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Buffer-layer-enhanced magnetic field effect in $La_{0.5}Ca_{0.5}MnO_3/LaMnO_3/SrTiO_3$:Nb heterojunctions

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The transport behaviors of $La_{0.5}Ca_{0.5}MnO_3/LaMnO_3/SrTiO_3$:Nb heterojunctions with a $LaMnO_3$ layer thickness of between 0 and 12 nm have been systematically studied. The effect of the magnetic field on the junction without the buffer layer is weak. The influence of the magnetic field on the junction is maximized when the layer thickness is ∼3 nm, demonstrated by a significant field-induced increase in current when the bias voltage is fixed. The corresponding magnetoresistance of the junction is negative, and its maximal value is ∼−32% for a field change of 5 T at 80 K. Based on a quantitative analysis of the current-voltage characteristics, the interfacial barrier can be derived, and it shows a complex variation with an increase in layer thickness, first decreasing and then increasing. This is the apparent reason for the change in buffer-layer-enhanced negative magnetoresistance of the junctions for different layer thicknesses. *© 2011 American Institute of Physics*. [doi:10.1063/1.3562916]

I. INTRODUCTION

Manganite-based heterojunctions have received great attention due to their excellent magnetic-field-dependent rectifying characteristics and bias-dependent magnetoresistance (MR).[1,2] In manganites, the strong magnetic-conductive correlation[3] and the order–disorder transition of the spin, orbital, and charge degrees of freedom[4] have been severely depressed in junctions by the strong interface/surface effect. In order to incorporate the peculiar properties of manganite into corresponding devices, the undesired interfacial effect should be depressed. As has been well established, the interfacial state is extraordinarily important in the study of heterojunctions. Recently, enhancement of the interfacial ferromagnetism of $La_{0.6}Sr_{0.4}MnO_3$ film by adding a $LaMnO_3$ buffer layer has been explored by Yamada *et al*.[5] This indicates that a proper buffer layer may affect the interfacial state and, thus, the physical properties of the junction. In fact, a field-induced positive MR has already been observed by Lü *et al*.[6] in $La_{0.67}Ca_{0.33}MnO_3/LaMnO_3$ (LMO)/$SrTiO_3$:Nb (STON) junctions, in addition to an enhanced magnetic response of the current-voltage characteristics. This is a completely different effect comparing with the negative MR usually observed in the junction without a buffer layer.

As a typical phase separated manganite that has coexisting ferromagnetic and charge ordered antiferromagnetic phases, the half-doped manganite $La_{0.5}Ca_{0.5}MnO_3$ (LCMO) possesses various properties that are different from those of $La_{0.67}Ca_{0.33}MnO_3$.[7] Its magnetic state experiences complex variations as temperature decreases, transitioning first from the paramagnetic (PM) to the ferromagnetic (FM) state, and then from the FM to the antiferromagnetic (AFM) state. It is possible that the interfacial state of the LCMO-based junctions could be different from that of $La_{0.67}Ca_{0.33}MnO_3/SrTiO_3$:Nb. As observed, the LMO layer has a strong effect on $La_{0.67}Ca_{0.33}MnO_3/SrTiO_3$:Nb. It would be interesting to check what happens to the LCMO/LMO/STON junction. Based on these considerations, in this paper, we perform a systematical study of the magnetic field effect of the LCMO/LMO/STON heterojunctions with different LMO thicknesses.

II. EXPERIMENT

The LCMO/LMO/STON junctions were fabricated by growing, by the pulsed laser ablation technique, first a LMO layer with a thickness t between 0 and 12 nm, and then a LCMO layer of ∼150 nm on the (001) $SrTiO_3$ substrate doped by 0.05 wt.% Nb. During the deposition, the temperature of the substrate was kept at 720 °C, and the oxygen pressure was kept at ∼10 Pa (for the LMO film) or ∼80 Pa (for the LCMO film). The film thickness was controlled by the deposition time.

The x-ray diffraction (XRD) results of the junctions indicate that the LCMO films are single phase and highly textured (not shown). The atomic force microscope analysis shows that the LMO buffer layer is quite smooth with a terrace-structured surface. For example, the root-mean-square roughness is ∼0.13 nm for the film of $t = 6$ nm, and the peak-to-valley fluctuation is ∼0.4 nm, which guarantees the quality of the top LCMO film.

Two Cu pads were deposited on the LCMO films and the STON substrate as electrodes, and the junction area was 1×1 mm^2. The Cu–STON and Cu–LCMO contacts were ohmic, and the contact resistances at room temperature were ∼15 Ω and ∼200 Ω. The *I-V* curves were measured by a superconducting quantum interference device magnetometer equipped with a resistance measurement unit. The voltage directing from LCMO to STON was defined as positive.

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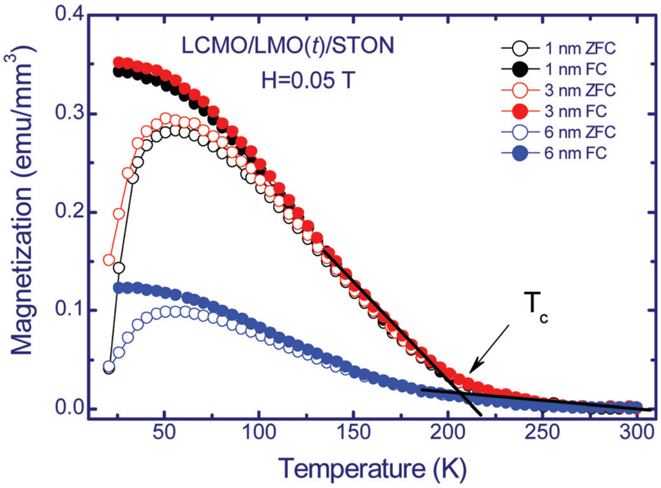

FIG. 1. (Color online) Temperature dependent magnetizations of the LCMO/LMO(t)/STON junctions for $t = 1$, 3, and 6 nm, respectively measured in the field cooled (FC) and zero-field cooled (ZFC) modes under an applied field of 0.05 T. Straight lines in the figure denote the Curie temperature.

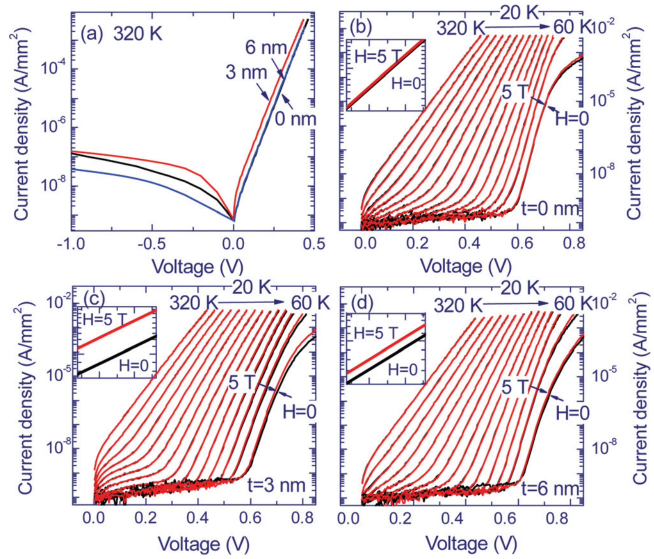

FIG. 2. (Color online) Semilogarithmic plot of the current-voltage curves measured without magnetic field for the LCMO/LMO(t)/STON junctions with typical LMO thicknesses of 0, 3, and 6 nm at 320 K (a). Current-voltage curves measured with and without magnetic field at different temperatures for the junctions with $t = 0$ (b), 3 (c), and 6 nm (d). Inset plots are close views of the *I-V* curves at 80 K.

III. RESULTS AND DISCUSSION

Figure 1 shows the thermomagnetization curves of the LCMO/LMO/STON junctions with buffer layer of 1, 3, and 6 nm, measured under a field of 0.05 T in the zero-field cooled (ZFC) and field cooled (FC) modes, respectively. The splitting of the FC and the ZFC curves at low temperatures reveals the presence of disorders in the films. The Curie temperature (T_c) of LCMO/LMO/STON shifts slightly to low temperatures with increasing LMO thickness. The ferromagnetic behavior of the film exhibits a complex variation, slightly enhancing when the LMO thickness increases from 1 to 3 nm and decreasing greatly when t changes from 3 to 6 nm. The latter suggests the depression of the FM order in the film. The slightly enhanced magnetization for $t = 3$ nm may be an indication of an increase of the FM region at the interface of the junctions. An ultrathin LMO layer may be hole-doped by LCMO due to the proximity effect and form a special interface phase. However, when the buffer layer is thick, the proximity effect is depressed. As a consequence, the magnetization is weakened when $t \geq 3$ nm again.

Figure 2(a) shows the semilogarithmic *I-V* characteristics of LCMO/LMO(t)/STON for $t = 0$, 3, and 6 nm, measured without magnetic field at 320 K. The asymmetric *I-V* curves show the excellent rectifying characters of the junctions. In general, the incorporation of the LMO layer does not affect the rectifying character of the junctions. However, an obvious leakage current appears when the LMO layer is 12 nm, which affects the transport behavior of the junction severely (not shown). However, the presence of the LMO layer enhances the magnetic field effect of the junction significantly. As shown by Figs. 2(b), 2(c), and 2(d), the log *I-V* curves exhibit an obvious upward shift upon the application of a magnetic field, indicating the negative MR effect. This effect enhances as t increases from 0 to 3 nm, and it weakens as t changes from 6 to 12 nm. The linearity of the log *I-V* curves implies that the electronic processes in the junctions remain unchanged in the presence of a magnetic field.

The MR of LCMO/LMO(t)/STON, defined as $J(0)/J(5\mathrm{T})$-1, is low when t is small. For the LCMO/STON junction of $t = 0$, the MR is about -1% at 80 K, even for positive MR at high temperature. Figure 3(a) shows the MR as a function of the bias voltage for the LCMO/LMO (3 nm)/STON junction. Figure 3(b) shows the MR as a function of LMO thickness. The MR grows rapidly with the increase of the LMO thickness, reaching, for example, a maximum value of $\sim -32\%$ at T = 80 K and $t = 3$ nm, and it decreases when the layer thickness exceeds 3 nm. The effects of temperature and electric bias on MR are also strong. In general, the MR increases with a decrease in temperature and reaches a maximum at an intermediate bias voltage.

In contrast to the tunneling process, the thermal process is generally insensitive to magnetic field. As a consequence, the MR is quite low for simple manganite junctions.[8,9] The enhanced MR observed in the LCMO/LMO/STON junctions is probably due to a change in hole density in the LMO/STON interface.

To get additional information on the interfacial state, in Fig. 4(a) we present the saturation current J_s as a function of temperature. J_s grows with temperature exponentially above 150 K. This result is also a signature of the thermal emission character of the transport process. As has been well established, the saturation current and interfacial barrier have a relation of $J_s \propto T^2\exp(-\Phi_B/k_BT)$,[10] where Φ_B is the interfacial barrier height and k_B is the Boltzmann constant. According to Suzuki *et al.*,[11] after introducing a LMO layer, $(1 - 1/n)V$ is dropped across the insulating layer and V/n is applied to the depletion layer. This suggests that LCMO/LMO(t)/STON can be treated as a Schottky junction and the *I-V* curves can be described by $J_s \propto T^2\exp(\Phi_B/k_BT)[\exp(eV/nk_BT) - 1]$ (Ref. 12) after considering the effect of the LMO layer by ideality factor. As shown in the inset of Fig. 4(a), the interfacial barrier experiences a small, yet definite,

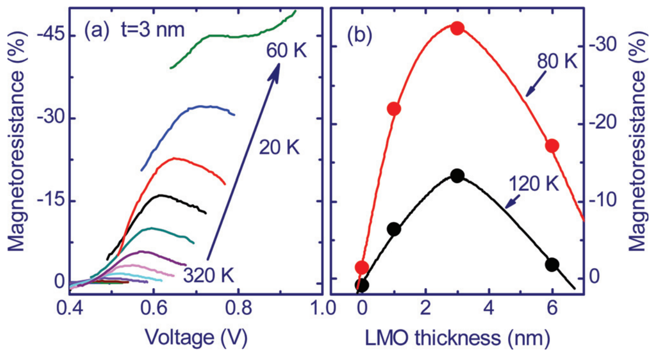

FIG. 3. (Color online) (a) MR as a function of bias voltage for the junction with $t = 3$ nm in the temperature range of 60 to 320 K. (b) MR as a function of the LMO layer thickness under a bias voltage of 0.7 V at temperatures of 80 and 120 K.

decrease with an increase in the LMO layer thickness, and it then increases when $t \geq 3$ nm again. The minimal Φ_B is $\sim$0.6 meV for $t = 3$ nm. These results indicate that the interfacial barrier shows a complex variation with increasing layer thickness, first decreasing and then increasing, which is the apparent reason for the change in the buffer-layer-enhanced negative MR of the junctions with different layer thicknesses in the present samples.

Figure 4(b) shows the ideality factor (n) of the junctions with $t = 1$ and 3 nm. In addition to the high quality of the junctions, the small ideality factor indicates that the dominant electronic process in the junction is the thermionic emission process. It is also obvious that the ideality factor experiences a slight, yet definite, increase with the incorporation of the LMO layer. This is plausible considering that part of the applied voltage drops on the LMO. It is interesting

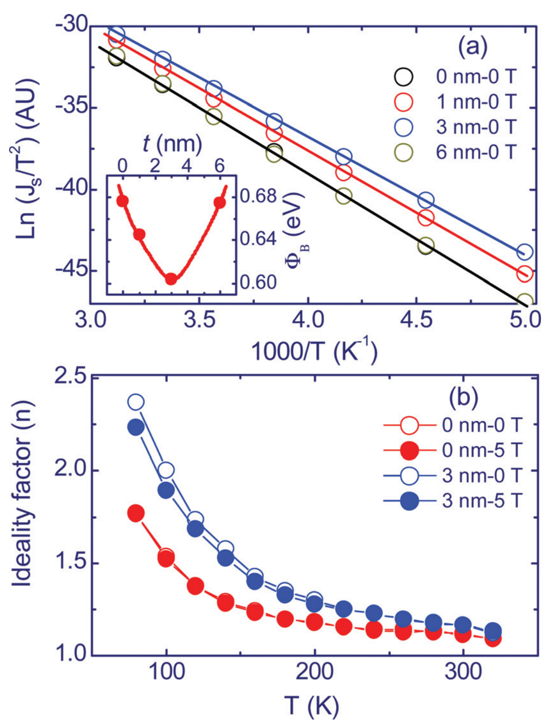

FIG. 4. (Color online) (a) Saturation current as a function of reciprocal temperature. (b) Ideality factor (n) as a function of temperature for LCMO/LMO(t)/STON. The inset plot in (a) shows the interfacial potential as a function of the LMO layer thickness without magnetic field.

that the applied field depresses the ideality factor, as shown in Fig. 4(b). This could be an indication of the improvement of the conductivity of the LMO layer.

IV. CONCLUSION

The enhanced MR is due to the LMO/STON interface. This effect is especially strong when t is small. However, when the buffer layer is thick, the proximity effect is depressed. In this case, the *I-V* characteristics of the junction are mainly determined by the LMO/STON interface. As a result, the MR effect is weakened because of the weak response of the LMO/STON interface to the magnetic field.

Magnetic field can improve the FM order of the manganite film, enhancing the mobility of the charge carriers. As a consequence, the density of the charge carriers that participate in the buildup of the interfacial barrier grow. This will cause a variation in depletion width. According to Xie *et al.*,[13] the depletion width undergoes a complex variation against the hole content. With the release of the localized holes, the depletion width can be either increased or decreased, depending strongly on the initial content of the mobile holes. It is possible that the enhancement of the MR in LCMO/LMO/STON is a combined result of the decrease of Φ_B and the depletion width. Different from $La_{0.67}Ca_{0.33}MnO_3/LaMnO_3/SrTiO_3$: Nb, the MR effect in LCMO/LMO/STON is negative. Obviously, the hole content near the LCMO/LMO interface could be different from that of the $La_{0.67}Ca_{0.33}MnO_3$/LMO interface, and it is probably higher. In this case, the different magnetic field effects in these two kinds of junctions are understandable.

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