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Published in:
Applied Physics Letters

DOI:
[10.1063/5.0051629](https://doi.org/10.1063/5.0051629)

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
2021

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Burema, A. A., & Banerjee, T. (2021). Temperature-dependent periodicity halving of the in-plane angular magnetoresistance in $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ thin films on LaAlO_3 . *Applied Physics Letters*, 119(1), [011901]. <https://doi.org/10.1063/5.0051629>

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Cite as: Appl. Phys. Lett. **119**, 011901 (2021); <https://doi.org/10.1063/5.0051629>

Submitted: 26 March 2021 . Accepted: 12 June 2021 . Published Online: 06 July 2021

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Submitted: 26 March 2021 · Accepted: 12 June 2021 ·

Published Online: 6 July 2021

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ABSTRACT

Strain-engineering is used as a tool to alter electronic and magnetic properties like anisotropy energy. This study reports the different angle-dependent magnetoresistance properties of the strain-engineered $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO) thin films, grown on LaAlO_3 , compared to their bulk analogs. Upon increasing temperature, a symmetry change from fourfold $[\cos(4\theta)]$ to twofold $[\cos(2\theta)]$ is observed in the angle-dependent resistance measurements. This systematic study with increasing temperature allows us to define three distinct temperature-dependent phases. The fourfold symmetric signal originates from magnetocrystalline anisotropy, whereas the twofold symmetric signal is believed to be the conventional anisotropic magnetoresistance. Our observations show that strain-engineering creates the possibility to manipulate the anisotropy, which, for example, can ultimately lead to observations of noncollinear quasi-particles like skyrmions in single layer thin films of LSMO.

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Doped manganites have been studied extensively since the first observation of the colossal magnetoresistance (CMR) effect^{1–3} in manganites. Other magnetoresistive effects, such as anisotropic magnetoresistance (AMR)^{2–6} and the planar Hall effect,⁷ have also gained attention. This interest resulted in various sensors^{8–13} and applications due to the rising popularity of Internet of Things,¹⁴ which increased the demand for electronic magnetoresistance sensors measuring anything from traffic mobility to water flow.¹² From a scientific point of view, the magnetoresistance effects form handles for measuring and/or manipulating properties of the material to form, for example, new magnetic textures like skyrmions. A first step toward creating magnetic textures can be the fine tuning of the anisotropy, which creates the ideal circumstances for finding magnetic textures.

One manganite, in particular, $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO), has attracted much attention due to its high Curie temperature of 360 K. Using pulsed laser deposition (PLD), epitaxial thin films of LSMO can be grown with minimal oxygen vacancies.¹⁵ Thickness-dependent thin film studies in LSMO led to new findings such as a shift in Curie temperature,^{16–18} coexisting magnetic phases,^{16,19} and the presence of magnetic dead layers.^{20–24}

In addition to controlling film thickness, the influence of strain induced by an underlying substrate⁶ on the angle-dependent

magnetotransport in LSMO thin films has been investigated. However, most (LSMO) films are grown on SrTiO_3 ,^{7,25–35} giving rise to tensile strain.^{36,37} Growing films on different substrates may change physical properties such as deforming the lattice, inducing oxygen octahedral tilt, or noncollinear magnetization, thus introducing novel physical properties in thin films of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$.^{20,36,38,39} On the other hand, replacing SrTiO_3 (001) (STO) with LaAlO_3 (001) (LAO) as a substrate induces a large in-plane compressive strain in the pseudocubic LSMO film, resulting in a deformation to a tetragonal structure with an elongation in the [001] direction. Compressive strain reduces Curie temperature and can result in out-of-plane magnetic anisotropy.^{36,40} The out-of-plane anisotropy can be understood as a result of compressive strain deforming the unit cell, tweaking the magnetocrystalline anisotropy (MCA) to a preferential out-of-plane direction.

Anisotropy effects are mainly studied with bulk characterization techniques, such as ferromagnetic resonance (FMR),^{35,41} and torque magnetometry,⁴² or a superconducting quantum interference device (SQUID) magnetometer^{23,42,43} to characterize the temperature-dependent magnetic anisotropy. Alternative to bulk characterization techniques, techniques, such as magnetic force microscopy,³⁶ are used to study the magnetization behavior locally (at small areas). Due to a Curie temperature above room temperature, various magnetism based

studies have been conducted with $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ at room temperature. Recent FMR studies at room temperature showed little MCA for LSMO on SrTiO_3 ,⁴¹ whereas in another study at 200 K, the MCA is clearly observed in thin films,³⁵ showing the need for systematic temperature-dependent studies. Both torque magnetometry and FMR can be used to quantify the exact anisotropy energy, whereas often it is desirable to observe and influence the effective anisotropy.

In this paper, we present the longitudinal magnetotransport data of strain-engineered LSMO thin films on LAO and investigate the temperature-dependent anisotropy, which provides information regarding the effective anisotropy.

The films used in our experiments have been grown using PLD with an excimer (KrF) laser using the previously published parameters.¹⁹ The growth of both films were monitored *in situ*, using reflection high-energy electron diffraction (RHEED), to verify a smooth epitaxial growth (see the [supplementary material](#), Fig. S1).

The thickness of the film is calculated from the Laue fringes and is found to be 9.1 nm (23 u.c.) and 5.5 nm (14 u.c.) for the thick and thin films, respectively. Laue fringes fitting on the XRD $\theta - 2\theta$ are shown in Fig. S3 of the [supplementary material](#). The thickness determined from XRD corresponds to the values obtained by counting RHEED intensity oscillations. Magnetic hysteresis loops taken at 10 and 300 K using a magnetic property measurement system (MPMS) are shown in Fig. 1(b). At 10 K, both films display typical ferromagnetic behavior with a coercive field of 30 mT and a saturation magnetization around 250 mT.

The magnetic properties of LSMO are further characterized by the temperature-dependent magnetotransport measurements. For this

purpose, the films are patterned with a Hall bar structure and etched using *aqua regia*.⁴⁴ Thereafter, we measured the temperature-dependence of the resistivity in the presence of a magnetic field. A DC of 5 μA is sent through the LSMO Hall bar to measure the longitudinal resistance in a 4-probe configuration as presented in Fig. 2(a), using a Keithley 2410. The longitudinal resistivity, ρ -T [in Fig. 2(b)], is measured while heating the sample from 10 to 300 K. The thin film (black squares) has two transition temperatures around 50 and 225 K. Below 50 and above 225 K, the film behaves as an insulator, while in the temperature range 50–225 K, the film behaves as a metal. The insulating behavior below 50 K is caused by a reduction in double exchange hopping and is more often observed in LSMO thin films of several unit cells thick.^{16,19} The transition at 225 K is close to the Curie temperature of this film (see Fig. S4 of the [supplementary material](#)). The thick film (green circles) is metallic through the entire temperature range, as depicted in Fig. 2(b). Other ρ -T measurements on the films (see Fig. S4 of the [supplementary material](#)) at higher temperatures allowed us to determine the maximum CMR to extract T_C as depicted in Table I.

For the angle-dependent magnetoresistance measurements (AdMR), the magnetic field is rotated around the [001] crystallographic axis, as shown in Fig. 2(a), while the resistance is measured in the four-probe configuration. The applied field of 0.7 T is at least one order of magnitude greater than the coercive magnetization field, as observed from the hysteresis loop in Fig. 1(b). Therefore, one can assume that the magnetization is parallel to the applied field direction ($\vec{M} \parallel \vec{B}$). The data obtained from the measurements [see Figs. 3(a) and 3(b) and [supplementary material](#) Fig. S5] are fitted with an equation based on the AMR energy terms³

$$E = A_0 + A_1 \cos^2(\theta) + A_2 \cos^4(\theta), \quad (1)$$

FIG. 1. (a) X-ray θ - 2θ diffraction of LSMO films. The Laue oscillations, clear from the thick film spectra, indicate a smooth and epitaxial growth of 23 unit cells, which is in close correspondence to the 25 unit cells observed with RHEED. (b) Normalized in-plane magnetization loops at 10 K for both films, with a coercive field of 30 mT and a saturation magnetization around 250 mT.

FIG. 2. (a) Optical microscope picture of the etched Hall bar from the thick LSMO film with the current direction either along the, equivalent, $[010]_{pc}$ or $[100]_{pc}$ pseudocubic crystallographic direction of LAO. In the picture, the contacts used in the four-wire setup are indicated. (b) Temperature dependence of LSMO's longitudinal resistivity (ρ -T) for both thin (black squares) and thick (green circles) films.

TABLE I. Summary of investigated thin films' characteristics.

	Thin film	Thick film
Thickness (nm)	5.5	9.1
H_{M_i} at 10 K (mT)	200	300
H_c at 10 K (mT)	30	30
Curie temperature (K)	195	320

where A_0 , A_1 , and A_2 are constants, and θ is the angle between current and applied magnetic field. For angle-dependent resistivity measurements, this equation is transformed into Eq. (2), as shown in literature,⁴⁵ to which we added a term to compensate for thermal drift. The resulting formula is used to fit the longitudinal resistivity

$$\rho_{\text{long}} = \rho_0 + \rho_{2f} \cos(2\theta - \delta_{2f}) + \rho_{4f} \cos(4\theta - \delta_{2f} - \delta_{4f}) + \rho_{\text{drift}} \exp[-(360 + \theta)/\tau_0], \quad (2)$$

where the longitudinal resistivity, ρ_{long} , in Eq. (2) depends on a baseline resistance ρ_0 , and on two cosine-modulated functions with amplitudes ρ_{2f} and ρ_{4f} . The terms δ_{2f} and δ_{4f} are offsets indicating the position of the magnetic hard-axis. The last term, ρ_{drift} , is added to compensate for thermal drift. More in depth information regarding the process undertaken to account for and eliminate the thermal drift effect is discussed in [supplementary material](#) Sec. 3. [Figures 3\(a\)](#) and [3\(b\)](#) show the angle-dependent resistivity and their respective fittings with Eq. (2) for the thin (black squares) and the thick (green circles) films.

[Figure 3\(c\)](#) shows the normalized AdMR ($\rho(\theta)/\rho_0 \times 100\%$) at different temperatures for the thin film. At temperatures 10–125 K, a

fourfold symmetry in the resistance is observed. Upon increasing the temperature, at 100 K, the signal starts to show an asymmetry. Increasing the temperature, from 125 to 250 K, causes the signal to show a twofold symmetry, which peaks close to the CMR maximum, at 195 K, for the thin film. Above 250 K, no angle-dependent signal is observed. For the thick film in [Fig. 3\(d\)](#), we observed at low temperatures no dominant periodicity. Upon increasing the temperature, the twofold signal becomes dominant around 150 K and remains so until 300 K. The twofold signal present in both films is observed in various magnetic materials and depends directly on the applied magnetic field strength and the angle between the applied magnetic field and the current direction.^{3,5}

To quantify the change in signal with temperature, the extracted amplitudes from a fit with Eq. (2) are plotted vs temperature in [Figs. 4\(a\)](#) and [4\(b\)](#). To follow similar conventions as in published literature, [Figs. 4\(c\)](#) and [4\(d\)](#) show the AMR percentage vs temperature based on the conventional AMR definition

$$\text{AMR} \stackrel{\text{def}}{=} \frac{\rho(90) - \rho(0)}{\rho(0)} \times 100\% \approx \frac{2A_{2f}}{\rho_0} \times 100\%, \quad (3)$$

where the AMR is defined as twice the extracted amplitude from Eq. (2) divided by ρ_0 . This definition is used for both the two- and fourfold signals in [Figs. 4\(c\)](#) and [4\(d\)](#).

The thin film, in [Figs. 4\(a\)](#) and [4\(c\)](#), confirms that the dominant signal is fourfold symmetric below 125 K. The observed peak at 50 K, in [Fig. 4\(c\)](#), is due to the increase in resistance below 50 K, decreasing the AdMR percentage for the measurements at 10 and 25 K. The twofold signal for both films shows a maximum around Curie temperature, which is expected for the conventional AMR effect.^{3,5} In addition, the CMR, shown in [supplementary material](#) Fig. S4, shows

FIG. 3. (a) and (b) AdMR signal at 50 and 250 K, respectively, for the thin (black squares) and thick (green circles) films with a fit (solid line) based on Eq. (3). For (c) and (d), the plotted resistance is corrected for thermal drift and normalized, $\rho(\theta)/\rho_{\text{average}}$, for multiple temperatures. Note that the indicated axes are allowed to be rotated by 90° as the axes $[010]$ and $[100]$ are equivalent. Plots (e) and (f) are the normalized data of (c) and (d), plotted in the polar plot format, respectively. The plot (e) shows a clear transition from fourfold to twofold without any change in peak positions, indicating that there is no magnetic easy-axis transformation. The legend in the right bottom corresponds to [Figs. 3\(c\)–3\(f\)](#).

FIG. 4. The thin film, in (a) and (c), shows three distinct phases with temperature. (a) The amplitude of the fitted signal. The fourfold amplitude is stable in size, around 0.4 mΩ cm, up to 50 K, after which it decreases to zero at 200 K. The thick film, in (b) and (d), shows two distinct regions. (c) and (d) The AMR strength based on Eq. (3). (all) The closed and open symbols correspond to the clockwise or counterclockwise rotation in the measurement, respectively.

the same trend in signal strength as does the conventional AMR, as commonly observed.

Similar trends, comparable to the temperature scaling, for both the two- and fourfold signals are shown with increasing magnetic field strength in [supplementary material](#) Fig. S6. The temperature and magnetic field dependence, for the fourfold symmetric signal, can be explained by a constant anisotropic energy in the LSMO thin film. The constant anisotropy energy ($E_{ani}|_{0K,0T} = E_{100} - E_{110}$) can be overcome by increasing either temperature, applied magnetic field, or thickness. Based on the crystallographic directions of the LaAlO_3 substrate and the Hall bar placement, the observed fourfold signal in our measurements follows the in-plane bi-axial magnetic easy axes along the $[110]_{pc}$ and $[\bar{1}\bar{1}0]_{pc}$ directions, similar to the fourfold observations reported previously in literature.^{6,46–48} Based on the temperature scaling in [Fig. 4\(a\)](#) and on existing literature review, we attribute the fourfold symmetric signal to the tetragonal MCA, as it fits all observed features.

In [Fig. 4](#), we observe a dominant MCA signal in the thin film, but not in the thick film. A possible explanation stems from a change in the conduction mechanism of LSMO. The two major conduction mechanisms found in LSMO are, first the double exchange hopping causing electron–electron ($\sim \rho T^2$) and electron–magnon ($\sim \rho T^{4.5}$) scattering below T_C , and second the polaronic conduction via hopping above T_C . Decreasing film thickness causes the resistance to increase, attributed to a decrease in the semiconducting bandwidth of the itinerant d-electrons.⁴⁹ The decrease in thickness eventually leads to a thermally activated hopping ($\sim e^{E_a/T}$),¹⁶ as observed below 50 K in our thin film. Rotating the magnetic field, for example, by orienting the magnetic field along an easy or hard axis, decreases or increases, respectively, the required hopping energy due to the MCA contribution. The net effect, MCA has on the conduction, is larger for resistive

films. This effect is intuitively understood for a metallic film, where the activation energy is zero; therefore, hopping is always possible irrespective of the orientation of the applied magnetic field.

To conclude, we report the following on the temperature-dependent AdMR signal vs temperature: At low temperature, the fourfold signal appears to be directly related to the MCA, with easy axes in $[110]_{pc}$ and $[\bar{1}\bar{1}0]_{pc}$ directions. At higher temperatures, a twofold signal is observed, caused by the conventional AMR signal of ferromagnetic compounds. We relate the increase in signal strength for AdMR and CMR to the increasing resistivity of the films caused by the decreasing thickness. These results indicate the feasibility to tune strain and thickness to control the anisotropy. Based on the results, we predict resistance changes due to strain-engineering to be best observed in thin samples, which still portray metallic behavior. Future experiments can aim to engineer an out-of-plane magnetization to deeper understand the relation between strain and the (out-of-plane) MCA observed by magnetotransport. Inducing a larger compressive strain on a thin (5.5 nm) film of LSMO can induce an out-of-plane anisotropy to facilitate the formation of magnetic textures like skyrmions in a single layer LSMO.

See the [supplementary material](#) for RHEED, magnetic hysteresis curves, fittings, and the full normalized AdMR data.

This work was realized using NanoLab NL facilities and is a part of the research program Skyrmionics: Towards New magnetic Skyrmions and Topological Memory (Project No. 16SKYR04). A.A.B. acknowledges financial support from the Netherlands Organisation for Scientific Research (NWO). We thank G. R. Blake, M. V. Mostovoy, A. Das, D. Ottomano, J. J. L. van Rijn, E. P. Vallabhaneni, P. Zhang, A. S. Goossens, and S. Chen for useful scientific discussions and J. G. Holstein and H. H. de Vries for technical assistance.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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