Vector analysis of electric-field-induced antiparallel magnetic domain evolution in ferromagnetic/ferroelectric heterostructures

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Abstract: Electric field (E-field) control of magnetism based on magnetoelectric coupling is one of the promising approaches for manipulating the magnetization with low power consumption. The evolution of magnetic domains under *in-situ* E-fields is significant for the practical applications in integrated micro/nano devices. Here, we report the vector analysis of the E-field-driven antiparallel magnetic domain evolution in FeCoSiB/PMN-PT(011) multiferroic heterostructures via *in-situ* quantitative magneto-optical Kerr microscope. It is demonstrated that the magnetic domains can be switched to both the 0° and 180° easy directions at the same time by E-fields, resulting in antiparallel magnetization distribution in ferromagnetic/ferroelectric heterostructures. This antiparallel magnetic domain evolution is attributed to energy minimization with the uniaxial strains by E-fields which can induce the rotation of domains no more than 90°. Moreover, domains can be driven along only one or both easy axis directions by reasonably selecting the initial magnetic domain distribution. The vector analysis of magnetic domain evolution can provide visual insights into the strain-mediated magnetoelectric effect, and promote the fundamental understanding of electrical regulation of magnetism.

Keywords: multiferroics; magnetoelectric effect; magnetic domains; magneto-optical Kerr effect (MOKE)

1 Introduction

The efficient manipulation of magnetic domains in

ferromagnetic films exhibits significant applications in magnetic memory and logic devices [1,2]. The conventional methods to control the magnetic domains involve the application of a magnetic field (H-field) or a spin-polarized electric current [3–6]. Both of the two methods inevitably utilize a large current to generate the H-field

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or the spin-polarized current, whose high energy dissipation impedes the development of high-density spintronic devices. Therefore, electric field (E-field), rather than electric current, has been proposed as an energy-efficient alternative to manipulate magnetization [7–9]. Significant progress has been made in E-field control of macroscopic magnetic properties of multiferroic heterostructures, including the magnetic anisotropy [10,11], the exchange bias [12,13], the magnetoresistance [14,15], the magnetic permeability [16], and the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction [17], etc. In consideration of the potential applications in micro/nano devices, recent attention has been turned toward the E-field manipulation of micromagnetic elements, such as magnetic domains [18–21]. The reversible electric-field-driven magnetic domain-wall motion [22,23], and E-field control of the domain-wall mobility [24,25], have been demonstrated in different multiferroic heterostructures with simple magnetic domain structures. However, magnetic domain evolution with complex initial magnetization distribution has rarely been investigated by vector imaging techniques in strain-mediated multiferroic heterostructures, which would be useful in understanding the response processes of magnetic domains to E-fields and the development of practical magnetoelectric devices.

Several imaging methods exhibit the ability to extract the magnetization directions of the magnetic domains, such as X-ray magnetic circular dichroism-photoemission electron microscope [21], scanning electron microscope with polarization analysis [26], Lorentz transmission electron microscope [27], and quantitative magneto-optical Kerr effect (MOKE) microscope [28,29]. Despite the relatively low resolution, quantitative MOKE has fewer restrictions on sample preparations and test conditions among the imaging techniques mentioned above. For vector imaging of magnetic domains via quantitative MOKE, calibration functions are necessary in two orthogonal sensitivity directions, which can be calculated based on the Kerr contrast of the measured single domains in saturated H-fields along different directions rotating from 0° to 360° . Then, the magnetization directions of the desired magnetic domains can be acquired by substituting domain patterns into the calibration functions. The deviation of the acquired magnetization directions is less than 5° [30], which is conducive to the investigation on the E-field-driven magnetic domain evolution with domain sizes of tens of micrometers.

In this work, we report the E-field-driven antiparallel magnetic domain evolution in FeCoSiB/PMN–PT(011) multiferroic heterostructures, revealed by an *in-situ* quantitative MOKE. The clearer domain patterns with more regular shape in the FeCoSiB layer are conducive to the analysis of magnetic domain variations by E-fields. With a bias H-field at 30 Oe, the magnetic domains can be induced towards both the 0° and 180° easy axis directions at the same time by E-fields, which is further clarified by pixel-by-pixel comparisons of domain patterns. The E-field-driven propagation of the magnetic domains depends on the energy minimization by the strain-mediated magnetoelectric coupling effect, which should be considered in the design of micro/nano magnetoelectric devices.

2 Material and methods

2.1 Sample preparation

Multilayer films with the structure of Cu (3 nm)/FeCoSiB (44 nm)/Ta (3 nm) were deposited on the 0.3 mm thick pre-polarized PMN–PT(011) piezoelectric single crystal substrates via dc magnetron sputtering at room temperature. The base pressure was below 1.5×10^{-7} Torr and the working Ar pressure was 3 mTorr. The back of the substrate was covered with a 20 nm Ta layer as the bottom electrode.

2.2 Experiment characterization

E-fields were applied through the thickness direction of the PMN–PT substrates by using a Keithley 6517B electrometer. Before tests, the sample was pre-polarized by an E-field of 16.67 kV/cm to induce the polarization along downwards in the out-of-plane direction. And during the electrical control, the applied E-fields were maintained in the same polarity with the polarization still pointing downwards. Magnetic domain patterns were measured by quantitative MOKE microscope (Evico Magnetics, em-Kerr-Highres, Germany) integrated with a commercial software that could analyze the magnetization directions of magnetic domains. The magnetization directions could be acquired by substituting the desired domains into the calculated calibration functions. The domains with different magnetization directions were marked with colors according to the color wheel. The variation angles of magnetic domains were acquired via pixel-by-pixel

comparisons between two domain patterns with and without E-fields.

3 Results and discussion

Figure 1(a) shows the configuration of the FeCoSiB/PMN-PT(011) multilayers, where E-fields are applied through the thickness direction of the PMN-PT(011) substrates. The magnetic hysteresis (M - H) loop with H-fields in the $[01-1]$ direction measured by MOKE is illustrated in Fig. 1(b), and orange stars and numbers are utilized to mark the H-fields at which the corresponding magnetic domain patterns are displayed in Fig. 1(c).

Due to the complexity of magnetization distribution, angles from 0° to 360° rotating counterclockwise are defined as the relative orientation of the magnetizations, as shown in the left of Fig. 1(c). In detail, magnetization directions of the magnetic domains under positive and negative saturated H-fields along the $[100]$ direction of PMN-PT(011) substrates are defined as 0° and 180° , and those in the $[01-1]$ direction are set as 90° and 270° , respectively. Additionally, the orientation of the PMN-PT(011) substrates corresponding to the defined angles is also displayed in the left as a supplement.

Figure 1(c) illustrates the magnetization reversal process when H-fields are parallel to the $[01-1]$ direction. The numbers mark the values of H-fields which are consistent with that in Fig. 1(b). By gradually increasing H-fields from -500 to -50 Oe, the magnetic domains rotate clockwise where the multi-domain state replaces the single-domain state along 270° . At the H-field of 0 Oe, ripple-like magnetic domains are acquired with a complex distribution of magnetizations, where it exists only negative magnetization components in the $[01-1]$ direction. When the H-field is ascended to 10 Oe, magnetic domains near 135° emerge and partial domains rotate towards 180° , resulting in both positive and negative magnetization components along the $[01-1]$ direction. At 15 Oe, the proportion of domains in 135° further increases, and magnetic domains along 45° (the green part) can also be found in the field of view. With the H-field of 30 Oe, all magnetization components are along the $[01-1]$ direction. Then the increase of H-fields induces the magnetic domains approximately in 45° to propagate in the form of domain-wall motion, accompanied by the rotation of magnetic domains until the single-domain state is restored where magnetic domains are parallel to 90° (the current external field direction). Additionally, the magnetization reversal process and the corresponding M - H loop with H-fields

Fig. 1 Schematic of multiferroic heterostructures and characterization of basic magnetic properties with H-fields along the $[01-1]$ direction. (a) Structures of FeCoSiB/PMN-PT(011) multilayers. (b) M - H loop measured by MOKE. (c) Dynamic magnetization reversal with H-fields from -500 to 500 Oe.

in the [100] direction are shown in Fig. S1 in the Electronic Supplementary Material (ESM), where the complex ripple-like magnetic domains suddenly rotate to mainly along 0° with H-fields increasing from 5 to 10 Oe.

M – H loops under different E-fields with H-fields lying in the [01-1] direction are shown in Fig. 2(a). It is revealed that E-fields make the initial hard axis direction ([01-1] direction) even harder to be magnetized. The normalized gray level values are equivalent to the ratios of magnetization to saturation magnetization (M/M_s). Furthermore, the values of M/M_s at different H-fields are extracted from the M – H loops with H-fields increased from -300 to 300 Oe and plotted as a function of the applied E-fields in Fig. 2(b). The approximately linear decrease of the absolute values of M/M_s with E-fields

along the hard axis direction suggests the switching of magnetizations towards the easy axis [31].

Figures 2(c)–2(e) illustrate the corresponding partial dynamic magnetization processes under various E-fields between 0 and 50 Oe when the H-fields are ascended from -300 to 300 Oe along the [01-1] direction. Under each fixed E-field, it is generally observed that the increase of H-fields induces the nucleation and elongation of magnetic domains near 45° (i.e., green colored region). Nevertheless, the H-fields required for the nucleation of magnetic domains in 45° ascend with the increase of E-fields. To be specific, for the case without E-fields, it already exists magnetic domains along 45° at 30 Oe. For the cases with E-fields at 6.67 and 10 kV/cm, the magnetic domains in this direction can only be acquired at 40 and 50 Oe, respectively.

Fig. 2 M – H loops and dynamic magnetization processes at different E-fields with H-fields parallel to the [01-1] direction via quantitative MOKE. (a) *In-situ* E-field control of M – H loops. (b) Values of the normalized graylevel at various H-fields as a function of E-fields extracted from the M – H loops. (c–e) Magnetic domain evolution with H-fields at 0, 6.67, and 10 kV/cm, respectively.

Furthermore, the domain components along the [01-1] direction at the same H-fields in different magnetization processes are descended with the increase in E-fields, which corresponds to the decrease of the absolute values of M/M_s . When H-fields are parallel to the [100] direction, the M - H loops and magnetization processes at different E-fields are illustrated in Fig. S2 in the ESM, where domains at the same H-fields are driven towards the single easy axis directions as the E-fields ascend.

Then, the *in-situ* magnetic domain evolution under E-fields is investigated at a fixed H-field. To exclude the environmental disturbance on magnetic domain patterns, domains at 0 and 5 min are imaged at various H-fields along the [01-1] direction, as shown in Fig. S3 in the ESM, which confirm that magnetic domains do not change with time. Therefore, the obvious variations in magnetic domain patterns observed under E-fields are caused by the strain-mediated magnetoelectric coupling effect.

Figures 3(a)–3(d) illustrate E-field control of magnetic domain propagation with the bias H-field keeping constant at 30 Oe, and the corresponding variation angles of domains at different E-fields via pixel-by-pixel comparisons are shown in Figs. 3(e)–3(g). Before the E-fields applied, the H-fields were increased from -300 to 30 Oe along the [01-1] direction. During the electrical control, the H-field of 30 Oe remains unchanged. Figure 3(a) displays the initial magnetization distribution ranging between 44° and 167° at 0 kV/cm, with both positive and negative magnetization components along the [100] direction. Considering the complex initial magnetization distribution, two

representative regions are selected and marked as Region 1 and Region 2. Region 1 is the upper part of the field of vision where magnetization directions are mainly between 44° and 81° , and Region 2 is the stripe-like area in the middle where magnetization directions are along 92° – 157° .

When an E-field of 3.33 kV/cm is applied as shown in Figs. 3(b) and 3(e), the magnetic domains in Region 1 rotate clockwise lying in 20° – 64° where the variation angles of domains between 0 and 3.33 kV/cm are located at -36° to -9° . Meanwhile, in Region 2, the range of magnetization directions is 120° – 180° , and the variation angles are between 5° and 40° . When the E-field continues to ascend to 6.67 kV/cm, magnetic domains in Region 1 are parallel to 1° – 34° via the E-field modulation induced clockwise rotation, as shown in Fig. 3(c). Figure 3(f) displays that the variation angles between 0 and 6.67 kV/cm are located between -60° and -28° . At the same time, in Region 2, magnetization directions are along 145° – 187° via anticlockwise rotation with the variation angles between 13° and 69° .

With the increase of the E-field to 10 kV/cm, most of the magnetic domains are driven along either the 0° or 180° easy axis directions, as shown in Fig. 3(d). The corresponding variations of the magnetization directions in Fig. 3(g) show significantly different behavior between Region 1 and Region 2. In Region 1, domains are lying in -24° to -9° with most of the magnetization directions are close to 0° easy axis direction via the clockwise rotation induced by the E-field and the switching angles of domains between 0 and 10 kV/cm are within -76° to -33° . In contrast, in Region 2,

Fig. 3 *In-situ* E-field induced antiparallel magnetization distribution with the bias H-field at 30 Oe. (a–d) Magnetic domain evolution with the increase of the E-field. Numbers are utilized to mark the representative regions. (e–g) Magnetization variation angles via pixel-by-pixel comparisons of domains at 3.33, 6.67, and 10 kV/cm with that at 0 kV/cm, respectively. (h) Ratios of domains along the 0° easy axis direction in Region 1 (green line) and 180° easy axis direction in Region 2 (purple line), respectively.

domains are driven lying in 159° – 193° , around the 180° easy axis direction where the range of domain change is in 22° – 82° . Figure 3(h) displays that the applied E-fields induce the ratios of domains lying around the 0° easy axis direction (-10° to 10°) in Region 1 from 0% at 0 kV/cm to 59% at 10 kV/cm, and drive that around the 180° easy axis direction (170° – 190°) in Region 2 from 2% to 62% simultaneously, which means that the electrical modulation can realize antiparallel magnetic domain evolution in multiferroic heterostructures. For the whole area of view, when the E-fields are ascended from 0 to 10 kV/cm, the electrical modulation drives the ratios of domains around the 0° and 180° direction increasing from 0% to 40% and 1% to 19%, respectively.

The antiparallel magnetic domain evolution is attributed to the requirement of lowest energy for magnetization rotation. According to the previous reports [32,33], the magnetoelastic energy reflecting the coupling between the magnetization and elastic strains, is one of the energy contributions to the total free energy in strain-mediated multiferroic heterostructures. The magnetoelastic energy density can be expressed as [33,34]:

$$f_{me} = \frac{1}{2} c_{ijkl} (\varepsilon_{ij} - \varepsilon_{ij}^0) (\varepsilon_{kl} - \varepsilon_{kl}^0) \quad (1)$$

where c is the elastic stiffness coefficient of the magnetic layer, and ε and ε^0 denote the total strain and eigenstrain from the spontaneous deformation, respectively. For the FeCoSiB/PMN–PT(011) multiferroic heterostructures, the strains in the ferroelectric phase induced by the applied E-fields can transfer to the ferromagnetic phase through the interface between them. Hence, the total strain ε would be changed by the piezo-strains under E-fields, resulting in the increase of the magnetoelastic energy of the FeCoSiB layer. To meet the minima of the total free energy, the eigenstrain ε^0 would correspondingly change to reduce the magnetoelastic energy. Thereinto, ε^0 is expressed as [33]:

$$\varepsilon_{ij}^0 = \frac{3}{2} \lambda_s \left(m_i m_j - \frac{1}{3} \delta_{ij} \right) \quad (2)$$

where λ_s is the saturation magnetostriction coefficient, m is the local magnetization vector, and δ_{ij} denotes Kronecker delta function. Thus, the local magnetization vectors may rotate under E-fields to seek the energy minima.

In the FeCoSiB/PMN–PT(011) heterostructures, m_3

is zero, and the shear deformation ε_{12} is assumed to be zero. After omitting term independent of m , the magnetoelastic energy can be expressed as

$$f_{me} = -\frac{3}{2} \lambda_s (c_{11} - c_{22}) (\varepsilon_{11} - \varepsilon_{22}) m_1^2 \quad (3)$$

that is [35,36],

$$f_{me} = K_{me} \sin^2 \theta_{[100]} \quad (4)$$

where $\theta_{[100]}$ denotes the angle between magnetization vectors and the $[100]$ direction, that is, the directions of magnetic domains as mentioned above where the $[100]$ direction is set as 0° . K_{me} is defined as a magnetoelastic factor:

$$K_{me} = -\frac{3}{2} \lambda_s Y (\varepsilon_{[01-1]} - \varepsilon_{[100]}) \quad (5)$$

where Y is the Young's modulus of FeCoSiB, and $\varepsilon_{[01-1]} - \varepsilon_{[100]}$ is the total effective strain. In the FeCoSiB layer, $\lambda_s = -25$ ppm [37] and $Y = 150$ GPa [38]. The total effective strain $\varepsilon_{[01-1]} - \varepsilon_{[100]}$ is directly proportional to the applied E-fields. As an E-field of 5 kV/cm is imposed on the PMN–PT(011) substrate, $\varepsilon_{[01-1]} - \varepsilon_{[100]}$ is equal to 583 ppm [15]. Thereby, K_{me} is calculated as the value of 3.3 kJ/m³ under 5 kV/cm. With the increase of the E-field to 8 kV/cm, $\varepsilon_{[01-1]} - \varepsilon_{[100]}$ is ascended to approximately 1000 ppm [15], and K_{me} correspondingly changes to 5.6 kJ/m³.

Therefore, in the FeCoSiB/PMN–PT(011) heterostructure, θ as 0° and 180° is energetically more favorable, which corresponds to the experimental results. Considering that the magnetoelastic energy is an even function of m from Eqs. (1) and (2), the 0° direction and 180° direction are equivalent in terms of energy. Within the domain rotation, the magnetization vectors cannot reach the minimum energy via crossing the point of the highest energy. Therefore, domains initially along 0° – 90° will rotate towards the 0° direction, and that in 90° – 180° to the 180° direction driven by the applied E-fields. In other words, E-fields can induce at most 90° magnetization rotation. Obviously, the key of antiparallel or parallel magnetic domain evolution by E-fields is the initial magnetization distribution at 0 kV/cm. For the case with the H-field at 30 Oe, on the basis of the initial magnetization between 44° and 167° , magnetic domains can be driven along both the 0° and 180° directions at the same time via the electrical modulation. And the range of H-fields to achieve antiparallel magnetic domain evolution is between 20 and 35 Oe with differences in the ratios along the 0°

and 180° directions.

In order to further clarify the antiparallel magnetic domain evolution by E-fields, Fig. 4 illustrates that E-fields induce overall magnetic domains to rotate to only one easy axis direction, 0° or 180°. With the bias H-field of 0 Oe, the heterostructures exhibit complex ripple-like magnetic domains along 168°–262° at 0 kV/cm, as shown in Fig. 4(a). The applied E-fields induce domains to rotate towards the 180° easy axis direction and when the E-field is ascended to 10 kV/cm, domains are driven along 170°–210°. At the initial state with 0 kV/cm under 60 Oe, most magnetic domains are in 50°–90°, as shown in Fig. 4(b). Under the electrical modulation, domains are induced rotating towards the 0° easy axis direction, and with the E-field increased to 10 kV/cm, magnetization directions range between 0° and 45°. The pixel-by-pixel comparisons of magnetic domains at 10 and 0 kV/cm with the bias H-fields of 0 and 60 Oe can be seen in Figs. 4(c) and 4(d), respectively, where variation angles of domains mainly locate between –65° and 12° at 0 Oe and –85° to –28° at 60 Oe, respectively. Figure 4(e) illustrates that electrical modulation can induce the ratios of domains lying around the 180° easy axis direction (170°–190°) from 13% at 0 kV/cm to 43% at 10 kV/cm under 0 Oe,

and drive that around the 0° easy axis direction (–10° to 10°) from 0% to 16% under 60 Oe.

4 Conclusions

In summary, the vector analysis of antiparallel magnetic domain evolution under *in-situ* E-fields was demonstrated in FeCoSiB/PMN–PT(011) multiferroic heterostructures by quantitative MOKE. At the bias H-field of 30 Oe, magnetic domains were driven to simultaneously align in both the 0° and 180° easy axis directions by E-fields, on the basis of the initial magnetization directions between 44° and 167°. In the selected representative regions, under the electrical modulation of 10 kV/cm, 59% domains clockwise rotated lying in –10° to 10° with variation angles between –76° and –33° in Region 1, and via anticlockwise rotation, 62% domains were located in the range of 170°–190° with variation angles in 22°–82° in Region 2. Meanwhile, E-field induced overall magnetic domains towards only one easy axis direction could be realized via the rational selection of initial magnetic domain patterns. For the case at 0 Oe that the initial magnetization directions located in the range of 168°–

Fig. 4 (a, b) *In-situ* E-field modulation of overall magnetic domains towards only the 180° easy axis direction at 0 Oe, and 0° easy axis direction at 60 Oe. (c, d) Pixel-by-pixel comparisons of magnetic domains at 10 and 0 kV/cm under 0 and 60 Oe, respectively. It should be highlighted that all tests remained in the same area. (e) Ratios of domains around the 180° easy axis direction at 0 Oe and the 0° easy axis direction at 60 Oe, respectively.

262°, domains were driven to along the 180° easy axis direction, and based on domains along 50°–90° initially at 60 Oe, electrical control could lead domains rotating towards the 0° easy axis direction. The different magnetic domain evolution paths under E-fields were attributed to the requirement of lowest energy for magnetization rotation in multiferroic heterostructures, which provided visual insights into the strain-mediated magnetoelectric coupling effect that is significant for advancing the fundamental understanding of the electrical modulation of magnetism.

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Electronic Supplementary Material

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