

Pinning of antiferromagnetic domain walls and domain structure in ultrathin $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ filmsG. Panchal ¹, F. Stramaglia ¹, M. Krummenacher ¹, A. Kleibert ¹, C. W. Schneider ², D. Backes ³,
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We report the evolution of the antiferromagnetic domain structure of epitaxial $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ (LSMO) ultrathin films with thickness in the range from 5–50 unit cells (uc), using x-ray photoemission electron microscopy (XPEEM). While the 5 uc thick LSMO shows no magnetic contrast down to ~ 80 K, thicker films display a multidomain antiferromagnetic configuration with a nonmonotonic variation in the characteristic domain size from 0.3 μm at 10 uc, to 3–5 μm at 15–16 uc and 1 μm for 50 uc. Post-growth annealing is found not to impact strongly the magnetic domain state. In contrast, for a 15 uc film grown on a substrate characterized by large atomic steps (~ 1 μm), we observe much larger antiferromagnetic domains and the presence of net magnetic moment in the form of stripes with alternating contrast, assigned to the signal from the top uncompensated spins of the A-type antiferromagnetic state of LSMO. From the combined antiferromagnetic domain structure and the net magnetic moment contrast, we determine the exact orientation of the Néel vector, including at domain walls. We describe the antiferromagnetic domain size distribution in terms of interface and bulk contributions to the density of defects that pin the antiferromagnetic domain walls and which determine the equilibrium domain configuration. Our results demonstrate the impact of thickness and defects on the antiferromagnetic domain size and constitute a stepping stone in controlling the antiferromagnetic domain state required for oxide antiferromagnetic device applications.

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Antiferromagnetic (AFM) materials are drawing significant interest as building blocks for spintronics on account of their faster spin dynamics compared to conventional ferromagnets (in the THz instead of GHz range), the absence of stray fields and their immunity to external magnetic field perturbations [1]. Antiferromagnetic materials are more common than ferromagnets and antiferromagnetism can be found in combination with a wide range of other electronic states, including, semiconducting, insulating, and superconducting, highlighting their potential for multifunctional device applications. One challenge is that the use of AFMs in device applications demands effective writing and reading of the information in the Néel vector $\mathbf{L}$, which is not possible by conventional application of a magnetic field or sensing the local average magnetization, respectively. However, several indirect mechanisms have been discovered suitable for

probing and controlling the local antiferromagnetic state, including by electric and spin currents, optical excitations, thermal gradients and structural defects [1–4]. For example, manipulation of AFM domains and domain walls has been attained by tuning both the direction of the electric current and polarity flowing through tetragonal CuMnAs thin film devices [5,6]. Another challenge relates to controlling the equilibrium antiferromagnetic domain state. In contrast to magnetic domains in ferro- and ferrimagnets, which have been extensively studied and are fairly well understood [7], the domain formation and the mechanism for domain wall motion in antiferromagnets remain largely unexplored, in part because of the significant hurdle of probing antiferromagnetism. However, control of the equilibrium antiferromagnetic domain state down to the nanometer or atomic scale is critical for spintronics applications and a better understanding of domain formation is of high interest.

In fact, one major challenge in antiferromagnetic spintronics is the complex antiferromagnetic multidomain structure observed in most systems, even in relatively simple collinear antiferromagnetic materials, such as LaFeO_3 [8–10], $\text{La}_{0.7}\text{Sr}_{0.3}\text{FeO}_3$ [11], NiO [12–15], CoO [16], $\alpha\text{-Fe}_2\text{O}_3$ [17,18], Cr_2O_3 [19,20], $\text{Ni}_2\text{MnTeO}_6$ [21], CuMnAs [6,22], and PtMn [23], where an intricate coexistence of micrometer and sub-micrometer scale domains are found. Such complex domain configurations are surprising in antiferromagnetic

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systems, since they cost exchange, magnetocrystalline, and magnetoelastic energy at no magnetostatic energy gain, which is the main driving force for multidomain states in ferromagnets [7,24,25]; they likely arise as a consequence of the freezing of fluctuating magnetic domains as the sample is cooled from above the Néel temperature, where local variations in the magnetic anisotropy, due to strain or defects, pin the antiferromagnetic domain walls and prevent the formation of large uniform domains [4,26–29].

One antiferromagnetic system that has been less investigated but which presents a number of unique properties is the mixed valence manganites, where several antiferromagnetic states are stabilized as a function of hole doping and strain [30–33]. For example, the antiferromagnetic equilibrium configuration of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ changes from A-type to C-type to G-type as the Sr doping increases from about 0.5 to 0.6 and higher, respectively [33]. In ultrathin films, epitaxial strain can be used as a further knob to control the equilibrium spin configuration in this system, where compressive strain favors hole hopping along the out of plane direction and C-type antiferromagnetism, while tensile strain favors in-plane e_g $x^2 - y^2$ orbitals and A-type antiferromagnetism [34,35]. Electrostatic doping has been shown as well to be highly effective in controlling the interfacial spin configuration, including between ferromagnetic and antiferromagnetic [36,37]. Recently, we reported the observation of relatively large antiferromagnetic domains and a strong x-ray magnetic linear dichroism (XMLD) contrast in a 15 unit cells (uc) thick $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ film employed to control the magnetic ground state of ultrathin $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ films between antiferromagnetic and ferromagnetic through the chemical potential mismatch between the two layers [38]. The large domains and the strong magneto-optic contrast makes the detection and measurement of the antiferromagnetic state of the doped manganites more straightforward. Those characteristics have motivated us to carry out a detailed study of the evolution of the antiferromagnetic state of $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ with film thickness, including the local spin structure and the impact of post-growth annealing using x-ray photoemission electron microscopy (XPEEM). While we find no magnetic contrast for the thinnest film studied, 5 uc, we observe the presence of AFM domains for 10 uc and above, with a characteristic lateral size that increases from 10 uc to 15–16 uc and then decreases again at 50 uc. We find that post-growth annealing does not affect markedly the equilibrium magnetic state. We ascribe the thickness evolution of the antiferromagnetic domain size to competing contributions from interface and bulk point defects that act to pin the antiferromagnetic domain walls.

II. SAMPLE GROWTH AND EXPERIMENTAL TECHNIQUES

The samples in this study consist of $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ films 5, 10, (three) 15, 16, and (two) 50 unit cells (uc) thick grown on TiO_2 -terminated $\text{SrTiO}_3(001)$ substrates. The samples were grown in an oxide molecular beam epitaxy (MBE) system with a base pressure of 2×10^{-10} mbar, equipped with five effusion cells, a reflection high-energy electron diffraction (RHEED) system and a load-lock chamber for sample transfer

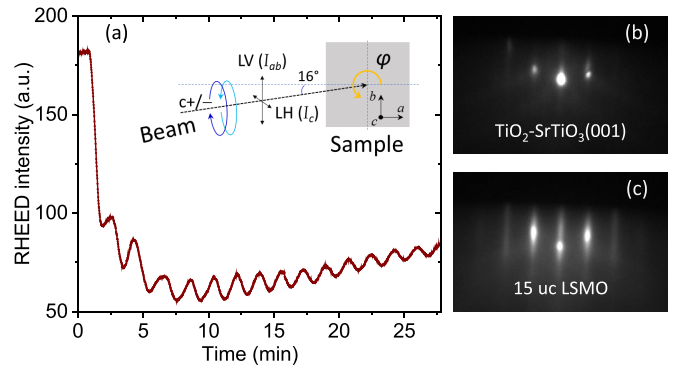


FIG. 1. (a) RHEED intensity oscillations as a function of time for one 15 uc $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ film. Inset shows the XPEEM measurement geometry. RHEED patterns along the $\langle 10 \rangle$ azimuth for (b) the TiO_2 -terminated $\text{SrTiO}_3(001)$ substrate before film growth and (c) $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ film immediately after growth. The energy of the incident electron beam was set to 15 keV.

[39]. $\text{SrTiO}_3(001)$ substrates with a miscut below $\pm 0.1^\circ$ (SurfaceNet GmbH) were chemically treated with a $\text{HCl}:\text{HNO}_3$ solution and annealed to 1000°C in air to provide single TiO_2 -terminated surfaces [40,41]. For the $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ deposition, we set the molecular oxygen partial pressure to $(5 \pm 1) \times 10^{-7}$ mbar, the substrate temperature to 720°C , and the $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ evaporation rate to about 0.5 uc/min. The thickness was monitored in real time by following the RHEED intensity oscillations observed during film growth, as illustrated in Fig. 1(a) for a 15 uc $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ sample, confirming layer-by-layer growth mode and the presence of atomically flat surfaces. The RHEED patterns and diffraction spot positions remained the same for the $\text{SrTiO}_3(001)$ substrate and the $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ films, confirming the expected cube-on-cube epitaxial relationship. The films were cooled down to room temperature under the same oxygen partial pressure used for the film growth. To study the impact of annealing on the antiferromagnetic domain structure we subsequently annealed the 10, one 15, 16, and one 50 uc films *ex situ* after growth under flowing O_2 at 600°C for 6 hours.

X-ray photoemission electron microscopy measurements were carried out at the SIM beamline of the Swiss Light Source, Paul Scherrer Institut (Switzerland), at the UE49PGM beamline at BESSY II (Germany), and at the I06 beamline of the Diamond Light Source (U.K.). Except where indicated otherwise, the measurements were carried out at the nominal base temperatures of 80 K at SIM, 50 K at UE49PGM, and 100 K at I06. In XPEEM, the sample is illuminated uniformly with a monochromatic x-ray beam and the local intensity of the photoemitted secondary electrons is recorded to obtain local maps of the sample x-ray absorption with ~ 50 nm lateral resolution [42–44]; the very short escape depth of a few nm of the detection of secondary photoemitted electrons makes XPEEM particularly surface sensitive [45]. To determine the full magnetic state, we use both the magnetic circular dichroic (XMCD) and the XMLD effect to probe the presence of a net magnetic moment or antiferromagnetic order, respectively [45,46]. Magnetization contrast images are obtained by pixel-wise division of drift-corrected x-ray absorption images taken with right (c+) and left (c−) circularly polarized light, giving

contrast maps of the magnetization via the XMCD effect; a similar procedure is carried out for images taken with linear horizontal (LH) and linear vertical (LV) polarized light, giving maps of the antiferromagnetic domains via the XMLD effect. In both instances, the energy of the x-rays is tuned to a value that maximizes the respective effect at the Mn $L_{2,3}$ absorption edges; in practice, this is determined by first measuring the x-ray absorption spectrum (XAS) to identify the peak positions and the expected XMLD maxima based on reference XMLD spectra (this procedure is required since the photon energy scale can differ between different beamlines). All measurements reported here are carried out at zero applied magnetic field. The magnetic contrast in XMCD is proportional to the projection of the magnetization on the x-ray propagation direction, $\mathbf{k} \cdot \mathbf{M}$ [45,46]; in our case, the x-rays impinge the sample surface at a grazing angle of 16° . The XMLD effect, in contrast, is proportional to quadratic terms of the magnetization or Néel vector projected along the electric field direction [47,48]. Typically, we collect 20–32 images per polarization, with each image corresponding to the average of 5 images, for a total integration time of about 10 min. Each image sequence was visually inspected and we found the presence of magnetic fluctuations in a few instances attributed to thermal excitations; however, the changes in the magnetic state during the course of the measurement was found not to be significant. In most cases, we acquired pairs of images at energies where the magnetic contrast is inverted and took the division of such images in order to further improve the signal to noise ratio and to remove illumination inhomogeneities (in such case, the resulting magnetic contrast corresponds to the difference of the contrasts at the two energies). The experimental sample geometry is such that the direction of the electric field for linear vertical light (LV) is along the film plane, while for linear horizontal (LH) the electric field is predominantly out of plane (Fig. 1, inset). X-ray absorption spectra were collected by taking consecutive images as a function of the photon energy for a given light polarization. We define here the XMLD contrast as $(I_{ab} - I_c)/(I_{ab} + I_c)$, where I_{ab} and I_c are the measured XPEEM intensities with electric field in-plane (LV) and out of plane (LH), respectively. The XMCD contrast is defined as $(I^- - I^+)/(I^- + I^+)$, where I^- and I^+ are the measured XPEEM intensities with circularly left and right polarized light, respectively (Fig. 1, inset).

Electrical resistivity measurements as a function of temperature were carried out in a home-built system based on a cold finger cryostat and gold-coated spring probe contacts. For the samples studied here, circular Au pads, 10 nm thick and 500 μm wide, were evaporated using a shadow mask to provide good electrical contact to the sample. Current versus voltage measurements were performed continuously as the temperature was ramped from 20 K to 350 K and the sample resistance was determined from a linear fit to the current versus voltage curves, with a maximum error in temperature of ± 0.3 K. The resistance values were multiplied by a factor of $9.06t$ to yield the resistivity, where t is the nominal film thickness and the numerical factor corrects for the measurement geometry [49].

X-ray diffraction measurements were carried out in a Seifert four circle diffractometer equipped with a Cu anode x-ray tube and a linear array detector. The measurements

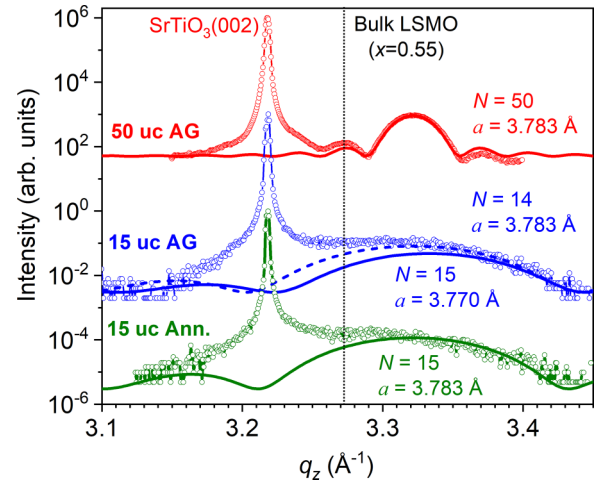


FIG. 2. X-ray diffraction patterns of as-grown and annealed 15 uc films and as-grown 50 uc $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ films near the (002) plane of SrTiO_3 (AG: as-grown; Ann: annealed). Solid and dashed lines are visual fits to the data using the expression for the diffracted signal from N atomic layers with spacing a . The spectra are normalized to the SrTiO_3 (002) peak amplitude and are shifted vertically successively by a factor of 10^3 for ease of display.

consisted of $\theta - 2\theta$ scans along the (002) diffraction peak to determine the out of plane lattice parameter. The atomic force microscopy measurements were performed in the tapping mode using a Bruker dimension icon 3100 instrument [50].

III. RESULTS AND DISCUSSION

We start by discussing the film misfit strain; in Fig. 2, we present the x-ray diffraction spectra of as-grown and annealed 15 uc films, and one as-grown 50 uc film, near the (002) peak of the SrTiO_3 substrate. For the 50 uc film, a clear peak to the right of the SrTiO_3 (002) peak is observed, which is readily identified as the pseudo-cubic (002) peak of $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$. In the figure we present, as full line, a visual fit to an expression of the form $A + B \sin^2(Nq_z a/2)/\sin^2(q_z a/2)$ expected for N atomic layers spaced by a , where q_z is the out of plane photon momentum transfer; A is the signal background and B an amplitude factor; in this expression, the out of plane lattice parameter and number of scattering planes determines the peak position and width, respectively. We find an excellent agreement for $N = 50$ and $a = 3.783$ Å. The pseudo-cubic lattice parameter of bulk $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ is 3.84 Å [30,33,52], indicating that the films are tensile strained, as expected from the lattice parameter of SrTiO_3 , 3.905 Å [51]. For the thinner films, there is a large intrinsic broadening in the signal due to the reduced number of scattering planes. We confirmed that the broad signal to the right of the SrTiO_3 (002) peak is from $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ from the measurement of a 15 uc film, where we observe a similar broad peak, and after removal of the film by polishing the sample, where the peak is absent. Further, visual fits to the expression for the diffracted signal, shown as full lines in Fig. 2, give good agreement to the data by using the nominal film thickness, corroborating that

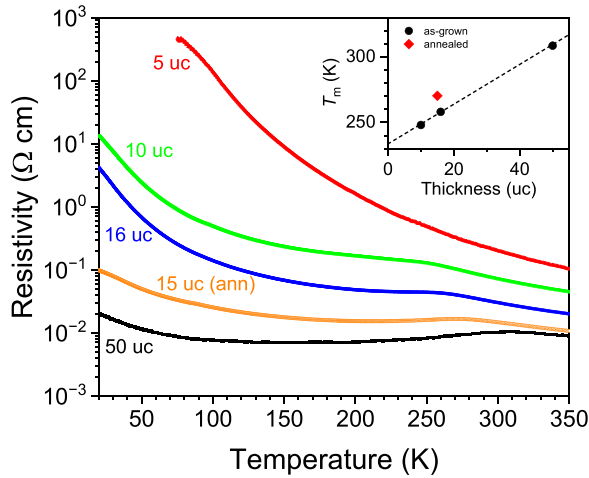


FIG. 3. Resistivity variation as a function of temperature for different film thicknesses. Inset shows the variation of the resistivity peak as a function of film thickness (dashed line is a guide to the eye).

the broadening is intrinsic to the scattering geometry. For the thinner films, the intrinsic broadening has the consequence that the diffraction peak starts to overlap with the $\text{SrTiO}_3(002)$ line, but the expression works remarkably well. For the as-grown $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ film, a better description of the data is obtained for $N = 14$ and $a = 3.783 \text{ \AA}$, shown as a dashed line. We conclude that, within the limit of accuracy of our measurements, all films are similarly strained.

Transport measurements were carried out to determine the magnetic critical temperature (T_N) by taking advantage of the characteristic peak in the resistivity (T_m) observed in mixed valence manganites as the system crosses the paramagnetic to magnetic phase due to the “colossal” magnetoresistance effect, including the antiferromagnetic transition near half doping [30,32,39,52]. In Fig. 3, we present the temperature variation of the resistivity for the various film thicknesses. We find that the 5 uc film exhibits an insulating behavior over the accessible temperature range (the sample is too insulating below 75 K for the sensitivity of our set-up). For the 10 uc, one observes a general increase in resistivity with decreasing temperature, however, with a peak at 248 K, which we identify with the critical temperature; the resistivity peak becomes more pronounced in the thicker films, with the 50 uc showing predominant metallic behavior with only a slight increase in resistivity at the lowest temperatures. Such increase in resistivity with decreasing temperature is often observed in thin films of the mixed valence CMR manganites and attributed to the effect of electron localization induced by A-site disorder [53]. We find as well that the resistivity increases with decreasing film thickness, suggesting a strong influence of the substrate interface to charge carrier scattering, including the presence of an insulating electric layer (estimated as ~ 3 uc at 300 K by extrapolating the conductivity to zero with film thickness) [54,55]. Finally, the variation of the resistivity peak with film thickness shows a linear increase from 10 to 50 uc, reaching $T_m = 308.5 \text{ K}$ for the 50 uc film (Fig. 3, inset). The magnetic behavior of the tensile-strained films is markedly different from the bulk, where a sequence of paramagnetic-

ferromagnetic-antiferromagnetic transitions (respectively, at 333 K and 230 K) with decreasing temperature is observed [32,33]. Effectively, in the thin LSMO films, we find that the ferromagnetic phase is suppressed and the onset of the antiferromagnetic phase is enhanced in temperature.

In order to probe the local antiferromagnetic state, we turn to the XPEEM results. According to the resistivity results, we expect that the $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ films are magnetic at the base XPEEM measurement temperature, with the possible exception of the 5 uc film. In Fig. 4, we present the evolution of the antiferromagnetic state as a function of $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ film thickness for both as-grown and annealed films. For these images, the in-plane electric field of the x-ray light is approximately parallel to the in-plane $\langle 110 \rangle$, where the XMLD signal is strongest, in contrast to the case when the light polarization is along the main crystal axis, where the contrast is very weak. The contrast scale is the same for all images and we attribute the strong variations in absolute contrast between the images to extrinsic factors associated with differences in the measurement conditions (carried out at different instruments). For the 5 uc, we find that the contrast in the image varies randomly from pixel to pixel, possibly due to limited signal statistics; it suggests either a magnetic state with domains with lateral size below about 50 nm, or a nonmagnetic state (consistent with the transport results; however, we also cannot exclude a C-type AFM state with Néel vector pointing along the out of plane direction, a configuration to which our measurement geometry is not sensitive, or that the critical temperature is below 80 K). For the 10 uc LSMO film, we observe an XMLD contrast characteristic of a multidomain antiferromagnetic state, with equally distributed small domains having a lateral size in the sub- μm range. At 15 and 16 uc, the LSMO films present a multidomain structure with much larger domains extending up to 5–10 μm , while at 50 uc the size of the antiferromagnetic domains again reduces to 2–5 μm . XMCD-PEEM images showed no discernible magnetic contrast for the as-grown LSMO films. We studied the impact of post-growth annealing on the magnetic domain structure to evaluate the role of oxygen stoichiometry in pinning the antiferromagnetic domain walls. Oxygen vacancies lower the free energy of the system by increasing the entropy, and are always present at finite temperature, with a density that depends on cation stoichiometry, substitutional impurities, temperature, and oxygen partial pressure [56]. In the manganites, they have activation energies on the order of a few eV [57], and their density can be controlled by adjusting the oxygen partial pressure during growth or by annealing [58]. Since the films are grown and cooled under an oxygen partial pressure of 5×10^{-7} mbar, one may expect a finite density of oxygen vacancies in the films, which should decrease after annealing at atmospheric pressure of oxygen. The impact of annealing is found to be weak, as shown in Figs. 4(g)–4(j), with the 10, 16, and 50 uc films showing similar domain morphology and size before and after annealing; the annealed 15 uc film (different sample from that of the as-grown 15 uc) shows a significantly different domain pattern, with much larger and regular domains, which suggests that a different factor either than thickness or post-growth annealing is playing a stronger role in determining the equilibrium magnetic state. In fact, a major difference to the other samples is the higher quality of the SrTiO_3 substrate for

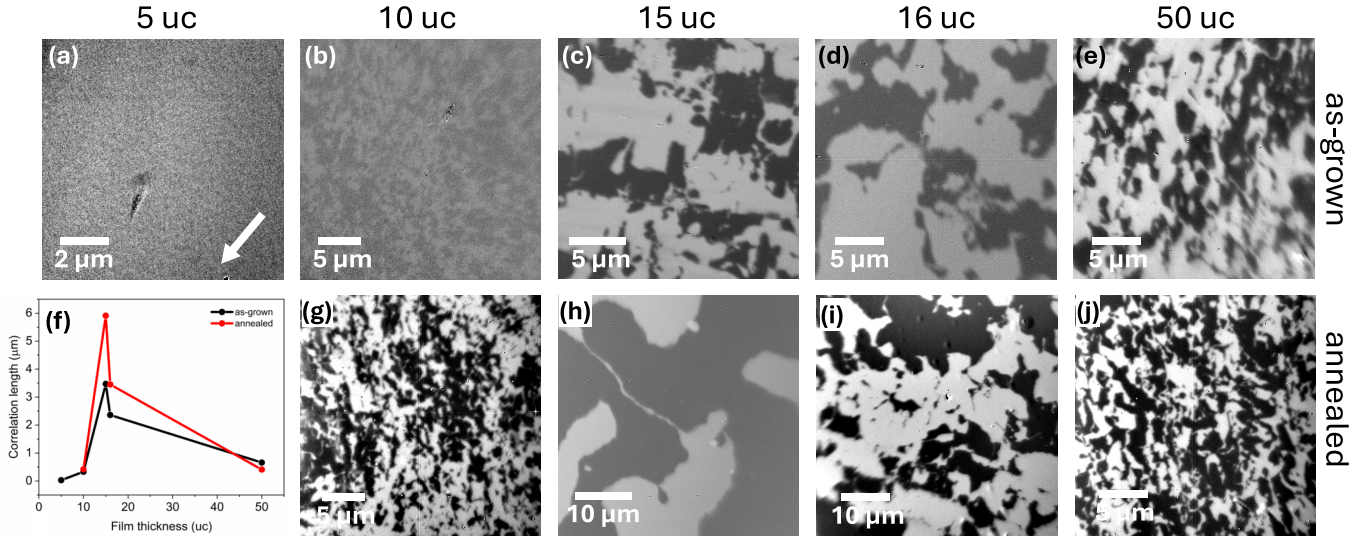


FIG. 4. XMLD contrast images of $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ as a function of thickness for (a)–(e) as-grown and (g)–(j) annealed samples, as labeled. The 10 and 16 uc films correspond to the same samples before and after annealing. The nominal measurement temperatures are 80 K for as-grown 5 and 60 K for as-grown 50 uc; 80 K for as-grown 15, 16 uc, and 100 K for as-grown 10 uc. White arrow indicates the direction of the incoming x-rays (common to all images). The intensity scale is set to ± 0.4 for all images. (f) Variation of the domain correlation length with film thickness determined from the XMLD images.

this sample, with an exceptionally small surface miscut, as determined from atomic force microscopy. The weak impact of post-growth annealing on the magnetic state of the LSMO films indicate that either the films are fully oxygenated after growth or that oxygen vacancies do not act as strong pinning sites for the domain walls.

To determine the orientation of the Néel vector, we consider the expression for the x-ray absorption for linearly polarized light in tetragonal symmetry (D_{4h}) [47,48]:

$$\begin{aligned}
 I_{D_{4h}} = & F_{a_{1g}^B}^{(0)}(\epsilon_x^2 + \epsilon_y^2) + F_{a_{1g}^A}^{(0)}\epsilon_z^2 + 2F_{e_g}^{(2)}(\epsilon_x\hat{m}_x + \epsilon_y\hat{m}_y)\epsilon_z\hat{m}_z \\
 & + 2F_{b_{2g}}^{(2)}\epsilon_x\hat{m}_x\epsilon_y\hat{m}_y + \frac{1}{2}F_{b_{1g}}^{(2)}(\hat{m}_x^2 - \hat{m}_y^2)(\epsilon_x^2 - \epsilon_y^2) \\
 & + F_{a_{1g}^B}^{(2)}(\hat{m}_z^2 - \frac{1}{3})\epsilon_z^2 + F_{a_{1g}^A}^{(2)}(\hat{m}_z^2 - \frac{1}{3})(\epsilon_x^2 + \epsilon_y^2),
 \end{aligned} \quad (1)$$

where the F coefficients represent independent XAS, and m_i , and ϵ_i are the cartesian components of the unit vectors of the magnetization and light polarization, respectively. The $F^{(0)}$ terms give the natural linear dichroic response, while the $F^{(2)}$ give rise to x-ray magnetic linear dichroic spectra. For the particular cases of $\epsilon_z = 0$ (LV) and $\epsilon_x = \epsilon_y = 0$ (LH):

$$\begin{aligned}
 I_{D_{4h}}^{\parallel} = & F_{a_{1g}^B}^{(0)} + 2F_{b_{2g}}^{(2)}\epsilon_x\epsilon_y\hat{m}_x\hat{m}_y + \frac{1}{2}F_{b_{1g}}^{(2)}(\hat{m}_x^2 - \hat{m}_y^2)(\epsilon_x^2 - \epsilon_y^2) \\
 & + F_{a_{1g}^B}^{(2)}(\hat{m}_z^2 - \frac{1}{3}),
 \end{aligned} \quad (2)$$

$$I_{D_{4h}}^{\perp} = F_{a_{1g}^B}^{(0)} + F_{a_{1g}^A}^{(2)}(\hat{m}_z^2 - \frac{1}{3}). \quad (3)$$

The XMLD images shown in Fig. 4 are the difference between images of the x-ray absorption taken with the electric field in-plane and out of plane. In the x-ray absorption images, we observe a strong intensity variation from the AFM domains in the former case, but almost no contrast when the electric field is out of plane, consistent with a uniform m_z^2 , i.e., either zero or with a constant projection along the anisotropy axis:

while both previous expressions have a term in m_z^2 , a uniform m_z^2 gives no contrast in the images. It follows also from (2) that the observation of strong contrast for $\epsilon_x = \epsilon_y$ (light polarization along [110]) and weak contrast for $\epsilon_x = 0$ (light polarization along [100]), implies that the in-plane component of the Néel vector is oriented along $\langle 110 \rangle$.

To obtain a fuller picture of the spectral evolution with energy, we consider in more detail the XAS response for the two linear light polarizations, vertical (electric field along in-plane $\langle 110 \rangle$) and horizontal (electric field predominantly along [001]), extracted from uniform magnetic regions of the sample, as shown in Fig. 5 for the 50 uc film. When scanning through the images as a function of energy, one observes strong variations in contrast across the image as the energy crosses the $L_{2,3}$ edges for vertical polarized light, as illustrated in the inset to Fig. 5(a) for an image taken at 640.7 eV where the contrast is strongest. The intensity contrast arises from the XMLD effect, more specifically, from different light absorption for antiferromagnetic domains with Néel vectors having different projections along the electric field direction [second term of (2)]. By plotting the XAS intensity as a function of energy for the regions with different contrasts highlighted in the inset to Fig. 5(a), one obtains the XAS and XMLD spectra shown in the main plot (effectively, $F_{b_{2g}}^{(2)}$). The two domains correspond to orthogonal in-plane orientations of the Néel vector and from the XMLD spectrum we determine the energy values of maximum contrast and where it is inverted. For linear horizontal light, both highlighted regions show a near identical XAS with only a nearly imperceptible variation in contrast [inset to Fig. 5(b)]; the linear dichroism between the two domains is a factor of about 10 smaller than that for the vertical light polarization and inverted in sign; it can be explained from the small in-plane electric field component present for this measurement geometry, with an XMLD signal smaller by factor of $-\sin^2 16^\circ \cong -0.076$, where the minus sign reflects the different projections of the Néel vector

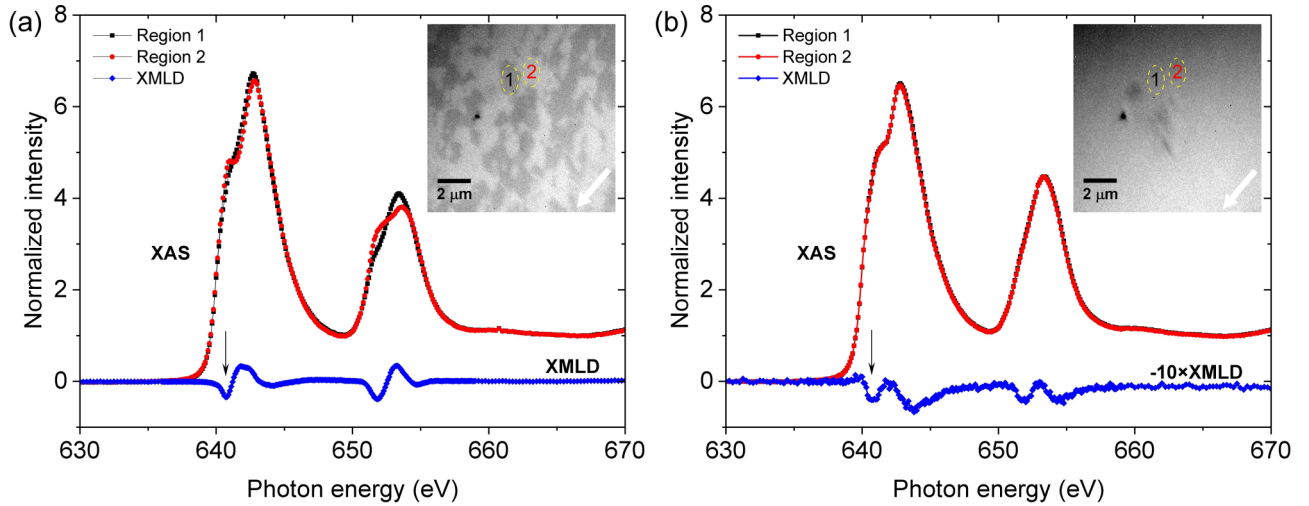


FIG. 5. Local XAS and XMLD spectra for (a) vertical and (b) horizontal light polarization extracted from the two regions marked in dashed ellipses in the figure insets, showing the respective original XPEEM images taken at 640.7 eV. The XAS have been normalised by the incoming light intensity and the XMLD spectrum in (b) has been multiplied by a factor of -10 for a better comparison with the spectrum shown in (a). The XMLD is calculated here as $I_1 - I_2$, where $I_{1(2)}$ is the integrated intensity over region 1(2) normalized to the post-edge. The arrows in the images indicate the direction of light propagation.

compared to the case with vertical light polarization. We note that the natural linear dichroism, including from the break in symmetry of the electron orbitals introduced by the surface and by epitaxial strain, gives a uniform contribution to the XMLD and therefore no contrast in the images. Hence, we conclude that the $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ films present either in-plane quadratic magnetic anisotropy with easy axes along in-plane $\langle 110 \rangle$ or near cubic magnetic anisotropy with easy axis along $\langle 111 \rangle$.

XPEEM results for both the XMLD and XMCD contrast maps for the annealed 15 uc film along different directions of the sample azimuth are shown in Fig. 6. Figure 6(a) shows the antiferromagnetic domain structure along the nominal $\langle 110 \rangle$ direction ($\phi = 0$). In comparison with the other films in this thickness range, one finds much wider uniform antiferromagnetic domains, extending over 10s of μm ; the domain walls are also more regular. With increasing azimuthal angle, one finds a reduced magnetic contrast at $\phi = 19^\circ$ and almost no contrast at 38° ; at this angle, the Néel vector has near identical projections along the in-plane electric field direction and only the antiferromagnetic domain walls become visible (indicating that the sample was misaligned by $\sim 7^\circ$ after mounting). The domain boundaries seen in Fig. 6(c) display either a bright or dark contrast, consistent with domain walls where the Néel vector rotates within the plane of the film.

In Figs. 6(d)–6(f), we show the corresponding XMCD contrast images, where clear stripe patterns are observed. At 0° , they are almost absent in the black antiferromagnetic domain regions and become more pronounced at 38° , where they appear over the whole image. The contrast amplitude is much weaker than that typical for ferromagnetic $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$; similar magnetic contrast images were reported for antiferromagnetic 3 uc $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3/\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ bilayers [38], where the presence of XMCD contrast was explained as due to the termination of alternating spin planes of the A-type antiferromagnetic $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ due to the substrate miscut, which exposes atomic terraces one unit cell in

height for TiO_2 -terminated $\text{SrTiO}_3(001)$ substrates, as illustrated in Fig. 7(a). Atomic force microscopy measurements carried out for this sample confirms the presence of wide atomic terraces, $\sim 1 \mu\text{m}$ wide on this region of the sample, Fig. 7(b), which agree with the periodicity of the magnetic stripe pattern, 54 half-periods over $50 \mu\text{m}$, as seen in Fig. 6(f). We attribute the observation of a magnetic contrast in XMCD to the surface sensitivity of XPEEM, where the top spin layer gives a stronger contribution to the XAS signal; this result also suggests that the $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ spin surfaces are fully uncompensated, i.e., with single spin termination, as expected for A-type antiferromagnetic order of tetragonal distorted $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ [34,35].

In fact, the magnetic imprint seen in the XMCD images can be used as a proxy for the orientation of the Néel vector. For example, the observation of XMCD stripes in the white domain at 0° , Figs. 6(a) and 6(d), shows that the Néel vector in these domains is oriented along the direction of the light propagation, while the black domains, where no XMCD stripes are observed, have an orthogonal Néel vector orientation. At 38° , Figs. 6(c) and 6(f), the XMCD contrast along the stripes does not change across the AFM domain walls with white contrast, while they flip contrast across the AFM domain walls with black contrast. A color-coded zoomed-in view of the region where the white and black antiferromagnetic domain walls meet is given in Fig. 7(c), where these changes in magnetic contrast can be better visualized. The spin configuration that is consistent with these data is schematically shown in Fig. 7(d). These results indicate that the spins rotate in the plane of the film across the 90° antiferromagnetic domain walls. This approach of combining both XMLD and XMCD contrasts provides a unique and powerful method to determine the exact Néel vector orientation, including across domain walls.

To better quantify the changes in the magnetic domain morphology, we estimate the characteristic domain size from the length decay of the autocorrelation function (correlation length) of the magnetic contrast, plotted in Fig. 4(f) for the

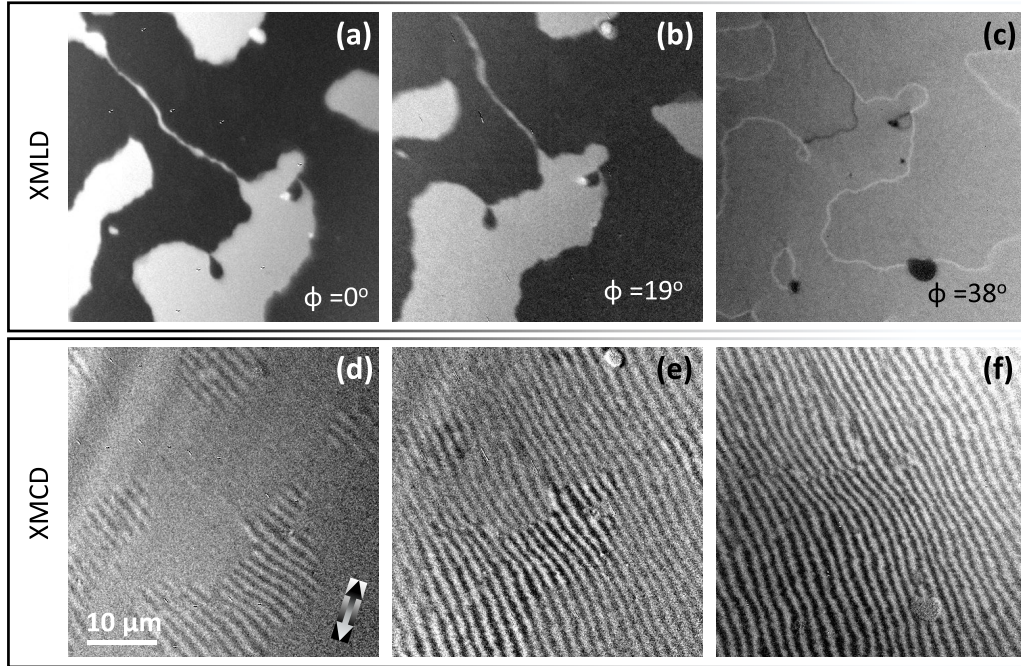


FIG. 6. (a)–(c) XMCD contrast of the annealed 15 uc $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ thin film as a function of the angle ϕ between the x-ray light direction and the nominal $\langle 110 \rangle$ sample direction (contrast range set to ± 0.015). (d)–(f) Respective XMCD contrast images (contrast range set to ± 0.003). Note the slight vertical shift in the image position for $\phi = 38^\circ$. The scale bar and the direction of the x-ray light indicated by the double arrow in (d) are the same for all images. The intensity gradient along the x-ray beam direction and the stripes seen in (d) are artifacts introduced by inhomogeneities in illumination and light interference with the exit slits, respectively.

images (b–e) and (g–j). For the as-grown films, the correlation length changes from $0.32 \mu\text{m}$ for 10 uc to $4.8 \mu\text{m}$ for 15 uc and $3.1 \mu\text{m}$ for 16 uc, and drops to $1.2 \mu\text{m}$ for 50 uc. (For the 5 uc, the correlation length is of the order of the pixel size and below the expected spatial resolution.) These values quantify the qualitative observation of an increase in the domain size from 10 to 15–16 uc, and a reduction at 50 uc. We attribute the large change in the domain size to the role of defects that locally modify the magnetic anisotropy and act to pin the antiferromagnetic domain walls. Possible defects include substitutional, antisite (cations), and vacancies (oxygen) point

defects in the bulk of the films and defects arising from the substrate interface, including epitaxial strain (including oxygen octahedra relaxation) [59–61], step-edges [62], interdiffusion [63], or possible charge-transfer effects [64–66], including between Mn and Ti cations [67,68]. Extended defects, such as grain boundaries [69], phase separation in ultrathin manganite films [55], or nonmagnetic inclusions [70] can also act as pinning centers for the domain walls. Direct access to the identity and number of point defects is in general difficult [71,72], however, we can estimate their density from the relative area size occupied by the domain walls. During

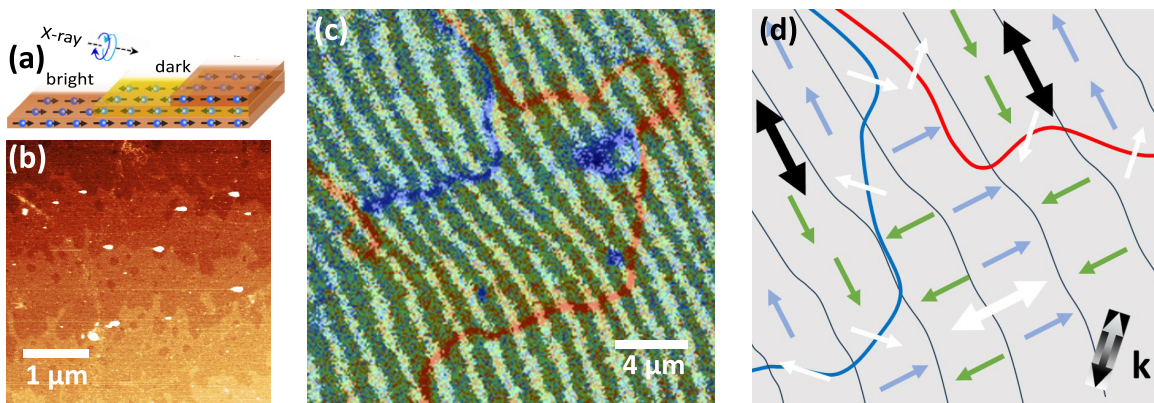


FIG. 7. (a) Schematic diagram of the antiferromagnetic spin structure across unit cell high-surface terraces. (b) Atomic force microscopy image of the annealed 15 uc $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ film. (c) Color-coded superposition of XMCD (blue and red) and XMCD (green and cyan) contrast images [detail of Fig. 6(c) and 6(f)]. (d) Schematic of the terminating spin configuration where domain walls of opposite contrast meet. White and black double arrows indicate the orientation of the Néel vector in the different domains, while simple arrows indicate the spin orientation of the terminating spin layer.

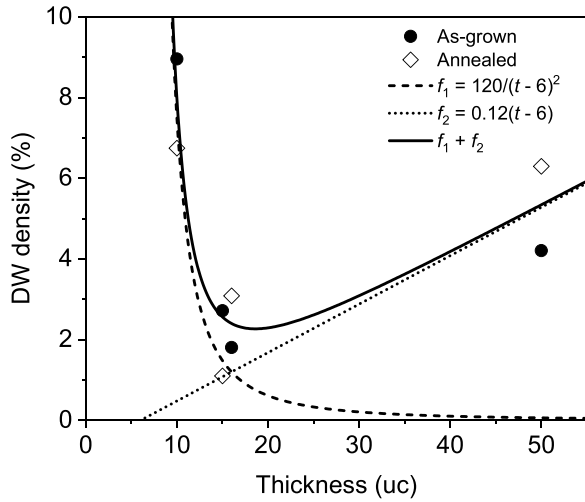


FIG. 8. Variation of the relative antiferromagnetic domain wall density with $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ film thickness (symbols). Full line is the sum of an interfacial term with an inverse quadratic variation (dashed line) and a bulk linear contribution (dotted line).

the cooling process, local domains are formed at nucleation sites and expand as the temperature is reduced; their final distribution is largely determined by the strength of pinning sites that arrest their expansion. For example, in the case of the 10 uc film, the domain pattern suggests a short propagation length for the domains after nucleation. In Fig. 8, we present the variation of the total antiferromagnetic domain wall length relative to the total area with film thickness. This information was extracted from the images by setting a threshold intensity of $\approx 50\%$ to identify the domains, finding the edges, and ‘skeletonizing’ the image (reducing the boundaries to a single pixel), and finally, determining the ratio of pixels occupied by the domain walls to the total area of the image. The plot aims at quantifying what is readily visible in the magnetic images and shows a large density of domain walls at 10 uc, a quick drop at 15, 16 uc and a slight increase at 50 uc. While there is some variation between as-grown and annealed samples, the overall trend with thickness is similar for both cases. The observed variation can be described as originating from two contributions, one interfacial, related to the onset of magnetism at $\approx 5\text{--}6$ uc, and a bulk contribution, varying linearly with thickness. The interfacial contribution can be described as rapid inverse quadratic variation associated to the strain field of point defects [73,74], while the linear term originates from a uniform density of defects in the bulk of the film. While tentative, the present picture may serve as starting point for future more detailed studies of the contribution of point defects into the pinning of antiferromagnetic domain walls in low-anisotropy antiferromagnets.

While we have no direct access to the local defects at the pinning sites, it is helpful to discuss their possible origin. A significant source of point defects in the oxide perovskites is oxygen vacancies [37,75,76], which tend to impact negatively the magnetic properties of the manganites [58,71]; for example, thin films grown under reducing conditions exhibit depressed saturation magnetization, magnetic critical temperature and metal to insulator transition [77–81], while direct

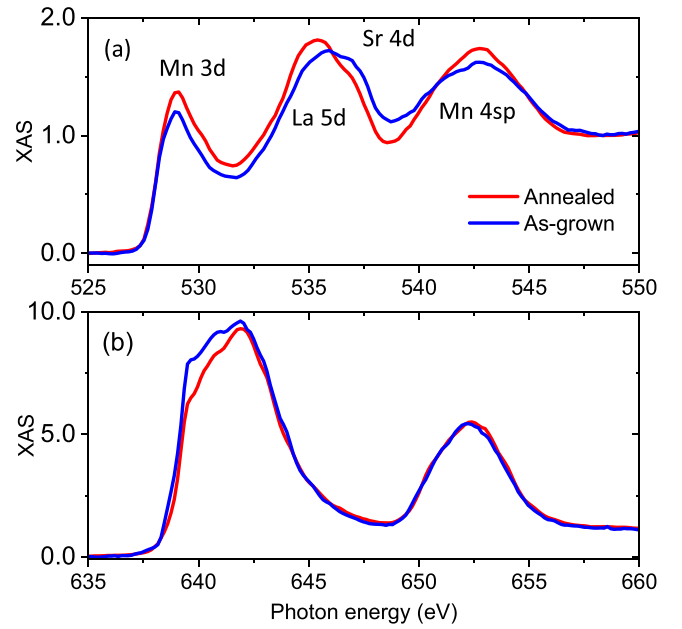


FIG. 9. Isotropic x-ray absorption spectra recorded at (a) O K edge and (b) Mn L edge at 250 K for as-grown and annealed 15 uc $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ films.

modulation of the density of oxygen vacancies in manganite layers induced by applied electric fields shows as well reduced magnetic characteristics with increased density of oxygen vacancies [82–86]. Furthermore, there is a close link between epitaxial strain in thin films and the density of oxygen vacancies, where tensile strain tends to favor the formation of oxygen vacancies in the MnO_2 planes [87–89], which may also act to relieve the in-plane stress of the film [90,91]. Contributing factors for the degraded magnetic properties include the reduction in atomic coordination, which affects the local atomic structure and the double exchange and superexchange interactions; modifications in the valence state of the cations, which affects not only the spin state, but can lead also to significant local lattice distortions due to differences in cationic size (chemical pressure) [92]; and changes in charge carrier density, since each oxygen vacancy donates two electrons to the conduction band (or traps two holes), which can lead to changes in the transport and magnetic properties when double exchange is active. Given the impact of oxygen vacancies on the local atomic and magnetic structure, it may be expected that they act to modify the local anisotropy landscape and pin magnetic domain walls. As discussed above, we find that post-growth annealing in oxygen does not significantly impact the magnetic properties of the $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ films; however, oxygen vacancies may still be present to compensate cation off-stoichiometry and low-valence impurities.

We checked the oxidation state of two, as-grown and annealed, 15 uc $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ films by measuring the XAS at the O K edge and Mn L edge, shown in Fig. 9; these measurements were carried out at 250 K, with circularly right polarized light, and give approximately the isotropic spectra for these samples. The signal was averaged over sample areas in the range from $10\text{--}30\text{ }\mu\text{m}^2$ and corrected for the energy dispersion plane in the case on the Mn signal (the x-ray optics

project the image of the exit slit on the XPEEM and, depending on the exit slit settings and XPEEM field of view, the energy dispersion of the x-ray monochromator can be present in the field of view; while the effect is small, it was corrected here, since the signal was taken from different positions in the image for the two samples) [93,94]. The samples were grown in separate runs and therefore some sample-related variations are expected; nevertheless, we find that the spectral differences are relatively small. At the O K edge, Fig. 9(a), the spectra differ mostly in a slightly higher intensity for the annealed sample at the spectral features associated with Mn hybridized states and in the inversion in intensity between La and Sr hybridized states [95], which may suggest a slightly different La:Sr content between the samples. The Mn spectra, shown in Fig. 9(b), differ in a slightly reduced intensity at the leading edge of the Mn L_3 peak for the annealed sample, consistent with a slightly higher oxidation state of Mn. A typical signature of oxygen vacancies in the manganites is the presence of a Mn^{2+} peak, which we do not identify in our samples, indicating that the samples are near fully oxygenated following film growth. Hence, we interpret the XAS to indicate a lower density of oxygen vacancies in the annealed film, which may however, originate from a better cation stoichiometry match compared to the as-grown film.

Concerning the contribution from the interface, we can rule out misfit dislocations, since all films are fully strained to the substrate; however, oxygen octahedra in LSMO/SrTiO₃(001) are known to relax to the bulk configuration within 2–10 uc [59,60,66]; local variations in the tilt angle could give rise to an interfacial-like source of pinning. Step edges arising from the surface miscut (0.1° corresponding to terraces ~200 nm in width) could also modify locally the magnetic anisotropy [96–98], however, the lack of directionality in the domain wall pinning and the results for the annealed 15 uc film showing that the domain walls do not follow the contours of the step edges (cf. Fig. 6), indicate that the impact of step-edge anisotropy may not be significant. Diffusion of Sr or impurities such as Ca and Si [99] from the substrate during film growth could locally modify the interface magnetic properties and, more generally, a less atomically smooth starting template for the film growth from the SrTiO₃ substrate could give rise to local variations in the film profile that locally results in local nonuniformities that pin the antiferromagnetic domain walls. Given the low impact of sample annealing and the significant difference in surface morphology found in the 15 uc film, this last aspect may be the main source of pinning from the interface. Our results suggest, therefore, that the main source of pinning in the LSMO films arises from the substrate interface, possibly due to a more disordered surface

or stronger diffusion of Sr or other impurities in a different substrate batch.

IV. CONCLUSIONS

In summary, the thickness evolution of the antiferromagnetic domain state in epitaxial ultrathin La_{0.45}Sr_{0.55}MnO₃ films studied by direct x-ray magnetic microscopy shows a complex, nonmonotonic behavior. We find that at 5 uc the La_{0.45}Sr_{0.55}MnO₃ film is insulating and exhibit no magnetic contrast down to the lowest measurement temperature (80 K), while films 10 uc and thicker exhibit a strong XMLD contrast. The domain size first increasing from 10 uc to 15–16 uc and decreases at 50 uc. We find that post-growth annealing has a minimal impact on the magnetic state, indicating that the films are fully oxygenated after growth. In one sample, characterized by a small substrate miscut, the antiferromagnetic domains are much larger and more regular compared to the other samples. For this sample, we observe as well the presence of XMCD contrast, originating from the single termination of the A-type antiferromagnetic configuration, an effect that is exploited to determine the exact Néel vector configuration of the film. The changes in the domain structure are attributed to the role of defects in pinning the domain walls, highlighting their impact of the equilibrium domain state of La_{0.45}Sr_{0.55}MnO₃. Our results demonstrate that control of the domain size can be achieved by tuning the film thickness and by controlling the density of defects and is expected to guide future work in controlling the antiferromagnetic domain state of oxide antiferromagnetic systems more generally.

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DATA AVAILABILITY

The data that support the findings of this study are openly available [50].

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