of the sample is also uniform, as indicated by circular dichroism of the (0,0,3) reflection (not shown here), indicating that the sample now comprises a single ferrotoroidal domain. Upon reversal of the magnetic field, once again the signs of the circular dichroism for both (0,0,3) and (0,0,1.5) reflections reverse (Fig. 7), indicating a global switching of both magnetization and magnetic polarity, while the direction of the ferrotoroidal moment remains the same—consistent with our bulk measurements in which the electric polarization fully switches on inversion of the applied magnetic field (Fig. 2).

3. Linear polarization analysis

Scans over the (0,0,3) and (0,0,4.5) reflections (TC phase) are displayed in Fig. 8. Measurements were performed at 40 K and at $+250$ mT in each of the four linear polarization channels (defined in the basis of orthogonal linear polarization states shown in Fig. 3). The (0,0,3) reflection [Fig. 8(a)] scatters most intensely in the $\sigma - \sigma'$ channel and is therefore predominately of charge origin, as expected. Weak intensity in both rotated channels ($\sigma - \pi'$ and $\pi - \sigma'$) demonstrates that there is scattering of magnetic origin at this location (being the only scattering process which can rotate the plane of polarization of incident light). Conversely, for the (0,0,4.5) reflection [Fig. 8(b)] the intensity in the $\sigma - \sigma'$ channel is zero, but scattering is observed in every other linear polarization

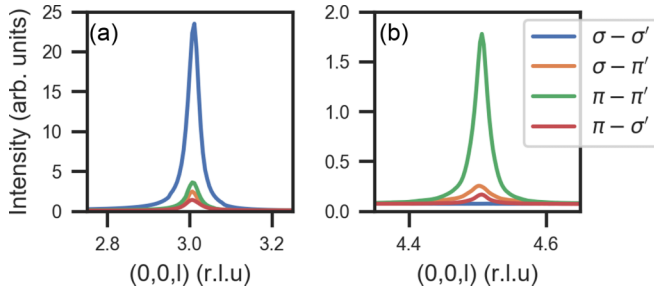


FIG. 8. Intensity of the (0,0,3) and (0,0,4.5) reflections [(a) and (b), respectively] measured in each linear polarization channel (color). Measurements were performed at 40 K and with a 250 mT magnetic field applied parallel to [2,1,0].

channel. This scattering is therefore of *pure* magnetic origin and its properties must, therefore, be linked to the magnetic structure of BSMFO. The importance of this observation becomes apparent when considering that there is a precedent for the commensurate reflections of the *Y*-type hexaferrites to be of *mixed* charge and magnetic scattering [10].

C. Dynamical measurements during magnetic field switching

The (0,0,3) and (0,0,4.5) diffraction intensities were measured in each linear polarization channel as the magnetic field was rapidly swept (6.25 mTs^{-1}) through several hysteresis loops, enabling one to track how different components of the magnetic structure changed during the switching process.

The most salient features of these data (shown in Fig. 9) are the presence of linear asymmetry (i.e., a step in the scattering intensity between positive and negative magnetic fields), a large, transient increase or decrease in the scattering intensities occurring at precisely the same field magnitude (23 mT and -26 mT , denoted by dashed gray lines/shaded areas in Fig. 9) for all linear polarization channels, and significant field hysteresis. Under slow magnetic field switching ($\sim 0.1 \text{ mTs}^{-1}$, not shown) the transient features are greatly suppressed and are therefore to be associated with the dynamic polarity switching process.

Ishiwata *et al.* proposed a mechanism for the magnetic polarity reversal in BMFO (the parent compound of BSMFO), by which the magnetic structure coherently rotates around \mathbf{P} (*y*), corresponding to a smooth transformation of the TC phase into the LC phase, and then back to a TC phase of opposite polarity in the oppositely applied magnetic field [7]. Our measurements categorically demonstrate that this cannot be the case in BSMFO. Instead, the peaked intensity in the (0,0,3) $\pi - \pi'$ channel uniquely identifies the evolution of a *y* component of the magnetization upon switching, indicating that the magnetic structure rotates about the *c* axis (*z*) as opposed to \mathbf{P} . Furthermore, a simple calculation of the diffraction intensity in all linear scattering channels based upon the block-model TC structure (not shown), showed that all peaked features observed upon switching are fully consistent with a coherent rotation of the magnetic structure around *c*.

D. Beyond the block model: The true nature of the TC phase

Although the peaks near zero field could be accounted for by the block TC model, we emphasise that this model *does*

not reproduce the large linear asymmetry (i.e. the ‘steps’ in the scattering intensity) observed in the rotated polarization channels ($\sigma\pi'$ and $\pi\sigma'$) for both the (0,0,3) and (0,0,4.5) peaks (Fig. 9). We have carefully considered the possibility that the steps might arise from the $F^{(2)}$ term, which we have thus far ignored (see Appendix A 4 for a detailed calculation of this term). It is not difficult to show that, for the TC phase, the magnetic interaction rank-2 tensor associated with the (0,0,3) peak is purely diagonal and does not contribute to intensity in the rotated channel. A more detailed calculation, reported in Appendix C shows that this term does not produce linear asymmetry in the (0,0,4.5) peak either. Another possibility is that the steps might arise from magnetic field misalignment with respect to the scattering plane. This possibility can also be ruled out, at least for the simple block-TC model: The observed asymmetry can only occur if M_x and M_z are partially in phase, and if only one of them switches upon field reversal (Sec. III B 1), and this combination can never be obtained with field misalignment for either of the reflections.

Since other possibilities fail, we must come to the conclusion that the observed asymmetry in the rotated channels *must* arise from a sizable internal distortion of the magnetic structure from the block model considered thus far. Taking the TC model as a starting point, the simplest way to reproduce the observed steps is to introduce a *scalar modulation* of the magnetic structure with commensurate propagation vector $\mathbf{k}_z = (0,0,1.5)$. In this context, scalar modulation simply means a modulation of the magnetic cone angles and/or of the helical pitch. Even without performing additional calculations, it is evident that such modulation would *simultaneously* introduce an M_z component at the Γ point (possibly associated with a small net moment in the *z* direction), *and* an M_x component with \mathbf{k}_z propagation vector, both having the correct phase to produce asymmetry in the rotated channels. We emphasise that the large size of the asymmetry steps means that the M_x and M_z components for both propagation vectors must be of the same order of magnitude, which constitute a significant deviation from the ‘standard’ TC model.

Indeed, the observed dichroism, asymmetries, and switching behavior are well reproduced [Figs. 9(g)–9(i)] by such a ‘modified’ 2-fan transverse conical model (m-TC), which was constructed with the minimal modifications required to explain the data while remaining physical. There are two significant points of departure from the ‘standard’ TC model: (1) The L block is split into two noncollinear subblocks, L1 and L2 (representing the bottom and the top of the L block, respectively), making an angle $2\phi_1$ with each other, and (2) the out-of-plane lifting angle of the S block is no longer constant, but varies by $\pm\psi_1$. As shown in more detail in Appendix B 3, the effective magnetic moment of each block is constant, and so is the dot product between adjacent blocks, provided that the following constraint is fulfilled:

$$\cos(\psi - \psi_1) \cos(\phi + \phi_1) = \cos(\psi + \psi_1) \cos(\phi - \phi_1) \quad (16)$$

Thus, the m-TC model has only the additional parameter ϕ_1 , since ψ_1 can be calculated from Eq. (16). The m-TC model has two characteristics making it strongly reminiscent of the zero-field LC phase: it has a net chirality (there are

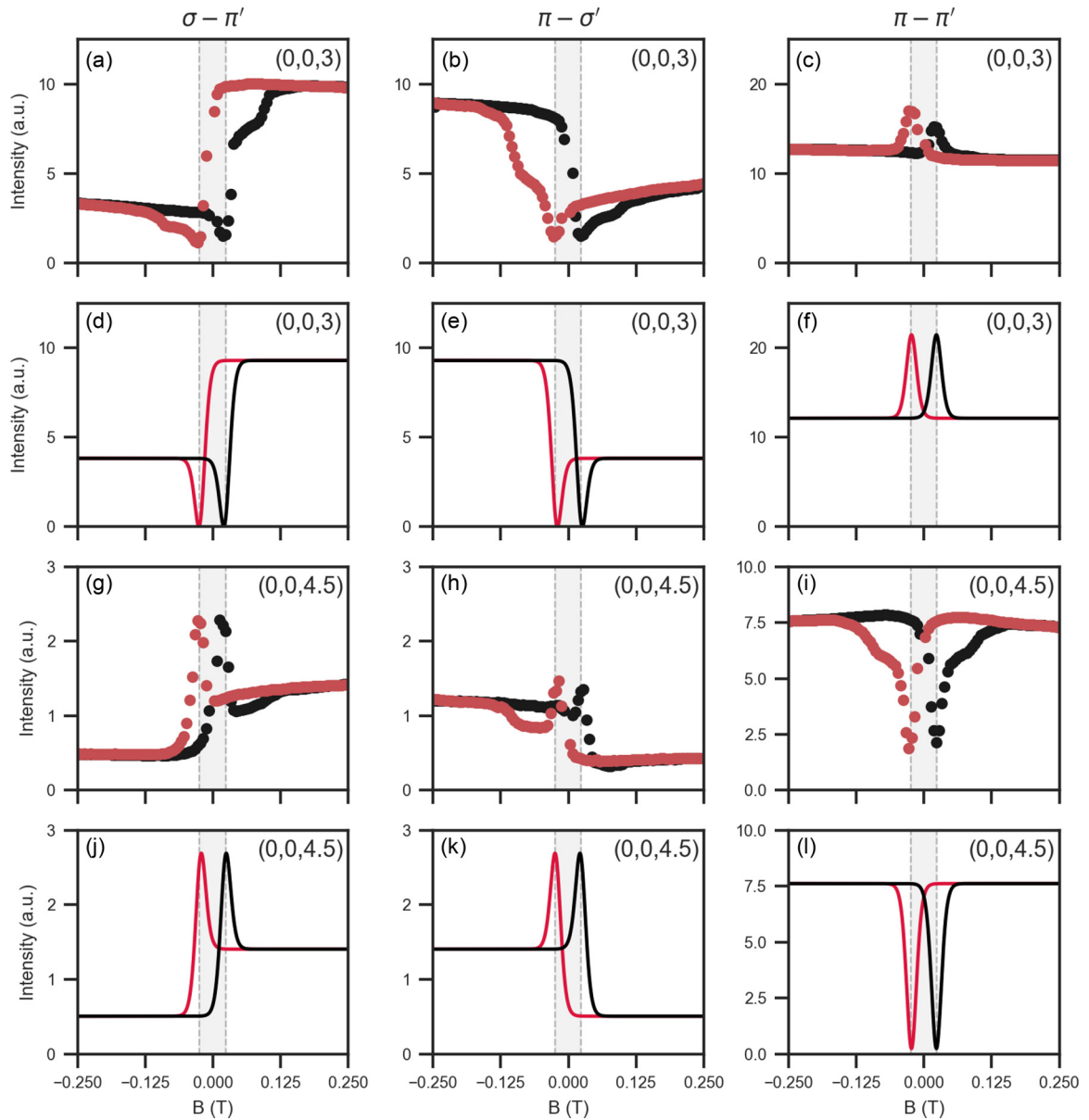


FIG. 9. (a)–(c),(g)–(i) Measured scattering intensity for the (0,0,3) and (0,0,4.5) reflections, respectively, in three linear polarization channels during the magnetic polarity switching process at 40 K. The magnetic field was swept at 6.25 mTs^{-1} . Black data points show the scattering intensity measured on increasing magnetic field and red points on decreasing. The pair of dashed gray lines are plotted at the same field values (23 mT and -26 mT) across all channels. (d)–(f),(j)–(l) Calculated intensity of the (0,0,3) and (0,0,4.5) reflection, respectively, in three linear polarization channels for the m-TC structure as the magnetic structure is coherently rotated about c . Parameters used in the calculations are those discussed in Table II with the addition of arbitrary scale factors and a parameter controlling the width of the hysteresis during switching. Color (red or black) denotes the direction of the field sweep (decreasing and increasing field, respectively).

left-handed and right-handed versions of it), and also a small net ferrimagnetic moment along the z axis in addition to the main x -axis magnetization. In the m-TC model, these two facts are related, since a structure possessing both net chirality and ferrotoroidal moment along z must also, by symmetry, possess a ferromagnetic moment along z .

Table II lists the experimental and calculated values of dichroism and asymmetry in different linear and circular channels for the m-TC model. Since the main fan angle was set to a typical literature value for the TC phase ($\phi = 38^\circ$) [12], there are only two variable parameters (ψ and ϕ_1), which were adjusted to obtain an exact agreement with the (0,0,4.5)

experimental values of δ_{ave} and $A_{\text{asy}\sigma\pi'}$. The agreement we obtain with the other experimental values is quite reasonable, especially considering that the model is overconstrained by the data. We emphasise again that the deviations from the standard TC model, represented by the angles ϕ_1 and ψ_1 , are very large. Undoubtedly, this is in part a consequence of the simplicity of the model, which fails to capture the full internal complexity of the hexaferrite structure.

Since we only have three magnetic Bragg peaks at our disposal (albeit with several polarization channels) and employ a technique that it is not entirely bulk-sensitive, proposing a full model of the ‘true’ magnetic structure of BSMFO in

TABLE II. Experimental and calculated values of dichroism and asymmetry in different channels for the modified 2-fan transverse conical model (m-TC). The calculations were performed for a negative-dichroism domain (a ‘blue’ domain in Fig. 6). The main fan angle was set to $\phi = 38^\circ$ —a typical value for the standard TC phase; ψ and ϕ_1 were adjusted to obtain precise agreement with the (0,0,4.5) experimental values of δ_{ave} and $Asy_{\sigma\pi'}$, while all the other values are unconstrained outputs of the calculations. Optimal values were $\psi = 36.5^\circ$ and $\phi_1 = 28.0^\circ$. The value of $\psi_1 = 29.3^\circ$ was calculated from Eq. (16). The circular dichroism and asymmetry for the (0,0,3) reflection depend on the unknown ratio $F^{(0)}/F^{(1)}$ and could not therefore be calculated.

Parameter	Reflection: (0,0,3)		Reflection: (0,0,4.5)	
	Expt.	Calc.	Expt.	Calc.
δ_{ave}	0.49		-0.59	-0.59
Asy_δ			0.28	0.25
$Asy_{\sigma\pi'}$	0.52	0.41	0.47	0.47
$Asy_{\pi\sigma'}$	-0.42	-0.41	-0.47	-0.47
$Asy_{\pi\pi'}$	-0.04	0	-0.00	0

its magnetoelectric phase would be incautious at best. Nevertheless, we speculate that all noncollinear magnetic structures of *Y*-type hexaferrites are significantly more complex than is currently believed, and that some of the internal degrees of freedom of these structures are fixed by competing exchanges in a way that is not captured by the block models, and that these features are preserved upon applications of magnetic field, adding unexpected ‘twists’ to the magnetic structure of the magnetoelectric phases.

V. CONCLUSIONS

We have shown that resonant x-ray microdiffraction can be used to measure the magnetic polarity domain configuration of the *Y*-type hexaferrite $\text{Ba}_{0.5}\text{Sr}_{1.5}\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$. We provide direct evidence of the magnetic polarity switching during inversion of the applied magnetic field and show that electric fields can be used to prepare a single magnetic polarity domain. By dynamic measurement of the scattering during the switching process we were able to propose a mechanism for the polarity switching—a coherent rotation of the magnetic structure about the *c* axis, in contention with the switching mechanism originally proposed for the parent compound BMFO [7]. We also showed that the established magnetoelectric structures are inconsistent with our data, and we constructed a minimally modified magnetic structure model that reproduces quantitatively the behavior of all the x-ray polarization channels upon field reversal.

ACKNOWLEDGMENTS

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APPENDIX A: BASIC ASPECTS OF MAGNETIC SCATTERING THEORY

In this section we reproduce the essential, well-known results of the resonant magnetic scattering theory, as it appears, for instance, in J. P. Hill and D. F. McMorrow [26], but we recast them in the compact formalism that we employed throughout the paper.

1. X-ray polarization

Using the Cartesian coordinates presented in Fig. 3, we can define the incident and scattered polarization of the x rays as follows:

$$\begin{aligned}
 \epsilon_\sigma &= \epsilon'_\sigma = (0 \quad 1 \quad 0) \\
 \epsilon_\pi &= (\sin \theta \quad 0 \quad \cos \theta) \\
 \epsilon'_\pi &= (-\sin \theta \quad 0 \quad +\cos \theta) \\
 \epsilon_{c+} &= \frac{1}{\sqrt{2}}(\sin \theta \quad i \quad \cos \theta) \\
 \epsilon_{c-} &= \frac{1}{\sqrt{2}}(\sin \theta \quad -i \quad \cos \theta). \tag{A1}
 \end{aligned}$$

2. Resonant magnetic x-ray diffraction

Here, we employ a slightly different formalism with respect to the classic treatment by J. P. Hill and D. F. McMorrow [26], with the aim of separating the geometrical terms (which are experiment dependent) from the magnetic interaction vectors/tensors, which can be evaluated once and for all for a given magnetic structure. For this purpose, we re-write Eq. (7) in McMorrow & Hill [26] in tensorial form as follows:

$$f_{\text{nEI}}^{\text{XRES}} = \epsilon'_\alpha \epsilon_\beta T_{\text{tot}}^{\alpha\beta} \tag{A2}$$

where

$$T_{\text{tot}}^{\alpha\beta} = F^{(0)}\delta^{\alpha\beta} - iF^{(1)}\epsilon^{\alpha\beta\gamma}M_\gamma + F^{(2)}T^{\alpha\beta} \tag{A3}$$

and the three terms in Eq. (A3) represent (from left to right) the contributions from Thomson/anomalous charge scattering, $F^{(1)}$ scattering (proportional to the magnetic moments), and $F^{(2)}$ (proportional to the square of the magnetic moments), $F^{(0)}$, $F^{(1)}$, and $F^{(2)}$ being the resonant scattering factors (in general, complex numbers). For simplicity, we have included into $F^{(0)}$ the ordinary Thomson structure factor, which is well approximated by a real number (the crystal structure is close to being centrosymmetric). The values of ϵ can be taken directly from Appendix A1 for our specific experimental configurations. In Eq. (A3), M_γ are the components of the ordinary magnetic interaction vector, a complex vector that also appears in the calculations of magnetic neutron scattering cross sections, so that $\epsilon^{\alpha\beta\gamma}M_\gamma$ is an antisymmetric rank-two tensor, while $T^{\alpha\beta}$ is the symmetric magnetic interaction rank-two tensor associated with $F^{(2)}$. The calculations of M_γ and $T^{\alpha\beta}$ for a generic magnetic structure and allowed Bragg peaks are reported in the remainder of this Appendix, for both commensurate and incommensurate magnetic structures.

3. Magnetic interaction vector

The definition of the complex magnetic interaction vector \mathbf{M} naturally arises from the calculation of the x-rays magnetic scattering cross section. In the case of RXD, the relevant term is

$$-iF^{(1)}\epsilon^{\alpha\beta\gamma}M_\gamma = -iF^{(1)}\begin{pmatrix} 0 & M_z & -M_y \\ -M_z & 0 & M_x \\ M_y & -M_x & 0 \end{pmatrix}. \quad (\text{A4})$$

When calculating the scattering amplitude, one needs to perform the following sum, running over the lattice nodes n and the sites in the unit cell m :

$$\mathbf{A} = \sum_{n,m} e^{i\mathbf{Q}\cdot(\mathbf{R}_n+\mathbf{r}_m)} \mathbf{m}_{nm} \quad (\text{A5})$$

where the \mathbf{m}_{nm} are the real magnetic moments at site m in unit cell n . \mathbf{R}_n denotes the position vector of the origin of unit cell n , while \mathbf{r}_m denotes the position vector of site m . Using the propagation vector formalism,

$$\mathbf{m}_{nm} = \sum_{\mathbf{k},m} \mathbf{S}_{\mathbf{k},m} e^{-i\mathbf{k}\cdot\mathbf{R}_n} + \bar{\mathbf{S}}_{\mathbf{k},m} e^{+i\mathbf{k}\cdot\mathbf{R}_n} \quad (\text{A6})$$

where $\mathbf{S}_{\mathbf{k},m}$ are the complex-vector Fourier component of the magnetic moment at site m and for propagation vector \mathbf{k} . After performing the lattice sum, one obtains:

$$\mathbf{A} = \delta(\mathbf{Q} - \mathbf{k} - \boldsymbol{\tau}) \mathbf{M}_{\mathbf{k}}^+ + \delta(\mathbf{Q} + \mathbf{k} - \boldsymbol{\tau}) \mathbf{M}_{\mathbf{k}}^- \quad (\text{A7})$$

where \mathbf{Q} is the scattering vector, $\boldsymbol{\tau}$ is a reciprocal lattice vector, and the two magnetic interaction vectors are

$$\begin{aligned} \mathbf{M}_{\mathbf{k}}^+ &= \sum_m e^{i\mathbf{Q}\cdot\mathbf{r}_m} \mathbf{S}_{\mathbf{k},m} \\ \mathbf{M}_{\mathbf{k}}^- &= \sum_m e^{i\mathbf{Q}\cdot\mathbf{r}_m} \bar{\mathbf{S}}_{\mathbf{k},m}. \end{aligned} \quad (\text{A8})$$

In the most general case of \mathbf{k} within the Brillouin zone, each Bragg peak develops two satellites, one at position $\boldsymbol{\tau} + \mathbf{k}$ and the other at $\boldsymbol{\tau} - \mathbf{k}$. In the special case of $2\mathbf{k} = RLV$ (a reciprocal lattice vector) then both terms will contribute to the same satellite, with the magnetic interaction vector

$$\mathbf{M}_{\mathbf{k}} = 2 \sum_m e^{i\mathbf{Q}\cdot\mathbf{r}_m} \Re(\mathbf{S}_{\mathbf{k},m}). \quad (\text{A9})$$

4. Magnetic interaction rank-two tensor

Similarly, the scattering amplitude for the $F(2)$ term are performed using the magnetic interaction rank-two tensor:

$$\begin{aligned} B^{\alpha\beta} &= \delta(\mathbf{Q} - 2\mathbf{k} - \boldsymbol{\tau}) T_{2\mathbf{k}}^{\alpha\beta+} + \delta(\mathbf{Q} + 2\mathbf{k} - \boldsymbol{\tau}) T_{2\mathbf{k}}^{\alpha\beta-} \\ &+ \delta(\mathbf{Q} - \boldsymbol{\tau}) T_0^{\alpha\beta} \end{aligned} \quad (\text{A10})$$

where

$$\begin{aligned} T_{2\mathbf{k}}^{\alpha\beta+} &= \sum_m e^{i\mathbf{Q}\cdot\mathbf{r}_m} \mathbf{S}_{\mathbf{k},m}^\alpha \mathbf{S}_{\mathbf{k},m}^\beta \\ T_{2\mathbf{k}}^{\alpha\beta-} &= \sum_m e^{i\mathbf{Q}\cdot\mathbf{r}_m} \bar{\mathbf{S}}_{\mathbf{k},m}^\alpha \bar{\mathbf{S}}_{\mathbf{k},m}^\beta \\ T_0^{\alpha\beta} &= \sum_m e^{i\mathbf{Q}\cdot\mathbf{r}_m} 2\Re(\mathbf{S}_{\mathbf{k},m}^\alpha \bar{\mathbf{S}}_{\mathbf{k},m}^\beta). \end{aligned} \quad (\text{A11})$$

So, there are contributions at $\boldsymbol{\tau} \pm 2\mathbf{k}$ and also at the Γ point $\boldsymbol{\tau}$. Once again if $2\mathbf{k} = RLV$ then there is only a contribution at the Γ point $\boldsymbol{\tau}$, with the following magnetic interaction tensor:

$$T^{\alpha\beta} = 4 \sum_m e^{i\mathbf{Q}\cdot\mathbf{r}_m} \Re(\mathbf{S}_{\mathbf{k},m}^\alpha) \Re(\mathbf{S}_{\mathbf{k},m}^\beta). \quad (\text{A12})$$

APPENDIX B: MAGNETIC MODELS

In this section, we calculate the Fourier components \mathbf{S} , the real magnetic moments \mathbf{m} , and the magnetic interaction vectors \mathbf{M} for all the phases and reflections of interest for this paper. We employ either the standard block model with small (S) and large (L) blocks or the modified block model, with the large block split into L1 and L2. In all cases the values of the magnetic moments depend on the z component of the unit-cell position vector \mathbf{R}_n (herewith denoted as R_n), where R_n can assume all values such that $3R_n = \text{integer}$. This is because we employ the hexagonal conventions for the propagation vector, whereas the primitive unit cell is rhombohedral.

1. Helix/LC

The calculations for the helical and longitudinal-conical (LC) phases are very similar, and are here performed together, the only difference being that the LC phase has a component at the Γ point. The other propagation vector is along the Λ line in the Brillouin zone, i.e. $\mathbf{k} = (0, 0, k_{\text{hel}})$, with k_{hel} in the range 0.6–0.8. The chirality of the helix is denoted by γ and is equal to ± 1 . Herewith, m_S^\perp and m_L^\perp denote the z axis components of the magnetic moments, while m_S^\parallel and m_L^\parallel denote the xy plane components.

$$\begin{aligned} \mathbf{S}_S^\Gamma &= \begin{bmatrix} 0 \\ 0 \\ \frac{1}{2}m_S^\perp \end{bmatrix} \\ \mathbf{S}_S^\Lambda &= \begin{bmatrix} -\frac{1}{2}m_S^\parallel \\ i\gamma\frac{1}{2}m_S^\parallel \\ 0 \end{bmatrix} \\ \mathbf{m}_S &= \begin{bmatrix} -m_S^\parallel \cos(2\pi k_{\text{hel}}R_n) \\ m_S^\parallel \gamma \sin(2\pi k_{\text{hel}}R_n) \\ m_S^\perp \end{bmatrix} \end{aligned} \quad (\text{B1})$$

$$\begin{aligned} \mathbf{S}_L^\Gamma &= \begin{bmatrix} 0 \\ 0 \\ -\frac{1}{2}m_L^\perp \end{bmatrix} \\ \mathbf{S}_L^\Lambda &= \begin{bmatrix} \frac{1}{2}m_L^\parallel (\cos(\frac{\pi k_{\text{hel}}}{3}) - i \sin(\frac{\pi k_{\text{hel}}}{3})) \\ \frac{1}{2}i\gamma m_L^\parallel (\cos(\frac{\pi k_{\text{hel}}}{3}) - i \sin(\frac{\pi k_{\text{hel}}}{3})) \\ 0 \end{bmatrix} \\ \mathbf{m}_L &= \begin{bmatrix} m_L^\parallel \cos(\frac{1}{3}\pi k_{\text{hel}}(6R_n + 1)) \\ m_L^\parallel \gamma \sin(\frac{1}{3}\pi k_{\text{hel}}(6R_n + 1)) \\ -m_L^\perp \end{bmatrix} \end{aligned} \quad (\text{B2})$$

The magnetic interaction vectors for the three reflections are:

$$\begin{aligned}\mathbf{M}_{-k} &= \begin{bmatrix} -\frac{1}{2}(m_L^{\parallel} + m_S^{\parallel}) \\ \frac{1}{2}i\gamma(m_L^{\parallel} - m_S^{\parallel}) \\ 0 \end{bmatrix} \\ \mathbf{M}_{+k} &= \begin{bmatrix} -\frac{1}{2}(m_L^{\parallel} + m_S^{\parallel}) \\ -\frac{1}{2}i\gamma(m_L^{\parallel} - m_S^{\parallel}) \\ 0 \end{bmatrix} \\ \mathbf{M}_3 &= \begin{bmatrix} 0 \\ 0 \\ m_S^{\perp} - m_L^{\perp} \end{bmatrix}\end{aligned}\quad (\text{B3})$$

2. TC—Standard block model

The standard TC model has two Fourier components at the Γ and Z points of the Brillouin zone, where $\mathbf{k}_Z = (0, 0, 1.5)$. The main fan angle of the L block is ϕ , while ψ denotes the out-of-plane lifting angle of the S block.

$$\begin{aligned}\mathbf{S}_S^{\Gamma} &= \begin{bmatrix} -\frac{1}{2}m_S \cos \psi \\ 0 \\ 0 \end{bmatrix} \\ \mathbf{S}_S^Z &= \begin{bmatrix} 0 \\ 0 \\ \frac{1}{2}m_S \sin \psi \end{bmatrix} \\ \mathbf{m}_S &= \begin{bmatrix} -m_S \cos \psi \\ 0 \\ m_S \cos(3\pi R_n) \sin \psi \end{bmatrix} \\ \mathbf{S}_L^{\Gamma} &= \begin{bmatrix} \frac{1}{2}m_L \cos \phi \\ 0 \\ 0 \end{bmatrix} \\ \mathbf{S}_L^Z &= \begin{bmatrix} 0 \\ \frac{1}{2}m_L \sin \phi \\ 0 \end{bmatrix} \\ \mathbf{m}_L &= \begin{bmatrix} m_L \cos \phi \\ m_L \cos(3\pi R_n) \sin \phi \\ 0 \end{bmatrix}\end{aligned}\quad (\text{B4})$$

$$\mathbf{m}_L = \begin{bmatrix} m_L \cos \phi \\ m_L \cos(3\pi R_n) \sin \phi \\ 0 \end{bmatrix}\quad (\text{B5})$$

The magnetic interaction vectors for the three reflections are:

$$\begin{aligned}\mathbf{M}_{1.5} &= \begin{bmatrix} 0 \\ im_L \sin \phi \\ m_S \sin \psi \end{bmatrix} \\ \mathbf{M}_{4.5} &= \begin{bmatrix} 0 \\ -im_L \sin \phi \\ m_S \sin \psi \end{bmatrix} \\ \mathbf{M}_3 &= \begin{bmatrix} m_L \cos \phi - m_S \cos \psi \\ 0 \\ 0 \end{bmatrix}\end{aligned}\quad (\text{B6})$$

3. m-TC—Modified block model

The modified TC model, described in some detail in the main text, has the same Fourier components of the standard

TC model. The S lifting angle is modulated by ψ_1 with modulation wave vector \mathbf{k}_Z . The L block is split into ‘lower’ L1 and ‘upper’ L2 blocks, each modulated by ϕ_1 in opposite direction and with modulation wave vector \mathbf{k}_Z . We obtain

$$\begin{aligned}\mathbf{S}_S^{\Gamma} &= \begin{bmatrix} -\frac{1}{2}m_S \cos \psi \cos \psi_1 \\ 0 \\ \frac{1}{2}m_S \cos \psi \sin \psi_1 \end{bmatrix} \\ \mathbf{S}_S^Z &= \begin{bmatrix} \frac{1}{2}m_S \sin \psi \sin \psi_1 \\ 0 \\ \frac{1}{2}m_S \sin \psi \cos \psi_1 \end{bmatrix} \\ \mathbf{m}_S &= \begin{bmatrix} m_S (\cos(3\pi R_n) \sin \psi \sin \psi_1 - \cos \psi \cos \psi_1) \\ 0 \\ m_S (\cos(3\pi R_n) \sin \psi \cos \psi_1 + \cos \psi \sin \psi_1) \end{bmatrix}\end{aligned}\quad (\text{B7})$$

$$\begin{aligned}\mathbf{S}_{L1}^{\Gamma} &= \begin{bmatrix} \frac{1}{4}m_L \cos \phi \cos \phi_1 \\ -\frac{1}{4}m_L \cos \phi \sin \phi_1 \\ 0 \end{bmatrix} \\ \mathbf{S}_{L1}^Z &= \begin{bmatrix} \frac{1}{4}m_L \sin \phi \sin \phi_1 \\ \frac{1}{4}m_L \sin \phi \cos \phi_1 \\ 0 \end{bmatrix} \\ \mathbf{m}_{L1} &= \begin{bmatrix} \frac{1}{2}m_L (\cos(3\pi R_n) \sin \phi \sin \phi_1 + \cos \phi \cos \phi_1) \\ \frac{1}{2}m_L (\cos(3\pi R_n) \sin \phi \cos \phi_1 - \cos \phi \sin \phi_1) \\ 0 \end{bmatrix}\end{aligned}\quad (\text{B8})$$

$$\begin{aligned}\mathbf{S}_{L2}^{\Gamma} &= \begin{bmatrix} \frac{1}{4}m_L \cos \phi \cos \phi_1 \\ \frac{1}{4}m_L \cos \phi \sin \phi_1 \\ 0 \end{bmatrix} \\ \mathbf{S}_{L2}^Z &= \begin{bmatrix} -\frac{1}{4}m_L \sin \phi \sin \phi_1 \\ \frac{1}{4}m_L \sin \phi \cos \phi_1 \\ 0 \end{bmatrix} \\ \mathbf{m}_{L2} &= \begin{bmatrix} \frac{1}{2}m_L (-\cos(3\pi R_n) \sin \phi \sin \phi_1 + \cos \phi \cos \phi_1) \\ \frac{1}{2}m_L (\cos(3\pi R_n) \sin \phi \cos \phi_1 + \cos \phi \sin \phi_1) \\ 0 \end{bmatrix}\end{aligned}\quad (\text{B9})$$

From this, by performing explicit dot products, we can easily deduce that the structure has constant moments as a function of R_n , and that the angle between L1 and L2 is $2\phi_1$, and is also unmodulated. Moreover, in order for the angles between S and L1 in the same unit cell and between L2 and S in adjacent unit cell ($\Delta R_n = 1/3$) to be the same, Eq. (16) must be fulfilled, thereby defining a relation between ψ_1 and ϕ_1 . The magnetic interaction vectors for the three reflections are:

$$\begin{aligned}\mathbf{M}_{1.5} &= \begin{bmatrix} m_S \sin \psi \sin \psi_1 \\ im_L \sin \phi \cos \phi_1 \\ m_S \sin \psi \cos \psi_1 \end{bmatrix} \\ \mathbf{M}_{4.5} &= \begin{bmatrix} m_S \sin \psi \sin \psi_1 \\ -im_L \sin \phi \cos \phi_1 \\ m_S \sin \psi \cos \psi_1 \end{bmatrix} \\ \mathbf{M}_3 &= \begin{bmatrix} m_L \cos \phi \cos \phi_1 - m_S \cos \psi \cos \psi_1 \\ 0 \\ m_S \cos \psi \sin \psi_1 \end{bmatrix}\end{aligned}\quad (\text{B10})$$

APPENDIX C: $F^{(2)}$ FOR THE TC PHASE

In this section, we will demonstrate that the $F^{(2)}$ term cannot produce the observed linear asymmetry for either the (0,0,3) or the (0,0,4.5) peaks. We need to compute the two magnetic interaction tensors, one for each reflection. We have:

$$T_0^{\alpha\beta} = 4 \begin{pmatrix} m_{Lx}^2 + m_{Sx}^2 & 0 & 0 \\ 0 & m_{Ly}^2 & 0 \\ 0 & 0 & m_{Sz}^2 \end{pmatrix}$$

$$T_k^{\alpha\beta} = 4 \begin{pmatrix} 0 & \pm im_{Lx}m_{Ly} & m_{Sx}m_{Sz} \\ \pm im_{Lx}m_{Ly} & 0 & 0 \\ m_{Sx}m_{Sz} & 0 & 0 \end{pmatrix} \quad (C1)$$

where $m_{Lx} = m_L \cos \phi$ etc., and the \pm is for the (0,0,1.5) and (0,0,4.5) peaks, respectively.

We can immediately see that the term $T_0^{\alpha\beta}$ at the Γ point is diagonal and therefore cannot contribute to scattering in the rotated channels (it is of the same form as an anisotropic Thomson scattering). The term $T_k^{\alpha\beta}$ is completely off-diagonal and does produce scattering in the rotated channels, but it produces no asymmetry. To show this, we calculate the scattering cross section for the $\sigma\pi'$ channel as an example:

$$I_{\sigma\pi'} = |i \sin \theta (F^{(1)}m_{Sz} \mp F^{(2)}m_{Lx}m_{Ly})|^2$$

$$= \sin^2 \theta (|F^{(1)}|^2 m_{Sz}^2 + |F^{(2)}|^2 m_{Lx}^2 m_{Ly}^2 + 2\Re(F^{(1)}\overline{F^{(2)}}m_{Sz}m_{Lx}m_{Ly})). \quad (C2)$$

It is easy to see that this term does not change by rotation of the TC structure by 180° around the z axis.

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