

Microscopic study of thermally and electrically driven magnetic changes in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films on BaTiO_3 substrates

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Epitaxial films of the ferromagnetic manganite $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ on substrates of the ferroelectric perovskite BaTiO_3 are known to display sharp magnetic changes and large magnetoelectric effects when the film is strained by the substrate undergoing thermally driven structural transitions and ferroelectric domain switching, respectively. However, only a single component of the in-plane magnetization has been hitherto imaged. Here we present magnetic vector maps—obtained from photoemission electron microscopy images with magnetic contrast from x-ray magnetic circular dichroism—to show that the electrically and thermally driven changes of local and global magnetization are deterministically influenced by the state of the substrate while also being complex and sample dependent. Our findings, supported by ferromagnetic resonance data and vibrating sample magnetometry, reveal that the behavior of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films on BaTiO_3 substrates is not well predicted from knowledge of each system, probably due to long-range strain between BaTiO_3 domains. In the future, it would be interesting to reduce complexity by patterning the film into regions between which magnetic communication is negligible.

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I. INTRODUCTION

The magnetization of thin films can be thermally and electrically controlled via strain from ferroelectric substrates in multiferroic heterostructures that display magnetoelectric coupling [1] due to strain [2–16], charge [17–20], or exchange bias [18–20]. Studies of these systems typically involve macroscopic magnetic measurements [21–24], while magnetic imaging is possible using second-harmonic generation [25–27], magnetic force microscopy [28], and photoemission electron microscopy (PEEM) [29–31]. The latter technique permits antiferromagnetism and ferroelectricity to be imaged via x-ray linear dichroism (XLD) contrast [29,31], whereas ferromagnetism can be imaged using x-ray magnetic circular dichroism (XMCD) [30,31].

Epitaxial films of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) on ferroelectric substrates of BaTiO_3 (BTO) represent a model system (LSMO/BTO) in which strain-mediated coupling at a large planar interface yields large jumps of film magnetization when there are structural phase transitions in the substrate [21]

and the magnetoelectric coupling of up to $2.3 \times 10^{-7} \text{ s m}^{-1}$ is large [24]. Although PEEM with XMCD contrast has been used to image the local film magnetization after varying temperature and electric field in order to change the orientation/phase of BTO domains and thus modify local film magnetization [16], each image only presented a single component of magnetization, such that one cannot identify thermally and electrically induced changes in the direction and magnitude of the in-plane magnetization vector.

Here we present magnetic vector maps of in-plane LSMO magnetization by combining XMCD-PEEM images obtained while the grazing-incident beam impinges upon the sample before and after a 90° in-plane rotation. In support, we also present data from ferromagnetic resonance (FMR) and vibrating sample magnetometry (VSM). Our magnetic vector maps were acquired after varying the local magnetic anisotropy in the LSMO films via BTO substrate strain due to either thermally driven structural transitions or electrically driven ferroelectric domain switching. Comparing the magnetic vector maps with macroscopic magnetization data confirms the role of substrate microstructure on the macroscopic magnetic properties of the films. However, our findings reveal a hitherto unexpected complexity that we attribute to long-range strain between BTO domains.

II. METHODS

LSMO films were grown at 1048 K in 15 Pa flowing oxygen ambient by pulsed laser deposition ($\lambda = 248 \text{ nm}$, 8 cm target-substrate distance, 1.7 J cm^{-2}) on one-side-polished

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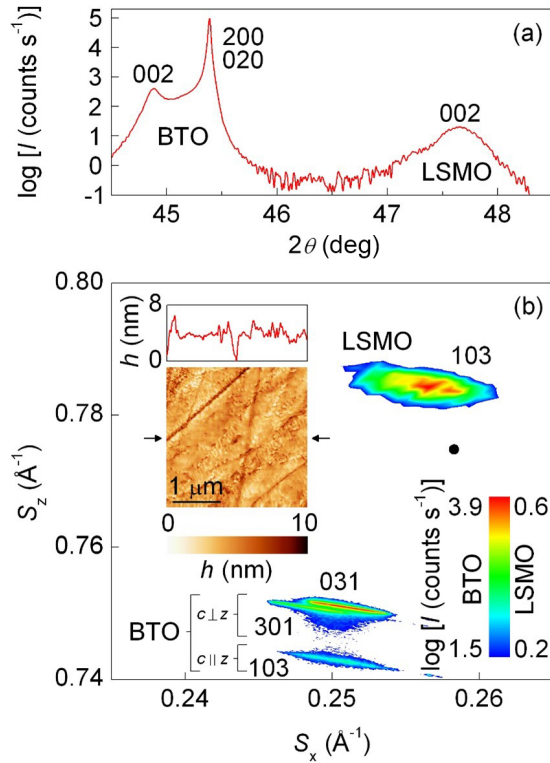


FIG. 1. LSMO/BTO structure at room temperature. (a) High-resolution XRD 2θ - ω scan showing out-of-plane reflections for BTO (200, 020, and 002) and pseudocubic LSMO (002). (b) High-resolution XRD reciprocal space map showing BTO reflections from three domains (031, 301, and 103) and a broad reflection for pseudocubic LSMO (103). The 103 pseudocubic reflection of bulk LSMO (•) is identified from the unit cell ($a = 5.5085 \text{ \AA}$ and $c = 13.37171 \text{ \AA}$, $R\bar{3}c$) [32]. Reciprocal lattice units S_x and S_z correspond to inverse lattice spacings. Inset: AFM image of the LSMO surface, and transect between arrows.

BTO substrates ($4 \text{ mm} \times 4 \text{ mm} \times 0.5 \text{ mm}$). The substrates were received unpoled and fixed with silver dag to the heater block in the deposition chamber. Prior to growth, the substrates were annealed for one hour at 1023 K. After deposition, the samples were annealed for 30 min at 973 K in an oxygen ambient of 55 kPa. Sample temperature was subsequently reduced to room temperature at -10 K min^{-1} . Electric fields were applied across the substrate between the LSMO film and a sputter-deposited Pt back electrode.

Film lattice parameters and epitaxy were studied by x-ray diffraction (XRD) using a Philips X'Pert high-resolution diffractometer (Cu $K\alpha$ radiation) with a primary monochromator. A film thickness of 55 nm was determined via x-ray fringes using macroscopically flat films that were codeposited on SrTiO_3 (001) substrates. Film topography was studied with a Digital Instruments Nanoscope III atomic force microscope (AFM) operated in tapping mode. Film magnetization was measured using a Princeton Measurements Corporation VSM, after removing silver dag with emery paper to prevent spurious magnetic signals from material that is transferred from the heater block to the samples. Mass-normalized magnetization data are presented after correcting for diamagnetic

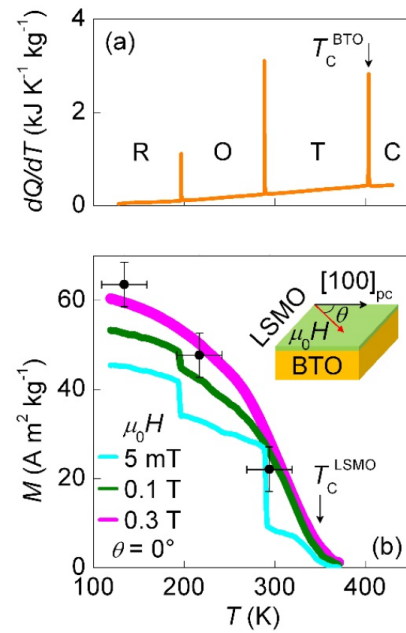


FIG. 2. Thermally driven magnetic and structural transitions in LSMO/BTO. (a) Differential endothermic heat dQ/dT measured in zero applied magnetic field on heating. (b) Magnetization M versus temperature T measured on heating at constant values of magnetic field H that are small, medium, and large. The measurement axis and field direction lie along a $[100]_{pc}$ direction of both materials. Magnetic jumps near $T_{R-O} \sim 200 \text{ K}$ and $T_{O-T} \sim 300 \text{ K}$ occur below the film Curie temperature of $T_C^{\text{LSMO}} \sim 350 \text{ K}$. The three black data points were obtained by choosing a single factor to scale the average magnitude of the magnetization in the Figs. 3(b), 3(e), and 3(h) maps.

background contributions (film mass was calculated assuming the bulk LSMO density of 6.4 g cm^{-3}). The structural phase transitions in the BTO substrate were studied using a TA Instruments DSC Q2000 differential scanning calorimeter (DSC) with a ramp rate of $+10 \text{ K min}^{-1}$. In-plane and out-of-plane FMR measurements were carried out between 134 K and 294 K using an X-band ELEXSYS E500 EPR spectrometer operated at 9.44 GHz.

PEEM with XMCD contrast was performed on beamline I06 at Diamond Light Source with the incident x-ray beam at a grazing incidence of 16° , using an Elmitec SPELEEM-III microscope to image the local zero-field magnetization to a probe depth of $\sim 7 \text{ nm}$ and a lateral resolution of $\sim 50 \text{ nm}$. Each image pixel gives the XMCD asymmetry $(I^+ - I^-)/(I^+ + I^-)$ and is proportional to the projection of the local magnetization onto the incident-beam polarization vector. Here, $I^\pm = I_{\text{res}}^\pm/I_{\text{off}}^\pm$ is the ratio of the secondary electron intensity arising from x-ray absorption on $(I_{\text{res}}^\pm)$ and off $(I_{\text{off}}^\pm)$ the Mn L_3 resonance using right (+) and left (−) circularly polarized light. Vector maps of local magnetization direction, and the corresponding maps of magnetization magnitude, were calculated by vectorially combining for each pixel the XMCD asymmetry from two images obtained before and after rotating the azimuthal angle of the sample by 90° , such that the in-plane projection of the incident-beam direction onto the sample was rotated by 90° .

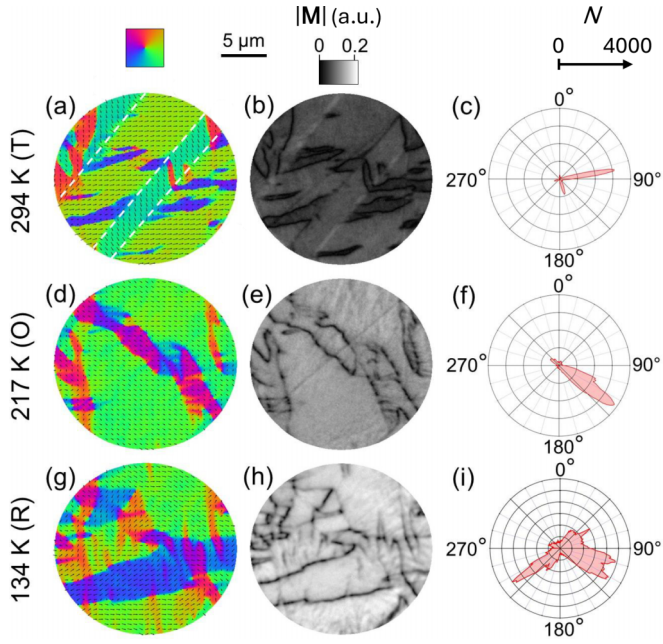


FIG. 3. Thermally driven changes of local magnetization in LSMO/BTO. (a),(d),(g) Vector maps of the in-plane magnetization direction (color wheel) for a single region of the LSMO film. Arrow directions and white dashed lines guide the eye; arrow magnitudes carry no meaning. (b),(e),(h) The corresponding maps showing the magnitude of the magnetization. (c),(f),(i) The corresponding polar plots of magnetization direction, where the shaded area of an infinitesimal angular sector at angle ϕ is proportional to the square of the number of pixels N with magnetization direction ϕ . The BTO substrate is (a),(b),(c) tetragonal (T) at 294 K, (d),(e),(f) orthorhombic (O) at 217 K, and (g),(h),(i) rhombohedral (R) at 134 K. The $[100]_{pc}$ direction in LSMO is roughly vertical.

III. RESULTS

Room-temperature XRD data for LSMO/BTO reveal weak and broad film reflections and confirm the presence of 90° domains in BTO (Fig. 1). The relative population of BTO domains varies between substrates, with (001) domains representing the minority species in all samples studied [e.g., Fig. 1(a)]. A reciprocal space map [Fig. 1(b)] shows that the film is almost fully relaxed and that each type of substrate domain is subject to $<1^\circ$ misorientations. From Fig. 1(a), we find out-of-plane lattice parameters of $a = 3.993(3) \text{ \AA}$ and $c = 4.036(1) \text{ \AA}$ for the a and c domains in BTO, respectively, and we find an out-of-plane lattice parameter of $3.81(1) \text{ \AA}$ for the pseudocubic unit cell of LSMO. The broad LSMO pseudocubic 103 reflection in Fig. 1(b) implies an in-plane lattice parameter of $3.90(5) \text{ \AA}$ for the pseudocubic unit cell of LSMO. By contrast, bulk LSMO has a unit cell ($a = 5.5085 \text{ \AA}$ and $c = 13.37171 \text{ \AA}$, $R\bar{3}c$) [32] that would imply a reflection at the point identified via the symbol $\bullet$ in Fig. 1(b). AFM revealed film surfaces to be smooth with a roughness of $\sim 0.5\text{--}1.0 \text{ nm}$ away from twin boundaries (Fig. 1, inset).

BTO undergoes a well known [21] series of thermally hysteretic structural transitions that we observed via calorimetry on heating [Fig. 2(a)]: it is rhombohedral (R) below 190 K,

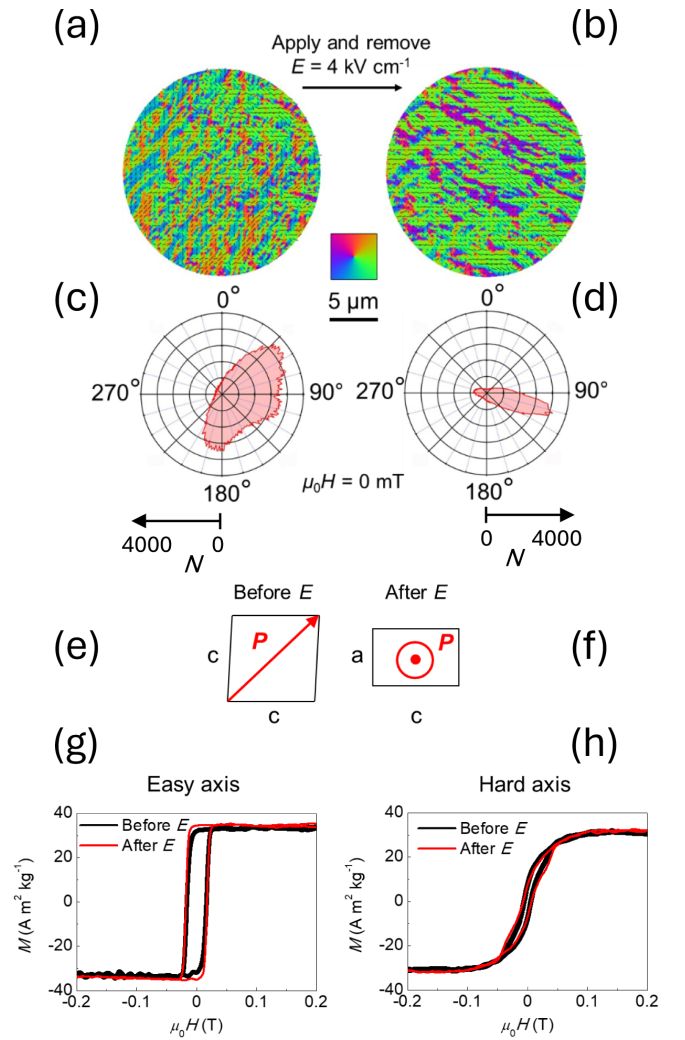


FIG. 4. Magnetoelectric effects in LSMO on O-phase BTO. (a),(b) Vector maps of in-plane LSMO magnetization and (c),(d) the corresponding polar plots of magnetization direction, (a),(c) before and (b),(d) after the application of 4 kV cm^{-1} across the BTO substrate at 217 K. The Fig. 3 caption gives more information about the vector maps and polar plots. (e),(f) Schematics of the BTO surface presented to the film (e) before and (f) after the electric field reorients polarization P from in-plane to out-of-plane [21]. (g), (h) Macroscopic magnetization M versus applied field H along the in-plane (g) easy and (h) hard axes, before (black) and after (red) the application of 4 kV cm^{-1} .

orthorhombic (O) up to 280 K, tetragonal (T) up to the BTO Curie temperature $T_C^{\text{BTO}} \sim 390 \text{ K}$, and cubic (C) at higher temperatures. As shown in Fig. 2(b), the R-O and O-T transitions in BTO, which lie below the LSMO Curie temperature $T_C^{\text{LSMO}} \sim 350 \text{ K}$, produce jumps in film magnetization M versus temperature T provided that a collinear magnetic field is small enough to avoid magnetic saturation ($\mu_0 H \leq 0.1 \text{ T}$); the jumps are suppressed by an applied field of $\mu_0 H = 0.3 \text{ T}$. The jumps in film magnetization arise due to changes of magnetic anisotropy that are driven by strain associated with the phase transitions in the substrate [21].

Magnetic vector maps [Figs. 3(a), 3(d), and 3(g)] and the corresponding polar plots of magnetization direction

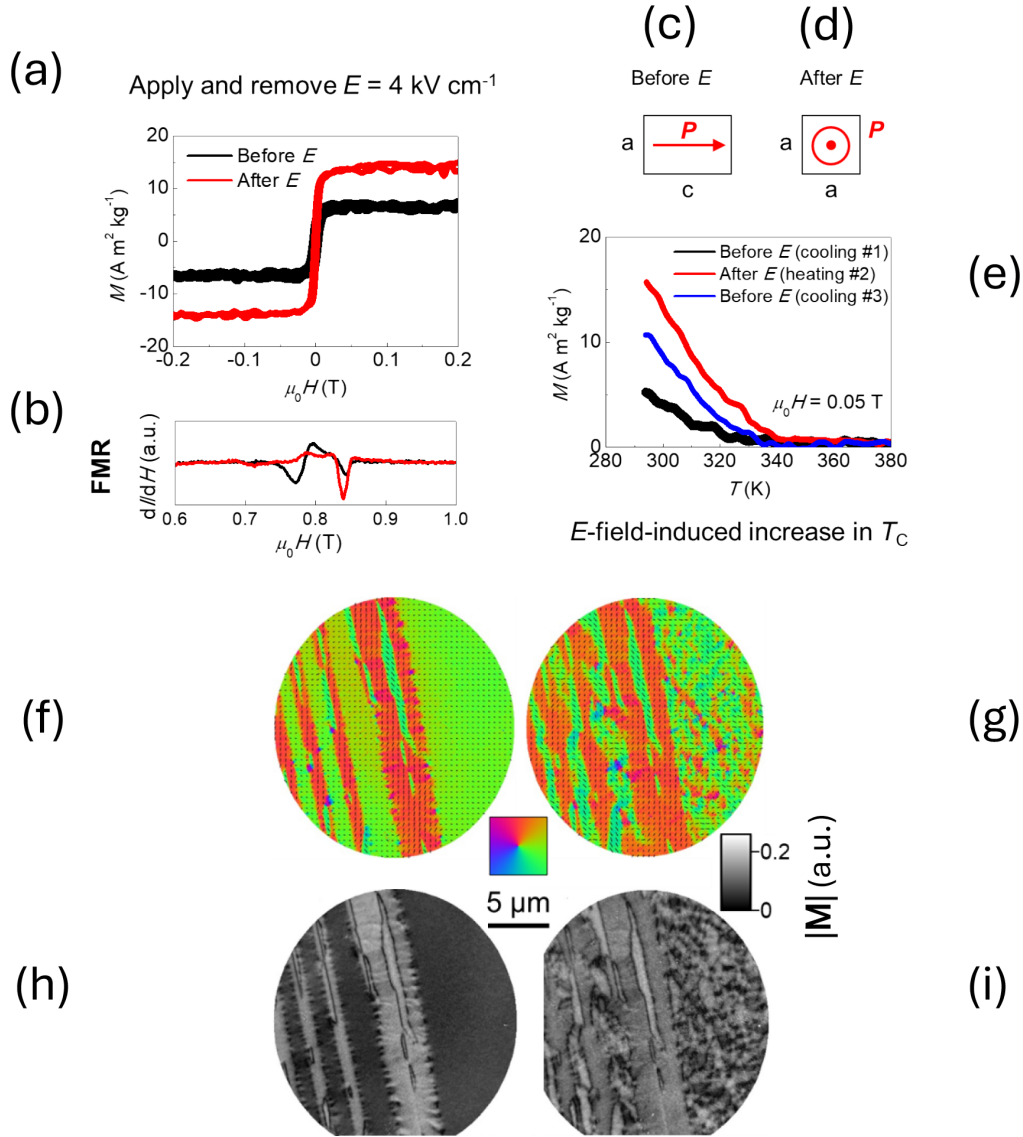


FIG. 5. Magnetoelectric effects in LSMO on T-phase BTO. (a) Macroscopic magnetization M versus applied field H along the in-plane easy axis before (black) and after (red) the application of 4 kV cm^{-1} across the BTO substrate at 294 K. (b) The corresponding FMR data. (c),(d) Schematics of the BTO surface presented to the film, (c) before and (d) after the electric field reorients polarization P from in-plane to out-of-plane. (e) Plots of $M(T)$ for different electrical and thermal histories. (f),(g) Vector maps of in-plane LSMO magnetization and (h),(i) the corresponding magnetization magnitude, (f),(h) before and (g),(i) after the application of 4 kV cm^{-1} at 294 K. The Fig. 3 caption gives more information about the vector maps and magnitude maps.

[Figs. 3(c), 3(f), and 3(i)] show that LSMO domain magnetizations lie primarily along orthogonal directions when the BTO is in the T phase [Figs. 3(a) and 3(c)], lie primarily along an intervening direction when BTO is in the O phase [Figs. 3(d) and 3(f)], and adopt a more complex pattern when BTO is in the R phase [Figs. 3(g) and 3(i)]. Magnetic domain walls are readily apparent in maps that represent the corresponding magnitude of the local magnetization [Figs. 3(b), 3(e), and 3(h)], where apparent domain wall width is enhanced because stray fields at domain walls affect secondary electrons and imaging. After averaging the magnitude of the magnetization over the field of view in each map, a single scaling factor permitted all three values [black points, Fig. 2(b)] to fall near the high field of the $M(T)$ plot [pink data, Fig. 2(b)], thus

demonstrating good equivalence between the macroscopic and microscopic data.

Using different samples, we now investigate magnetoelectric effects that arise due to the application and removal of 4 kV cm^{-1} across the BTO substrate in the O phase (Fig. 4) and the T phase (Fig. 5). For O-phase BTO at 217 K, XMCD-PEEM data reveal that exposure to the field tends to align magnetic domains, whose magnetizations then lie in a narrower range of angles [Figs. 4(a)–4(d)]. These magnetic changes arise because the polarization in the substrate is switched from in-plane to out-of-plane, such that the unit cell c - c template becomes an a - c template [21] [Figs. 4(e) and 4(f)]. The magnetic changes are also seen from in-plane magnetization loops along orthogonal directions, for which

the electric field produces opposite changes of squareness [Figs. 4(g) and 4(h)].

For T-phase BTO at 294 K, exposure to the 4 kV cm^{-1} increases the easy-axis saturation magnetization, as seen from magnetization loops [Fig. 5(a)]. The corresponding FMR data [Fig. 5(b)] show that the electric field converts two peaks (low-magnetization regions and high-magnetization regions) into one peak (high-magnetization regions only). These increases in magnetization arise due to an increase of Curie temperature in some regions that is driven by $\sim 1\%$ in-plane uniaxial contractions when the polarization in the substrate switches from in-plane to out-of-plane, such that the unit cell a - c template becomes an a - a template [Figs. 5(c) and 5(d)]. Measurements of magnetization versus temperature are concomitantly sensitive to the thermal and electrical history of the sample and the change of Curie temperature with electric field is apparent [Fig. 5(e)]. Microscopically, XMCD-PEEM data reveal that the effect of the field is to fragment monolithic ferromagnetic domains into regions whose magnetization varies in both direction [Figs. 5(f) and 5(g)] and magnitude [Figs. 5(h) and 5(i)], most notably in the right-hand third of the images. The low Curie temperature in the low-magnetization regions is enhanced by the field-induced strain, implying substantial caloric effects [33]. This field-induced variation in magnetization magnitude contrasts the field-induced homogenization of magnetization magnitude observed via FMR [Fig. 5(b)].

IV. DISCUSSION

LSMO/BTO represents a complex system in which the local LSMO magnetization is controlled by a variable epitaxial strain. This epitaxial strain is primarily governed by the aspect ratio of the template presented by the interfacial BTO domains to the LSMO film. Said aspect ratio is primarily determined by temperature-induced phase transitions and electric field-induced ferroelectric domain switching. However, the complexity we observe evidences subtle modifications due to long-range (normal and shear) strain between the BTO domains [34]. The interesting features to emerge are as follows: sample-to-sample variation is evident if we compare the XMCD-PEEM images in Figs. 3–5, the applied field can increase [Fig. 5(b)] or decrease magnetic homogeneity [Figs. 5(f) and 5(g)], and the magnitude of the observed magnetoelectric effects [Fig. 5(a)] is similar to the magnitude of magnetoelectric effects observed in similar LSMO/BTO samples [24] (Fig. 6). The enhancement of the magnetoelectric coupling at room temperature may be attributed to the use of a magnetic bias field that saturates the magnetization, whose magnitude is modified by strain. This contrasts traditional magnetoelectric effects (Fig. 4) that only arise in sufficiently low magnetic fields when electrically driven strain modifies the direction and not the magnitude of the local magnetization.

In the future, it would be interesting to perform a microscopic study of the interfacial magnetoelectric coupling that has been predicted for LSMO/BTO and other systems [35] if one can overcome the challenge of growing high-quality ultrathin epitaxial LSMO on BTO in order to probe both materials using PEEM (probe depth of $\sim 7 \text{ nm}$) with XMCD and XLD contrast, respectively. For example, it was recently shown that one can grow high-quality ultrathin epitaxial Fe

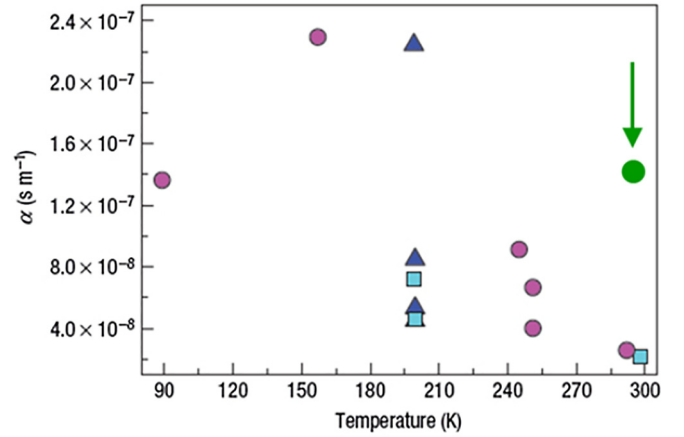


FIG. 6. Large magnetoelectric coefficient in LSMO/BTO at room temperature. The magnetoelectric coefficient $\alpha = \mu_0 dM/dE$ measured at 2 mT for different samples (color symbols) of LSMO/BTO at various temperatures is reproduced from Ref. [24], with the addition of high-field data from Fig. 5(a) (arrowed green data point, $\alpha = 1.47 \times 10^{-7} \text{ s m}^{-1}$).

on BTO and observe changes of magnetic order in the Fe film (imaged using XMCD-PEEM) due to changes of ferroelectric order in the BTO substrate (imaged using XLD-PEEM) [36]. It would also be interesting to perform similar studies on many LSMO/BTO samples to improve statistics and investigate the thermally and electrically driven changes in patterned elements of LSMO on BTO substrates such that magnetically independent LSMO islands can host single-domain states under thermal and electrical control.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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