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Spintronics exploits the magnetoresistance effects to store or sense the magnetic information. Since the magnetoresistance strictly depends on the magnetic anisotropy of a system, it is fundamental to set a defined anisotropy to the system. Here, we investigate half-metallic La$_{0.67}$Sr$_{0.33}$MnO$_3$ thin films by means of vectorial Magneto-Optical Kerr Magnetometry and found that they exhibit pure biaxial magnetic anisotropy at room temperature if grown onto a MgO (001) substrate with a thin SrTiO$_3$ buffer. In this way, we can avoid unwanted uniaxial magnetic anisotropy contributions that may be detrimental for specific applications. The detailed study of the angular evolution of the magnetization reversal pathways and critical fields (coercivity and switching) discloses the origin of the magnetic anisotropy, which is magnetocrystalline in nature and shows fourfold symmetry at any temperature. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/1.5020072

Half-metallic perovskite oxides promise great advantages over conventional spintronic metallic materials for applications such as magnetic sensors, magnetic random access memory (MRAM) devices, magnetic tunnel junctions (MTJs), and domain wall race-track memory devices. 1 Perovskite oxides, in general, appear to be new contenders for many novel applications that were considered traditionally beyond their range. 2 The conduction mechanism in these materials, in fact, strongly depends on the interplay between orbital and spin degrees of freedom 3 that may be exploited to add multiferroic or ferroelectric functionalities. 4,6 The complexity of such a mechanism, however, can determine entangled magnetotransport response which becomes undesired in some cases. For example, it has been observed that a switchable anisotropic magnetoresistance (AMR) response in manganites may be hidden by the colossal magnetoresistance (CMR) if the magnetic anisotropy of the system is not accurately designed. 7 Irrespective of the applications, one of the key properties that need to be considered for a ferromagnetic sample is the magnetic anisotropy that dictates the magnetization reversal pathways as well as the MR output. 8 While a defined uniaxial anisotropy is essential for magnetic field sensors based on AMR, 7 a biaxial anisotropy, which provides four stable magnetization states, has the capability to encode more information (four binary bits: “00,” “01,” “10,” “11”) and can be used in memory and logic devices. 9,10 However, especially in half-metallic perovskite compounds with fourfold magnetocrystalline anisotropy in bulk, it remains difficult to avoid either strain (induced by the substrate) or shape uniaxial anisotropy in thin films. 11,12

Among manganites, La$_{0.67}$Sr$_{0.33}$MnO$_3$ (LSMO) has arisen special interest for its peculiar properties such as nearly 100% spin polarization and room temperature (RT) ferromagnetism with a Curie temperature of about Tc ≈ 370 K, hence enabling RT spintronic applications. The magnetotransport properties of these compounds in thin films are strongly affected by external perturbations such as substrate-induced strain. 13 In general, in tensile (compressive) strained films, the electron occupancy in the “e$_g$” doublet favors in-plane (out-of-plane) x$^2$-y$^2$ (3z$^2$-r$^2$) orbitals 14 determining the in-plane (out-of-plane) magnetization easy axis. 15 In addition, tensile (compressive) strain reduces (enhances) Tc with respect to the bulk value. 16,17 Therefore, the choice of the substrate for the LSMO growth is extremely important. The most commonly used single crystal substrate is SrTiO$_3$ (STO) (aSTO = 0.3905 nm) with (001) crystallographic orientation since LSMO (aLSMO = 0.387 nm) grown epitaxially on STO exhibits low tensile strain (0.82%) and has good structural and morphological properties. 18 However, in the low thickness regime, a sizeable uniaxial (twofold) anisotropy contribution 19 due to surface steps and terraces originated from the mis-cut angle of the substrate 20,21 generally hides the biaxial (fourfold) magnetocrystalline anisotropy of the LSMO film. In the higher thickness regime, LSMO on STO (001) usually shows a competition between (biaxial) bulk 12 and shape or strain (uniaxial) magnetic anisotropy. 22 Another single crystal oxide that can be used as a substrate for the growth of LSMO films is MgO with (001) crystallographic orientation. However, the lattice mismatch between LSMO...
energetic laser pulses (1.4–1.7 J/cm²) were used for transfer-
using the KrF excimer laser of wavelength 248 nm. High
sition (PLD) from commercial stoichiometric targets by
grown on the MgO (001) substrate by pulsed laser depo-
an order of magnitude at 40 K.
shows dominant biaxial anisotropy even at RT, enhanced by
netic anisotropy symmetry in our STO buffered LSMO films
quality epitaxial LSMO film. The angular dependent mag-
the misfit strain and acts as a template for depositing a good
STO as a buffer layer. The inclusion of a 12 nm STO buffer
film on the cubic MgO (001) substrate and employed 12 nm
symmetry in LSMO thin films, instead of growing it directly
material) demonstrate low residual resistivity, Metal-
temperature measurements (see Fig. S1 in thesupplementary
layer and improves the film properties. The film sur-
the STO buffer layer helps to reduce structural defects in the
420 and 362 K,
and MgO (a_{MgO} = 0.4212) results in large tensile strain
(~8%). This generally degrades the film magneto-transport
and morphological properties that limited their use.
One of the utmost challenges is to control the magnetic
symmetry in LSMO thin films, instead of growing it directly
on cubic STO (001) crystals, we deposited a 50 nm LSMO
film on the cubic MgO (001) substrate and employed 12 nm
STO as a buffer layer. The inclusion of a 12 nm STO buffer
layer on the MgO substrate not only helps in incorporating
the structural and interfacial defects in it but also minimizes
the misfit strain and acts as a template for depositing a good
quality epitaxial LSMO film. The angular dependent mag-
the system via substrate-induced strain and surface
engineering. In this letter, to define magnetic anisotropy
in LSMO films is demonstrated by the
unit cell). The epitaxial structure of the STO and LSMO
roughness of about 0.35 nm (very smooth, i.e., less than one
unit cell). The epitaxial structure of the STO and LSMO
films are 0.3903 and 0.3873 nm, respectively, which
indirectly indicate the optimal oxygen composition in the
film. In order to determine the in-plane epitaxial relation-
ship between the LSMO film and the MgO (001) substrate,
azimuthal ϕ-scans are performed around the (0–24) MgO
and (0–13) LSMO asymmetric reflections, as shown in Fig.
1(b). The peaks with a separation of 90° observed for both
the MgO substrate and the LSMO film demonstrate the four-
fold symmetry with the [00] plane of the film parallel to the
[100] plane of the substrate.
In order to investigate in detail the LSMO film structure,
we have recorded the XRD reciprocal space map (RSM)
around the asymmetric (0–13) LSMO and (0–24) MgO
reflections. Interestingly, in the low-resolution mode [Fig.
2(a)], a single diffraction peak is evident, thus inferring the
cubic structure of the film (i.e., identical in-plane lattice
parameters, a = b). However, in the high-resolution mode
[i.e., using a Ge (220) double-bounce monochromator],
the RSM around the (0–13) LSMO crystallographic reflection
shows a double-peak structure along the Qx in-plane direc-
tion [inset of Fig. 2(a)]. Such a feature indicates the presence
of two slightly different in-plane lattice parameters (i.e.,
a ≠ b), thus supporting a possible orthorhombic or mono-
clinic structure for the LSMO film. The average in-plane
lattice parameter of LSMO is 0.388 nm, which is very close
to the relaxed pseudocubic lattice value. Figure 2(b) shows
the omega scans measured around the LSMO (002) peak at
different ϕ angles (0°, 45°, and 90°). At ϕ = 0° and 90°, the

![FIG. 1.](image)

(a) Theta-2 theta XRD scan of the 50 nm thick LSMO film on the
STO buffered MgO (001) substrate and (b) asymmetrical Phi scans around
the (0-13) peak of the LSMO (001) film and the (0-24) peak of the MgO
(001) substrate show 90° separation cube-on-cube epitaxy.
presence of the double peak along the LSMO out-of-plane direction indicates, as a first approximation, the presence of two LSMO domains with the c-axis direction tilted at \(\pm 0.4^\circ\) with respect to the [001]-MgO direction. However, the rocking curve taken at \(\phi = 45^\circ\) does not show any double-peak structure, which excludes a possible rhombohedral arrangement of the LSMO unit cells. Therefore, from the phi scans \((x = \beta = 90^\circ)\) and omega scans \((\gamma \neq 90^\circ)\), we deduce that the most probable scenario is that the LSMO structure is monoclinic with in-plane fourfold symmetry and out-of-plane rigid rotations of the LSMO cell.

The magnetic anisotropy of the film also reveals a fourfold symmetry. This was investigated at RT in detail by acquiring in-plane Kerr hysteresis loops by v-MOKE in the full angular range (i.e., \(0^\circ - 360^\circ\)) while keeping fixed the external magnetic field direction. At \(\theta = 0^\circ\), the applied external magnetic field is aligned parallel to the [100] crystallographic axis of the MgO (001) substrate. By exploiting the simultaneous acquisition of two in-plane magnetization components, i.e., parallel \(M_{||}(H)\) and perpendicular \(M_{\perp}(H)\), we can deduce the symmetry of magnetic anisotropy present in the film.\(^7,11,27\) In fact, by inspecting the change of sign in \(M_{\perp}(H)\), we can accurately locate the easy axis (e.a.) and hard axis (h.a.) directions. Figure 3 presents the normalized Kerr hysteresis loops of the LSMO (001) film acquired at and around e.a. and h.a., i.e., \(\theta = 45^\circ, 90^\circ, \text{and } \pm 9^\circ\). The square loop with sharp irreversible transitions in \(M_{||}(H)\) is found at \(\theta = 45^\circ\), whereas the \(M_{\perp}(H)\) component is almost zero. This indicates that the magnetization is aligned in the film plane either parallel or anti-parallel, which corresponds to a magnetization reversal proceeding by nucleation and further propagation of magnetic domains (characteristic of an e.a. behavior). When the field is applied away from the e.a., smoother reversible transitions are found, indicating that the reversal starts by rotation followed by propagation of domains. In particular, when the magnetic field is applied around the e.a., i.e., from \(\theta = 36^\circ\) to \(54^\circ\), the magnetization switches with just one irreversible transition and from the \(M_{||}(H)\) component, we could observe that there is a significant change of sign. This means that the magnetization rotates within the film plane in clockwise to anticlockwise directions or vice-versa with respect to the applied field angle. The remanence of \(M_{||}(H)\) at the e.a. is \(M_{R,||} \cong M_S\), and the coercive field is about 1.8 mT.

The \(M(H)\) loops taken at \(\theta = 81^\circ\) and \(\theta = 99^\circ\) show that the magnetization reversal takes place in three steps, which are marked with arrows in \(M_{||}(H)\) and circles in \(M_{\perp}(H)\) [Fig. 3 (top right)], respectively. Sweeping the field from positive to negative values, the reversal occurs by a first reversible magnetization rotation, followed by two irreversible transitions that take place by nucleation and propagation of two consecutive \(90^\circ\) domain walls.\(^28\) At h.a., i.e., \(\theta = 90^\circ\), as the field strength decreases, the \(M_{||}(H)\) loop shows rotation of the magnetization followed by sharp irreversible transition and final rotation towards the applied field direction. The reversal proceeds thus with one (two) irreversible transition, related to nucleation and propagation of \(180^\circ\) (\(90^\circ\)) domain.
walls, when the field is applied close to one of the two e.a. (h.a.) orientations of magnetization. These are the typical signatures of a fourfold magnetic symmetry.\textsuperscript{27} The experimental data have been properly reproduced in the whole angular range with a modified coherent rotation Stoner-Wohlfarth model\textsuperscript{11,27} by using exclusively a single magnetic anisotropy term with fourfold symmetry whose strength was extracted from the experimental curves in the hard axis ($H_K \approx 5$ mT). Note that, since this model is based on coherent rotation, it fails to reproduce the experimental data in the regions in which nucleation and propagation of domain mechanisms dominate (i.e., close to e.a.). However, it provides a qualitative and quantitative estimation of the relevant anisotropy contributions involved (in the h.a. region).

The fourfold symmetry of the magnetic anisotropy becomes more evident by plotting the angular dependent remanence and critical fields in the full angular range (Fig. 4). The normalized remanence magnetization plots ($M_{r,1}/M_S$ and $M_{r,2}/M_S$) extracted from the experimental $M_r(H)$ and $M_r(H)$ loops at the applied field $H=0$ as a function of angle “$\theta$” are shown in Fig. 4(a). Both the magnetization components show repeated features with the periodicity of 90°. In addition, $M_{r,1}$ changes its sign for every 45°, i.e., whenever it crosses the characteristic axes. The polar plots of $M_{r,1}/M_S$ and $M_{r,2}/M_S$ are presented in panels (c) and (d) of Fig. 4, revealing the fourfold symmetry. In particular, $M_{r,1}/M_S$ resembles a butterfly structure with the highest and lowest values pointing towards e.a. and h.a. of the film, i.e., along with the [110] and [010] crystallographic directions, whereas $M_{r,2}/M_S$ shows a four lobe symmetrical shape with positive and negative values which are depicted in solid and open circles.

Figure 4(b) shows the angular dependence of the critical fields, i.e., coercive ($H_C$) and switching ($H_S$) fields, extracted from $M_r(H)$ and $M_r(H)$ loops. The value of “$H_C$” ($H_S$) is higher (lower) at e.a., i.e., [110], and decreases (increases) as it approaches towards h.a., i.e., [010]. $H_C$ and $H_S$ coincide at and around e.a. which correspond to one irreversible transition (grey shaded area), leading to 180° domain wall reversals. As the field is applied away from the e.a. (i.e., approaching the h.a., white region), $H_S$ increases and reaches the maximum exactly at the h.a. In the white shaded regions, the magnetization reversal takes place with two irreversible transitions that are related to the nucleation and propagation of two consecutive 90° domain walls. The polar plots of $H_C$ and $H_S$ are presented in Figs. 4(e) and 4(f) which show symmetrical four lobes and an asteroid shape, respectively, with 90° periodicity. From these angular dependent analyses, it is evident that the angles between two adjacent e.a. and h.a. are orthogonal to each other and the minima of $M_{r,1}$ at the consecutive h.a. have identical values, which experimentally prove that no uniaxial magnetic anisotropy contribution exists.\textsuperscript{27} These features demonstrate the fourfold symmetry of the magnetic anisotropy in the LSMO/STO/MgO (001) structure, which is similar to the magnetocrystalline anisotropy of the bulk LSMO with easy axes aligned towards 45° [110].\textsuperscript{29} The analysis of both magnetization components therefore discloses the symmetry of the magnetic anisotropy with high accuracy. Note that, even though the authors in Ref. 12 claim to have biaxial anisotropy in the LSMO/STO film, the difference between the magnetization remanence states at two consecutive h.a. reveals an additional uniaxial contribution, as explained by the authors in Refs. 19 and 27.

In order to disclose the nature of the fourfold magnetic anisotropy in our systems, we performed temperature dependent studies. The normalized Kerr hysteresis loops [Fig. 5(a)] measured at 40 K in different in-plane magnetic field directions present similar features and symmetry with respect to the RT case. The e.a. and h.a. are present at 45° [110] and 90° [010]. Near h.a., i.e., at $\theta = 70°$ and 80°, two irreversible transitions are observed owing to nucleation and propagation of two consecutive 90° domain walls. In this case, the data have been reproduced by again using a single magnetic anisotropy term with fourfold anisotropy and the anisotropy field is one order of magnitude larger than the one found at RT ($H_{K}^{b} = 40$ mT). Temperature-dependent coercive fields [Fig. 5(b)] in the angular range of 0°–180° have 90° periodicity as the RT behavior but with a tremendous increase in the coercive fields of about one order of magnitude. Therefore, as the temperature decreases from 300 K to 40 K, the signatures of biaxial anisotropy become more evident. The cause of anisotropy is due to magnetocrystalline anisotropy of LSMO, which is usually dominant at low temperatures.\textsuperscript{30}

In summary, we have fabricated high-quality epitaxial LSMO thin films on the STO buffered MgO (001) substrate by the PLD technique and studied in-plane magnetic anisotropy properties at 300 and 40 K. We demonstrated that the use of the STO buffer layer improved the quality of the LSMO film by accommodating structural defects. The LSMO film has a 4-domain monoclinic structure with domains present along [100] and [010] directions. The magnetic anisotropy also

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{Angular evolution at 300 K of the magnetic properties of the 50 nm thick LSMO film grown on the STO buffered MgO (001) substrate. (a) Normalized remanence magnetization $M_{r,1}/M_S$ (black) and $M_{r,2}/M_S$ (red) and (b) critical fields (coercivity “$H_C$” (blue) and switching “$H_S$” (magenta) as a function of the applied field angle “$\theta$” show well-defined 90° periodicity, i.e., a pure bi-axial anisotropy. The grey shaded regions in (b) indicate that the system exhibits only one irreversible transition, whereas the white regions indicate that the system exhibits two consecutive irreversible transitions. (c)–(f) Polar plots of $M_{r,1}/M_S$, $M_{r,2}/M_S$, $H_C$, and $H_S$, respectively. Positive and negative values in (d) are represented by solid and open circles.}
\end{figure}
domains, i.e., 0
absent and the h.a. is aligned along the tilted monoclinic
sistance response.
play well-defined biaxial anisotropy, which can be useful in
substrates that show anomalies in magnetic anisotropy, films
origin. Unlike the LSMO films directly grown on STO (001)
of magnitude larger at 40 K, revealing its magnetocrystalline
strength of the biaxial anisotropy increases, which is one order
results of the Stoner-Wohlfarth model.
showed biaxial anisotropy with the e.a. direction which is
aligned towards 45° or [110], where the tilted domains are
absent and the h.a. is aligned along the tilted monoclinic
domains, i.e., 0° or [100]. As temperature decreases, the
strength of the biaxial anisotropy increases, which is one order
magnitude larger at 40 K, revealing its magnetocrystalline
origin. Unlike the LSMO films directly grown on STO (001)
substrates that show anomalies in magnetic anisotropy, films
that were grown onto STO buffered MgO (001) substrates dis-
play well-defined biaxial anisotropy, which can be useful in
bit logic devices based on purely anisotropic magnetore-
sistance response.

See supplementary material for the morphology, magneti-
c, and electrical transport properties of the LSMO/STO/
MgO (001) film.

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4F. A. Cuellar, Y. H. Liu, J. Salarafzana, N. Nemes, E. Iborra, G. Sanchez-
5M. Bubes, Nat. Mater. 11, 354 (2012).

FIG. 5. (a) Normalized magnetic hysteresis loops of the 50 nm thick LSMO film grown on STO buffered MgO (001) around the first half quadrant measured at 40 K and (b) angular dependent coercive field “HC” at 40 and 300 K. The cyan and green solid lines in (a) correspond to the simulation results of the Stoner-Wohlfarth model.