In-plane reversal of the magnetic anisotropy in (110)-oriented $LaCoO_3$/$La_{0.67}Sr_{0.33}MnO_3$ heterostructures

Jing Zhang, Xi Yan, Furong Han, Jine Zhang, Dan Liu, Baogen Shen, and Jirong Sun

Citation: AIP Advances **8**, 055809 (2018); doi: 10.1063/1.5007169
View online: https://doi.org/10.1063/1.5007169
View Table of Contents: http://aip.scitation.org/toc/adv/8/5
Published by the American Institute of Physics

In-plane reversal of the magnetic anisotropy in (110)-oriented $LaCoO_3/La_{0.67}Sr_{0.33}MnO_3$ heterostructures

Jing Zhang,[1,2] Xi Yan,[1,2] Furong Han,[1,2] Jine Zhang,[1,2] Dan Liu,[1,2] Baogen Shen,[1,2] and Jirong Sun[1,2,a]

[1]*State Key Laboratory of Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, People's Republic of China*

[2]*University of Chinese Academy of Sciences, Beijing 100049, People's Republic of China*

(Presented 8 November 2017; received 30 September 2017; accepted 30 October 2017; published online 18 December 2017)

The interface engineering of the complex oxides with strongly coupled degrees of freedom opens a wide space for the exploration of novel effects. $La_{0.67}Sr_{0.33}MnO_3$ is one of the most typical complex oxides used for atomic level material engineering. Herein we reported an in-plane reversal of the magnetic anisotropy in (110)-oriented $LaCoO_3/La_{0.67}Sr_{0.33}MnO_3$ (LCO/LSMO) bilayers grown on (110)-oriented $LaAlO_3$ substrates. Fixing the LSMO layer thickness to 8 nm and varying the LCO layer from 0 to 8 nm, totally six bilayers were fabricated. Without the LCO layer, the LSMO film exhibits an easy axis along the [1-10] direction. However, when the thickness of the LCO layer exceeds 1 nm, a signature of spin-reorientation appears; the easy axis turns from the [1-10] to the [001] direction below 225 K. This tendency is continuously enhanced by increasing the LCO. We reveal that lattice strains are different along these two directions. The magnetic anisotropy is not only controlled by lattice strain but also by structural distortion at interface. This work shows the great potential of the interface engineering with differently structured oxides for the exploration of novel functional materials. https://doi.org/10.1063/1.5007169

I. INTRODUCTION

The hole-doped manganites $R_{1-x}A_xMnO_3$ (R=trivalent rare-earth ion, A=divalent alkaline-earth ion), have received extensive attentions because of their unusual properties.[1–3] LSMO is attractive due to its colossal magnetoresistance effect (CMR),[4] high Curie temperature,[5] and its possible application to magnetic tunnel junction devices.[6,7] Unlike the bulk LSMO, the epitaxial LSMO films could be intentionally strained, resulting in a coupling between lattice strains and the spin, orbital, charge degrees of freedom, and finally affect magnetic anisotropy (MA).[8] Suzuki *et al.* have systematically studied the MA of LSMO films on different substrates,[9] and showed that lattice mismatch was the main cause to affect the MA. What' more, they claimed that MA of the films with the [110] and [001] orientations were twofold and fourfold symmetric,[10] respectively, and obtained a MA energy about $K_1=8.4\times10^4$ ergs/cm^3, higher than that of the LSMO single crystals ($K_1=1.8\times10^4$ergs/cm^3). Lecoeur *et al.*[11] have investigated the MA of $La_{0.7}Sr_{0.3}MnO_3$ films with Kerr microscopy. In addition to MA, the effects of lattice strains on magnetoresistive properties was also studied for epitaxial LSMO.[12,13] In these works, the adopted LSMO films were usually thick (30 nm to 500 nm) and usually focused on the (001)-orientation, for which the lattice strains are fourfold symmetric. However, the (110)-oriented manganite films deserve special attention, because the in-plane lattice strains along the [1-10] and

[a]Corresponding author: jrsun@iphy.ac.cn

[001] directions are no longer equivalent. This will provide us an opportunity to tune in-plane MA via strain engineering. LSMO-based multilayers are also attractive and the oxygen octahedra near interface can be tilted/distorted by interlayer coupling, causing anisotropic magnetic and transport behaviors.[14–16] Therefore, it is worthwhile to investigate the combination of lattice strains and oxygen octahedra tilting to effect the MA for the (110)-oriented LSMO-based bilayers.

Herein we are interested in the bilayers composed of the paramagnetic LCO and ferromagnetic LSMO. We fabricated different LCO/LSMO bilayers on (110)-oriented LAO. The x-ray diffraction (XRD) measurements indicated the epitaxial growth of our films, without lattice relaxation. A spin-reorientation characterized by the change of the easy axis from the original [1-10] to [001] was observed in the cooling process when the layer thickness of LCO exceeds 1 nm. This is different from the plain LSMO films which exhibit an easy axis along the [1-10] direction on (110)-LAO.[17] Our work indicated that the lattice strains were not the only factor affecting the MA of manganite films, and also the interlayer coupling played an important role.

II. EXPERIMENTS

LCO(t)/LSMO(8nm) bilayers were grown by the pulsed laser ablation technique with a 248 nm KrF excimer laser on (110)-oriented LAO substrates (3×5×0.5 mm^3). The adopted laser energy density was kept at 2 Jcm^{-2}, and the repetition rate was 2 Hz. The growth temperature was kept at 730°C and the oxygen pressure was fixed to 30 Pa. The LSMO layer was deposited first.

To calibrate deposition rate, a LSMO or LCO film about 30 nm was deposited, and its thickness was accurately measured by small angle X-ray reflectivity (XRR, as shown in Fig. 1(a)). The crystal structure and orientation of the film was determined by high-resolution XRD (D8 Discover, Cu Kα radiation). Magnetic measurements were performed on the Quantum-Designed vibrating sample magnetometer (VSM-SQUID), in the temperature interval from 10 K to 380 K.

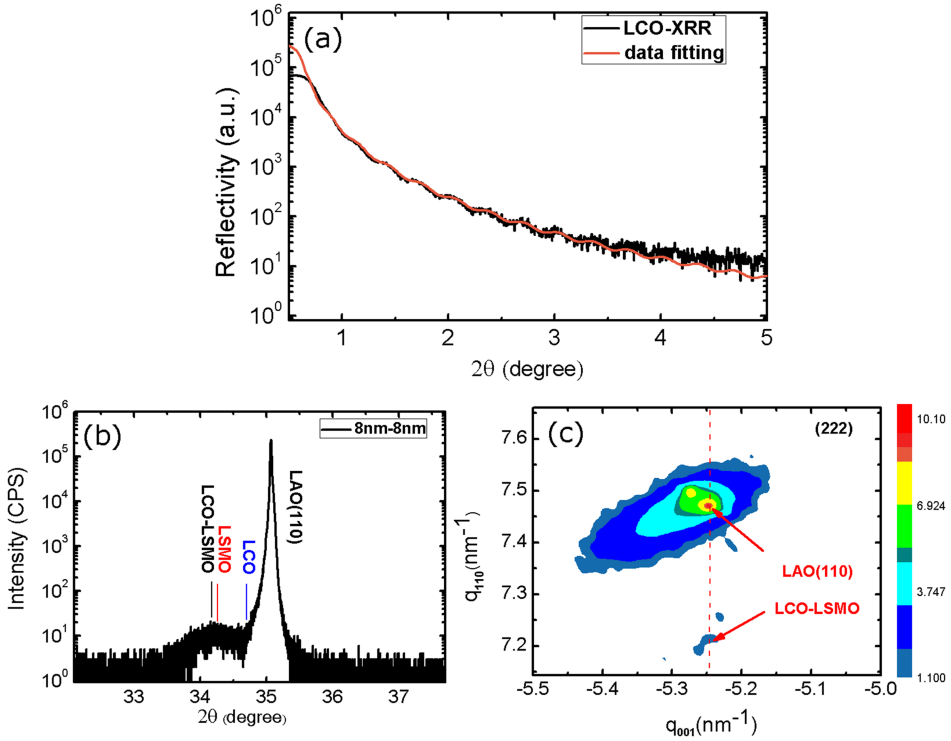

FIG. 1. (a) Small angle x-ray reflectivity of the LCO single layer and the corresponding results of data fitting that gives layer thickness. (b) XRD θ-2θ scan patterns of LCO (8nm)/LSMO(8 nm)/LAO (110). (c) Reciprocal space mapping (RSM) of the (222) reflection of LCO(8nm)/LSMO(8 nm)/LAO (110).

III. RESULTS AND DISCUSSION

As an example, in Fig. 1(a) we show the XRR spectrum and the fitting of single LCO/LAO(110) film. The regular and periodic oscillation indicates a smooth film surface and a homogeneous film thickness. Using Fast Fourier Transition fitting, the LCO thickness of 26.4 nm is deduced. Combined with deposition time, we got the growth rate of LCO and determined LSMO growth rate in the same way.

As a representative, in Fig. 1(b) we presented the θ-2θ XRD pattern of LCO(8nm)/LSMO(8nm) bilayers. Besides the (110) reflection of LAO substrate (~35.08°), a minor and broad diffraction peak can be clearly seen at a lower angle of 34.17° (marked by a red line). It belongs to the LCO/LSMO bilayers, which does not coincide with the single LCO (3.83 Å, marked by a blue line), but almost close to LSMO layer (3.87 Å, marked by a red line). We also measured the θ-2θ spectra of other bilayers (not shown here), and obtained similar results. According to the Bragg diffraction equation: $2d\sin\theta=\lambda$, we can calculate the out-of-plane lattice constant c=3.878Å of the film, which is slighter larger than the LAO substrate. On the basis of the invariant cell volume, it is concluded that the LCO/LSMO bilayers are slightly in-plane compressive and out-of-plane tensile.

In order to get further information on lattice strains, Fig. 1(c) exemplifies the reciprocal space mapping (RSM) of LCO(8nm)/LSMO(8 nm) of the (222) reflection. It is evident that the reflections of bilayer and substrate aligned vertically, meaning no lattice relaxation.

To determine the magnetic properties of the bilayers, we measured the temperature dependences of the magnetic moment (M-T relation) along [1-10] and [001] directions, respectively, with an applied field of 0.01 T in field-cooling (FC) mode. As shown in Fig. 2(a), along the [001] orientation we observed a magnetic behavior along hard axis when t=0. The magnetic moment is small even at very low temperature and undergoes a ferromagnetic to paramagnetic transition at ~310 K (marked by a red arrow), which is consistent with previous reports.[5,17] As the LCO layer becomes thicker, the magnetic moment grows rapidly, meaning that the spins become to favor the [001] direction when LCO is thick.

In sharp contrast to [001] direction, the LSMO shows a typical ferromagnetic behavior along the [1-10] direction when t=0 (Fig. 2(b)). It takes the highest value at low temperature, and slowly first and rapidly then when close to T_C. Obviously, the [1-10] direction is the easy axis of the (110)-LSMO film. As increasing the LCO, the magnetic moment displays a tendency towards decrease. This phenomenon is especially obvious when t>1 nm. A second magnetic transition appears when the sample is cooled down from 380K to ~225 K (marked by blue triangle), resulting in a steep decrease in M. This temperature is not the T_C of LCO (75 K),[18] and is resulted from spin-reorientation as will be discussed in detail later. We failed to find out the magnetic transition of the LCO layer. There are two possible reasons: firstly, the magnetic moment of the LCO layer much lower than that of the LSMO layer; secondly, the magnetic order in the LCO layer on LAO is weak.

Compared the data in Fig. 2(a) and 2(b), we found that the M-T curves along the two directions are mutually complementary; when the magnetic moment increases in one direction it will decrease

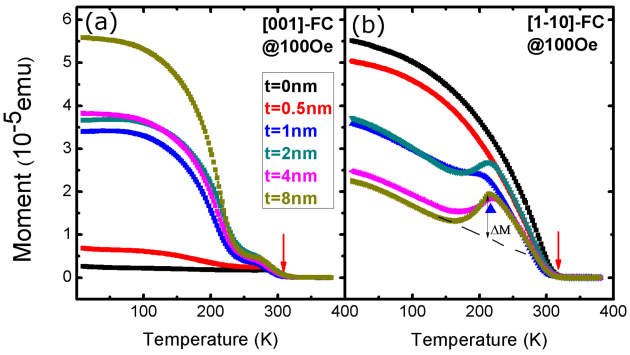

FIG. 2. The temperature-dependent magnetization of LCO (t nm)/LSMO (8 nm)/LAO (110) bilayers along (a) [1-10] and (b) [001].

in another direction, and vice verse. These results reveal the strong effect of the LCO layer on the MA of the LSMO layer. Without LCO, [1-10] is easy axis. When $t>1$, the easy axis is still in the [1-10] direction at high temperatures and changed to the [001] direction when below 225 K. At about 225 K, the LSMO layer in the bilayers undergoes a spin-reorientation which is absent in plain manganite films. Simply extrapolating the low temperature part of M-T curve to 310K, the ΔM is obtained, which actually presenting the existence of spin-reorientation.

As will be seen later, the easy/hard axis is indeed closely related to the relative thicknesses of the LCO and LSMO layers. By adjusting the layer thickness of LCO, we realized a reversion of the biaxial MA of LSMO.

To confirm the spin-reorientation suggested by the *M-T* curve, we performed further magnetic measurements at 10K. Fig. 3(a)–(f) display the magnetic loops of different samples, recorded with in-plane filed along [001] and [1-10] directions, respectively. All samples exhibit the typical FM behavior, showing a gradual transition of the easy axis with *t*. Based on the M-H curves in Fig. 3, we can give an intuitive description of the in-plane MA. When LCO is thin (t=0 and 0.5 nm), Fig. 3(a) and 3(b), the magnetic behavior is dominated by LSMO, and the [1-10] direction is the easy axis compared with [001]. This result indicates that, under compressive strains, spins of the LSMO layer prefer to lie along the [1-10] direction. This is a conclusion similar to that obtained by other groups.[17] When t=1 nm, the magnetic moments at the two directions are comparative. Possibly, the LCO cap layer and LAO substrate jointly compress the intermediate LSMO layer, causing the spins to aligning nearly equally long two directions. Interestingly, increasing the LCO thickness further, it is difficult to decide which direction is the easy axis intuitively.

To figure out the easy axis as LCO changes, we analyzed the coercive force (Hc) along two directions in Figure 4(a). The Hc increases with t along [001] (230Oe to 4000Oe), with a jump at about 1nm, but decline at [001] (1300Oe to 850Oe), meanwhile t=1 is critical point for spin-reorientation along [001] with Hc<1kOe. Deduced from Figure 2(b), we get the ΔM along [1-10], presenting the existence of spin-reorientation, which appearance indicates the [1-10] turned to hard axis. According to Fig. 4(b), the Ms are nearly at 3μB/Mn and the ΔM is perfectly in accordance with

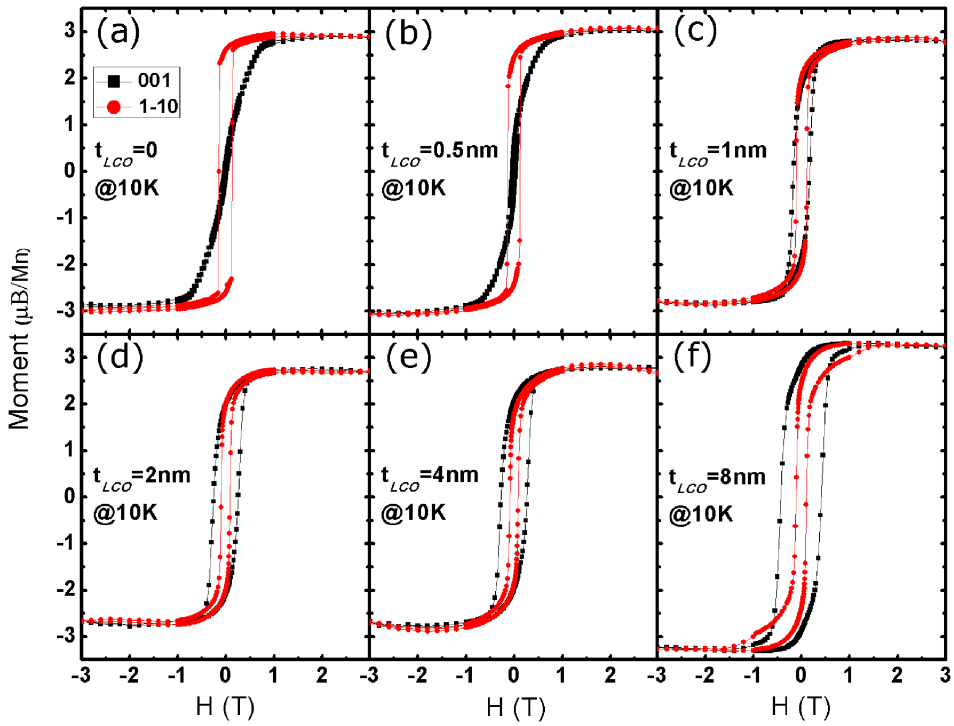

FIG. 3. The magnetic field dependence of the magnetization of LCO(*t*)/LSMO(8nm)/LAO(110) films. (a) t=0; (b) t=0.5; (c) t=1; (d) t=2; (e) t=4; (f) t=6.

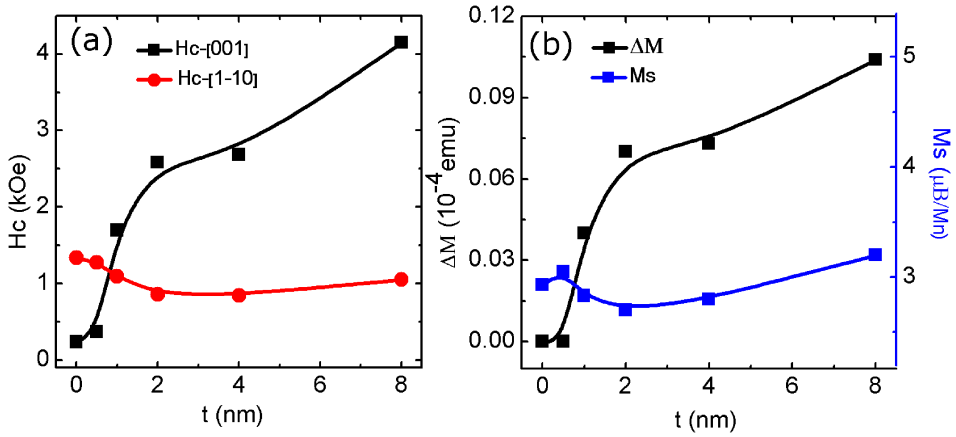

FIG. 4. The coercive force (Hc) along two directions (a) and magnetization (Ms) as a function of the LCO thickness t (b).

the Hc of [001] direction, indicating that the spin-reorientation occurs at the same time with the jump of the Hc of [001] direction. But the high value of Hc when t>1 nm makes the easy axis reversal unapparent. So we infer that increasing the LCO thickness further, [001]-axis gradually becomes the easy axis compared to the [1-10]-axis. However the high Hc(>1kOe) overspreads the easy axis for [001], which can be deduced from the existence of spin-reorientation. These results indicate that the LCO layer, together with the substrate, has modulated the arrangement of the magnetic moment of LSMO, i.e., the compressive strain is not the only reason determining magnetic orientation.

IV. CONCLUSIONS

In summary, we have carried out a systemic study on the correlation between the LCO thickness and the in-plane MA of LSMO films grown on LAO(110) substrates. As the temperature cooled, we find an interesting spin reorientation occurred in LCO/LSMO bilayers, which was absent either in single LCO layer or LSMO layer. What's more, we found that the magnetic anisotropy was sensitive to the LCO thickness. An in-plane reversed magnetic anisotropy of LSMO film by varying the layer thickness of LCO was obtained for first time. Combined the strain from substrate and LCO layer, we can conclude that: 1. strain along two directions is different, even though both are compressive. 2. strain is not the only incentive for spin arrangement. Moreover, there must be something special occurred at the LSMO and LCO interface, which maybe the structure distortion. The present work shows the great potential of the interface engineering to explore novel functional materials.

ACKNOWLEDGMENTS

This work has been supported by the National Basic Research of China (2016YFA0300701), the National Natural Science Foundation of China (11520101002, 51590880, 11374348, 11134007, 11574376, and 11574366), and the Key Program of the Chinese Academy of Sciences.

[1] S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science **264**, 413 (1994).
[2] E. Dagotto, T. Hotta, and A. Moreo, Physical Reports **344**, 1 (2001).
[3] J. C. Loudon, D. M. Neil, and A. M. Paul, Nature **420**, 797 (2002).
[4] A. Sadoc, B. Mercey, C. Simon, D. Grebille, W. Prellier, and M. Lepetit, Physical Review Letters **104**, 046804 (2010).
[5] A. Urushibara, Y. Moritomo, T. Arima, A. Asamisu, G. Kido, and Y. Tokura, Physical Review B **51**, 14103 (1995).
[6] R. Werner, A. Yu. Petrov, L. Alvarez Miño, R. Kleiner, D. Koelle, and B. A. Davidson, Applied Physics Letters **98**, 162505 (2011).
[7] M. Bowen, M. Bibes, A. Barthélémy, J. P. Contour, A. Anane, Y. Lemaître, and A. Fert, Applied Physics Letters **82**, 233 (2003).
[8] H. Boschker, M. Mathews, E. P. Houwman, H. Nishikawa, A. Vailionis, G. Koster, G. Rijnders, and D. H. A. Blank, Physical Review B **79**, 214425 (2009).
[9] Y. Suzuki, H. Y. Hwang, S.-W. Cheong, and R. B. van Dover, Applied Physics Letters **71**, 140 (1997).
[10] Y. Suzuki, H. Y. Hwang, S.-W. Cheong, T. Siegrist, R. B. van Dover, A. Asamitsu, and Y. Tokura, Journal of Applied Physics **83**, 7064 (1998).

[11] P. Lecoeur, P. L. Trouilloud, G. Xiao, A. Gupta, G. Q. Gong, and X. W. Li, Journal of Applied Physics **82**, 3934 (1997).
[12] S. Jin, T. H. Tiefel, M. McCormack, H. M. O'Bryan, L. H. Chen, R. Ramesh, and D. Schurig, Applied Physics Letters **67**, 557 (1995).
[13] C. Kwon, M. C. Robson, K. C. Kim, J. Y. Gu, S. E. Lofland, S. M. Bhagat, Z. Trajanovic, M. Rajeswari, T. Venkatesan, A. R. Kratz, R. D. Gomez, and R. Ramesh, Journal of Magnetism and Magnetic Materials **172**, 229 (1997).
[14] M. Ziese, I. Vrejoiu, E. Pippel, P. Esquinazi, D. Hesse, C. Etz, J. Henk, A. Ernst, I. V. Maznichenko, W. Hergert, and I. Mertig, Physical Review Letters **104**, 167203 (2010).
[15] A. Sadoc, B. Mercey, C. Simon, D. Grebille, W. Prellier and M. Lepetit, Physical Review Letters **104**, 046804 (2010).
[16] Z. Liao, M. Huijben, Z. Zhong, N. Gauquelin, S. Macke, R. J. Green, S. Van Aert, J. Verbeeck, G. Van Tendeloo, K. Held, G. A. Sawatzky, G. Koster, and G. Rijnders, Nature Materials **15**, 425 (2016).
[17] F. Tsui, M. C. Smoak, T. K. Nath, and C. B. Eom, Applied Physics Letters **76**, 2421 (2000).
[18] D. Fuchs, E. Arac, C. Pinta, S. Schuppler, R. Schneider, and H. v. Löhneysen, Physical Review B **77**, 014434 (2008).