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## Buffer layer-induced positive magnetoresistance in manganite-based heterojunctions

W. W. Gao,<sup>1</sup> W. M. Lü,<sup>1</sup> A. D. Wei,<sup>1</sup> J. Wang,<sup>1</sup> J. Shen,<sup>2</sup> B. G. Shen,<sup>1</sup> and J. R. Sun<sup>1,a)</sup>

<sup>1</sup>Beijing National Laboratory for Condensed Matter Physics and the Institute of Physics, Chinese Academy of Sciences, Beijing 100190, People's Republic of China

<sup>2</sup>Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, People's Republic of China

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Effects of the LaMnO<sub>3</sub> (LMO) buffer layer on the magnetoresistive behaviors of La<sub>0.67</sub>A<sub>0.33</sub>MnO<sub>3</sub>/LaMnO<sub>3</sub>/SrTiO<sub>3</sub>:0.05 wt% Nb (LAMO/LMO/STON, A = Ca, Sr) have been experimentally studied. In addition to an enhanced response to a magnetic field, the current-voltage relations show a downward shift in magnetic field, indicating an increase of the junction resistance. It is completely different from that observed in the junctions without buffer layer. The positive magnetoresistance (MR) is strongly dependent on the thickness of the LMO layer, increasing first then decreasing with the increase of layer thickness. Furthermore, it is significantly stronger in LCMO/LMO/STON than in LSMO/LMO/STON. The maximal MR at 50 K is ~90% for LCMO/LMO/STON and ~52% for LSMO/LMO/STON, occurring at the LMO thickness of 4 nm under the field of 5 T. The MR persists up to 350 K, and it is ~30% and ~24% for the LCMO and LSMO junctions, respectively. An analysis of the current-voltage characteristics indicates an increase in interfacial barrier in magnetic field, which is the origin for the positive MR. © 2012 American Institute of Physics. [doi:10.1063/1.3673858]

### I. INTRODUCTION

The heterojunctions composed of manganites and SrTiO<sub>3</sub>:Nb have attracted remarkable attention in recent years due to their excellent rectifying characteristics<sup>1</sup> and interesting properties such as bias-dependent magnetoresistance (MR)<sup>2,3</sup> and significant photoelectronic behaviors.<sup>4</sup> Because of the order-disorder transition of the spin, charge, and orbital degrees of freedom<sup>5</sup> and the strong magnetic-conductive correlation existing in the manganites,<sup>6</sup> magnetic field is expected to have an important impact on the behaviors of the manganite-based heterojunctions, which may greatly enhance the functionality of the corresponding devices. However, the magnetic field effects have been severely depressed in manganite-based devices such as the manganite junctions due to the presence of inactive interfacial layer. In order to incorporate the unusual properties of the manganite into corresponding the devices, the interfacial layer must be activated. As reported, the interfacial ferromagnetism of the La<sub>0.6</sub>Sr<sub>0.4</sub>MnO<sub>3</sub> film on SrTiO<sub>3</sub> can be recovered by introducing a LaMnO<sub>3</sub> buffer layer.<sup>7</sup> This means that a proper intermediate layer can modify the interface state, thus the physical properties of junctions significantly. In fact, the magnetic field induced positive MR has already been found by Lü *et al.*<sup>8</sup> in La<sub>0.67</sub>Ca<sub>0.33</sub>MnO<sub>3</sub>/LaMnO<sub>3</sub>(*t*)/SrTiO<sub>3</sub>:0.05 wt% Nb junctions, the enhanced positive MR is obtained, which is totally different from the negative MR effect in the simple junctions without a buffer layer.

We noted that this work has been conducted only for the LCMO-based junction. La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> (LSMO) is different

from LCMO in many aspects. For example, the Jahn-Teller distortions in the latter are less strong and, furthermore, the lattice mismatch between LSMO, LMO, and STON are less severe. It is therefore worthwhile to check whether the LMO-caused positive MR remains in the LSMO-based junctions. This work can provide us further information on the effect of interface decoration, which may lead to a deep understanding of this phenomenon. Based on these considerations, in this paper, we perform the systematic study on the effects of the LMO layer on the magnetoresistive behaviors of La<sub>0.67</sub>A<sub>0.33</sub>MnO<sub>3</sub>/LaMnO<sub>3</sub>/SrTiO<sub>3</sub>:0.05 wt% Nb (LAMO/LMO/STON, A = Ca, Sr), with different LMO layer thickness. In addition to an enhanced positive MR, the magnetic field effect is further found to be much stronger in LCMO/LMO/STO than in LSMO/LMO/STON.

### II. EXPERIMENTAL PROCEDURE

Two series of manganite-based heterojunctions, LAMO/LMO(*t*)/STON (A = Ca, Sr), were fabricated by growing, using the pulsed laser ablation technique, first a LMO layer with a thickness of 0 ~ 12 nm then a LAMO film of ~150 nm on the (100) SrTiO<sub>3</sub>:0.05 wt% Nb (STON). During the deposition, the substrate temperature was kept at 720 °C and the oxygen pressure at ~10 Pa, ~40 Pa, ~80 Pa for the LMO, LSMO, LCMO films, respectively. The film thickness was controlled by deposition time.

The x-ray diffraction (XRD) results indicate the LAMO films are single phase and epitaxially grown (not shown here). The surface morphology of LMO layer shows that the film is quite flat with a terrace-structured surface. The peak-to-valley fluctuation is ~0.4 nm and the root-mean-square roughness is ~0.13 nm for, for example, the LMO film

<sup>a)</sup>Author to whom correspondence should be addressed. Electronic mail: jrsun@iphy.ac.cn.

of 6 nm. This will guarantee the high quality of the top LAMO films.

As electrodes, two Cu pads were deposited on the top LAMO film and the STON substrate, respectively. The contacts of Cu-STON and Cu-LCMO (LSMO) film were ohmic. The current-voltage ( $J$ - $V$ ) characteristics were measured using a two probe method. The  $J$ - $V$  curves were recorded by a superconducting quantum interference device magnetometer.

### III. RESULTS AND DISCUSSIONS

The semilogarithmic plot of the  $J$ - $V$  relations for the LAMO/LMO( $t$ )/STON junctions with  $t = 1$  nm and 4 nm are presented in Figs. 1(a)–1(d). At first glance, the rectifying characters are excellent for all junctions, the current is very small even for the backward bias up to  $-6$  V, while growing rapidly for the forward bias beyond a threshold value, as shown in the inset plots of Figs. 1(a) and 1(c). An excellent linear relation between  $\log J$  and  $V$  is obtained for the LMO layer below 6 nm. This means that the incorporation of the LMO layer does not affect the rectifying behaviors of the junctions. However, an enhancement in leakage current was observed in the LAMO/LMO( $t$ )/STON for  $t > 10$  nm.

As well documented, the MR of the simple manganite junction is negative and quite low.<sup>3,9</sup> Through introducing an LMO layer, we obtained positive MR under the forward bias voltage. In Fig. 1 for the LAMO/LMO( $t$ )/STON ( $A = \text{Ca, Sr}$ ) junctions, the  $\log J$ - $V$  curves show an obvious downward shift in magnetic field when the junctions are positively biased, indicating the positive MR, defined as  $J(0)/J(5\text{ T}) - 1$ . The magnetic field response of the  $J$ - $V$  curves is strongly dependent on the LMO thickness. It is weak without LMO and strong in the presence of LMO. These features are observed in both the LCMO/LMO/STON and

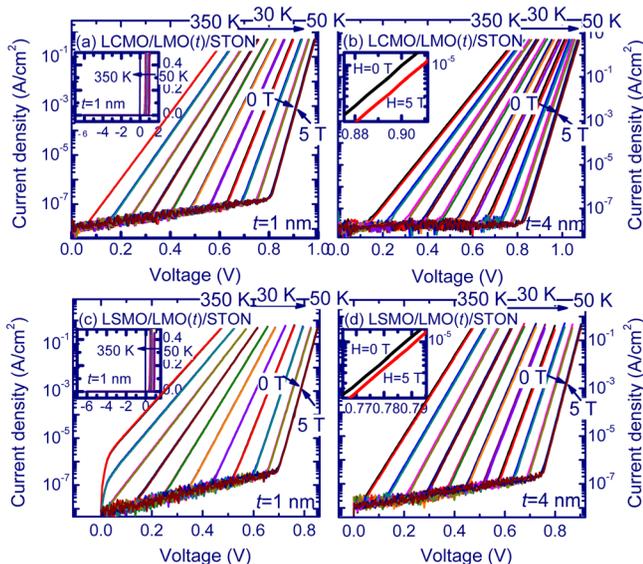


FIG. 1. (Color online) Semilogarithmic plot of the  $J$ - $V$  relations of the LCMO/LMO( $t$ )/STON junctions with  $t = 1$  nm (a) and  $t = 4$  nm (b) and the LSMO/LMO( $t$ )/STON junctions with  $t = 1$  nm (c) and  $t = 4$  nm (d), measured with and without magnetic field in the temperature range from 50 K to 350 K. The inset plots in Figs. 1(a) and 1(c) are the  $J$ - $V$  curves in linear scale accordingly, and 1(b) and 1(d) are close views of the  $J$ - $V$  curves at 80 K.

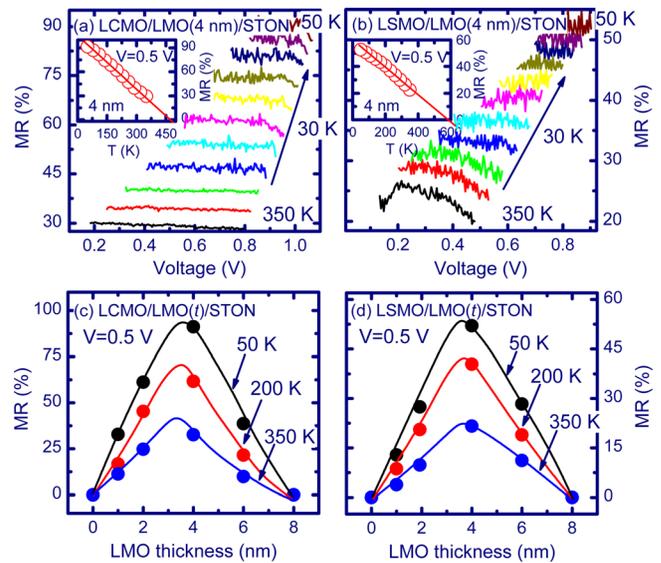


FIG. 2. (Color online) MR as a function of bias voltage for the LCMO/LMO(4 nm)/STON (a) and LSMO/LMO(4 nm)/STON (b) junctions measured in the temperature range from 50 K to 350 K. MR as a function of the LMO thickness, obtained at 0.5 V for the LCMO/LMO( $t$ )/STON (c) and LSMO/LMO( $t$ )/STON (d) junctions. The data for  $t = 6$  nm are extracted from the low bias process. The inset plots in Figs. 2(a) and 2(b) show the temperature dependence of the MR of LAMO/LMO(4 nm)/STON junctions at 0.5 V.

LSMO/LMO/STON junctions, although difference between the two series of junctions does exist. This indicates the generality of the positive MR in LMO buffer layer modified junctions.

To get a quantitative idea on the magnetic field effect, the MR of LCMO(LSMO)/LMO(4 nm)/STON junctions are presented in Figs. 2(a) and 2(b). The inset plots show the temperature dependent on the MR. Two features can be identified from these data. The first one is the monotonic increase of the MR upon cooling. It is  $\sim 30\%$  at 350 K and  $\sim 90\%$  at 50 K for LCMO/LMO/STON and  $\sim 24\%$  at 350 K and  $\sim 52\%$  at 50 K for LSMO/LMO/STON. It is interesting to note that the expected temperatures for zero MR are  $\sim 480$  K and  $\sim 520$  K for the LCMO/LMO/STON and LSMO/LMO/STON, respectively. Both temperatures significantly exceed the Curie temperature of LCMO or LSMO. This means that the interfacial state of the junctions is greatly different from that of the bulk

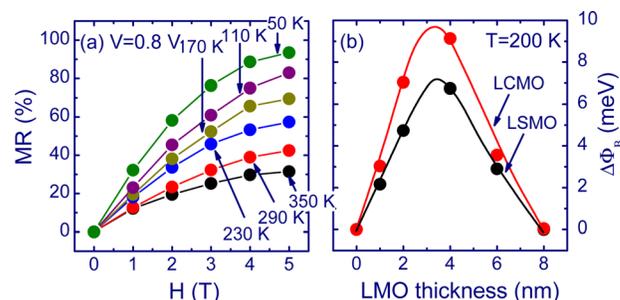


FIG. 3. (Color online) (a) MR as a function of magnetic field ( $H$ ) for the LCMO/LMO(4 nm)/STON junction at 50 K, 110 K, 170 K, 230 K, 290 K, and 350 K, respectively. (b) Magnetic field-caused change in interfacial barrier as a function of LMO thickness for LAMO/LMO/STON junctions at 200 K. Solid lines are guides for the eye.

manganites. The second feature is the difference of MR effect in the two series of junctions: The magnetic field effect is much stronger in LCMO/LMO/STON than in LSMO/LMO/STON. This could be ascribed to the different magnetic field responses of the LCMO and LSMO films. As well known, the MR is much stronger in the former, due to its strong Jahn-Teller effect, than in the latter. This result indicates that the interface layer of LAMO inherits the characteristics of the bulk counterpart.

Figures 2(c) and 2(d) show the dependence of the MR on the LMO thickness collected at 50 K, 200 K, and 350 K, respectively. Without the LMO layer, the MR is very low, and it increases as  $t$  grows from 1 to 4 nm, maximizing at the LMO layer of 4 nm. The MR disappears in the junctions with the LMO layer above 8 nm. It means that the coupling between the LAMO film and STON substrate can be separated completely by a layer of 8 nm.

To get further information on the magnetic field effect, we show the detailed dependence of the MR on magnetic field for LCMO/LMO(4 nm)/STON in Fig. 3(a). The MR is found to vary regularly with applied field, growing first rapidly then slowly with the increase of magnetic field. A tendency to saturation emerges closing the field of 5 T.

As is well known, there is a simple relation between saturation current and interfacial barrier,  $J_S \propto T^2 \exp(-q\Phi_B/k_B T)$ ,<sup>10</sup> where  $q$  is the electron charge,  $\Phi_B$  the interfacial barrier, and  $k_B$  the Boltzmann constant. According to Suzuki et al.,<sup>11</sup>  $(1-1/n)V$  is dropped across the insulating layer and  $V/n$  is applied to the depletion layer in the presence of the LMO layer. This suggests that LAMO/LMO/STON can be treated as Schottky junctions obeying the law of  $J \propto J_S [\exp(eV/nk_B T) - 1]$ <sup>12</sup> after considering the effect of the LMO layer by ideality factor. Figure 3(b) shows the variation of the interfacial barrier for the two series of junctions. It indicates a growth of  $\Phi_B$  in magnetic field, and this is the origin for the positive MR. Comparing the data in Figs. 2(c) and 2(d), we can clearly see the correspondence between MR and  $\Delta\Phi_B$ . The maximal  $\Delta\Phi_B$  is quite small,  $\sim 9$  meV, while the MR is significant,  $\sim 60\%$  at  $T = 200$  K. This is due to the exponential dependence of the junction resistance on interfacial barrier.

As is well established, the charge carriers near the interface are localized by lattice distortions. The presence of the LMO layer may awaken the dead layer by restoring the ferromagnetism of the interface. In this case, it may be easier for the magnetic field to affect the interface state, causing a change in Fermi level, thus in  $\Phi_B$ . The detailed relation between the positive MR and the awakening of the dead layer is an interesting issue and further studies in this regard are required.

#### IV. CONCLUSION

In conclusion, LMO buffer layer-induced positive MR in LAMO/LMO( $t$ )/STON ( $A = \text{Ca, Sr}$ ) have been experimentally studied. In addition to an enhanced magnetic response, the  $J$ - $V$  characteristics show a downward shift in magnetic field, indicating the positive MR. This is a phenomenon completely different from that observed in the junctions without LMO buffer layer. The positive enhanced MR is further found to show a systematic variation with the thickness of the LMO layer. It is weak when  $t$  is small, maximizes at  $t \approx 4$  nm, and disappears when  $t$  exceeds 8 nm. Furthermore, it is significantly stronger in LCMO/LMO/STON than in LSMO/LMO/STON. The maximal MR at 50 K is  $\sim 90\%$  for LCMO/LMO/STON and  $\sim 52\%$  for LSMO/LMO/STON, when LMO thickness is about 4 nm at 5 T. It is interesting that the MR persists up to 350 K, and it is  $\sim 30\%$  and  $\sim 24\%$  for the LCMO and LSMO junctions, respectively. The MR is further found to vary regularly with the applied field, growing first rapidly then slowly with the increase of magnetic field, a tendency to saturation emerges closing the field of 5 T. An analysis of the  $J$ - $V$  curves indicates an increase in interfacial barrier in magnetic field, which is the origin for the positive MR.

#### ACKNOWLEDGMENTS

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