Electronic transport and magnetoresistance in ultrathin manganite-titanate junctions

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We present a systematic study on the rectifying behaviors of the heterojunctions composed of a ultrathin $La_{0.67}Sr_{0.33}MnO_3$ film (~2 nm in thickness) and a $SrTiO_3$ substrate doped by 0.05 or 1 wt % Nb. These junctions exhibit excellent rectifying behaviors and a remarkable bias-dependent magnetoresistance (up to 60% under a field of 5 T). The transport behaviors are dominated by thermal process and tunneling process for the junctions with low and high Nb contents, respectively. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2828135]

Depositing manganite films on Nb-doped $SrTiO_3$ (a band semiconductor) substrates forms manganite-titanate junctions.[1–8] These junctions integrate the magnetoresistance (MR) of manganites and the rectifying behaviors of conventional semiconductor junctions and are potential spintronics devices. However, the understanding of the mechanisms of transport and MR of these junctions is limited because of the complexity of the manganites compared with the ordinary semiconductors.[9] It has been reported that there is an insulating layer at the interface of manganites and substrates, in which the transport and MR can be very different from that of bulk films.[10–12] It is a natural assumption that the physical properties of the junctions could be mainly determined by the layer near the interface rather than the bulk film. If treating manganite as an ordinary semiconductor, in most junctions the depletion region in manganite will be thinner than 1 unit cell. However, many authors suggested that, due to the electron localization by strong electron correlation, the depletion region of manganite should be much thicker.[1,13] Up to now, no experiments have been performed to show how the corresponding junction will behave when the thickness of the manganite film is reduced to several unit cells.

In this letter, we report on such a straightforward experiment on junctions based on ultrathin (2 nm, ~5 unit cells) $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) films. LSMO is the most canonical double-exchange manganite. Previous researches showed that the doping levels of substrates have significant influence on transport. For the lightly doped junctions thermal process is the most important transport mechanism,[5] while for the heavily doped junctions tunneling process is the most important.[8,13] To give an overall picture, junctions deposited on two types of substrates are compared—those deposited on 0.05 wt % Nb-doped (001) $SrTiO_3$ (0.05% STON) and on 1 wt % Nb-doped (001) $SrTiO_3$ (1% STON). These junctions exhibit excellent rectifying behaviors and a remarkable bias-dependent MR. The transport behaviors are dominated by thermal process and tunneling process for the junctions with low and high Nb contents, respectively. Remarkable MR (up to 60% under a field of 5 T) was observed in both kinds of junctions.

Pulsed laser ablation technique was used to deposit LSMO films on STON substrates. During the deposition the temperature was kept on 750 °C and the oxygen pressure was kept on 50 Pa. The details of the fabrication process of junctions have been described elsewhere.[8] The area of junction is 1 mm^2. Transport measurements were performed by standard two-electrode method. We first investigate the physical properties of single layer 2 nm LSMO film deposited on (001) $SrTiO_3$ substrate. X-ray diffraction reveals epitaxial growth of (001)-oriented LSMO film. Atomic force microscopy (AFM) analysis reveals a rather smooth film surface and the root-mean-square roughness is ~0.24 nm [Fig. 1(a)]. This result indicates a complete coverage of the substrate by thin film. Figure 1(b) shows the temperature-dependent resistivity and MR, under a field of 5 T, of the film. The resistivity exhibits an exponential increase with the decrease of temperature and is extremely large at low temperatures. The MR is enhanced at low temperatures. These are typical behaviors of thickness-reduced manganite films.

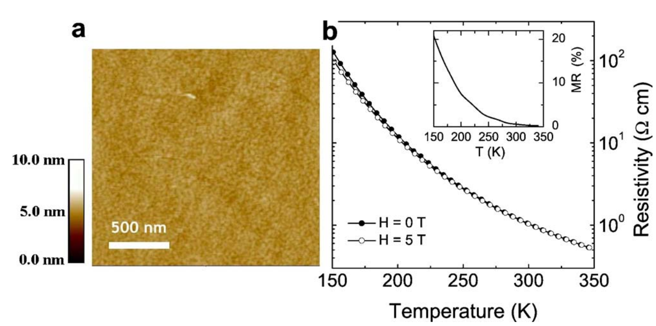

FIG. 1. (Color online) (a) AFM image of the 2 nm LSMO film. The root-mean-square roughness is ~0.24 nm. (b) In-plane resistivity ρ as a function of temperature T for the 2 nm LSMO film grown on (001) $SrTiO_3$ substrate. The inset shows the MR, defined as $[\rho(0\ \mathrm{T})-\rho(5\ \mathrm{T})]/\rho(0\ \mathrm{T})$, of the film.

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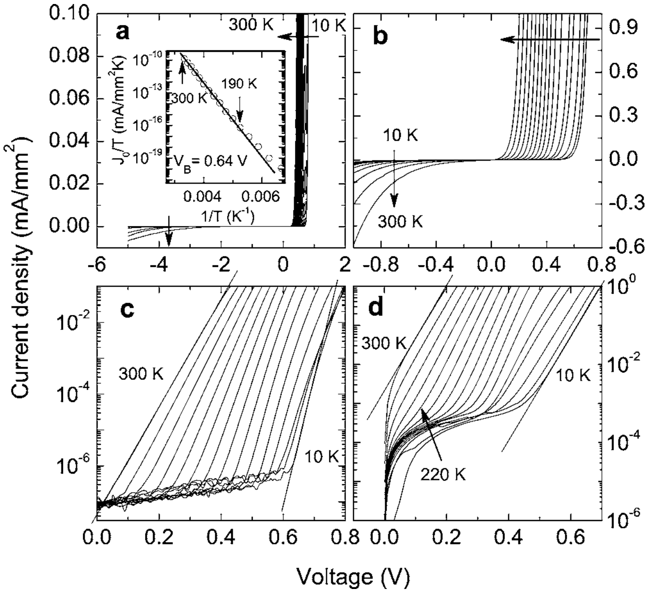

FIG. 2. *I-V* characteristics at different temperatures for junctions (a) LSMO/0.05% STON and (b) LSMO/1% STON. In (c) and (d), we redraw the results of (a) and (b) in semilog coordinates, respectively. The inset of (a) displays saturation current as a function of reciprocal temperature, based on which the built-in potential is determined. Thin lines are guide for eyes.

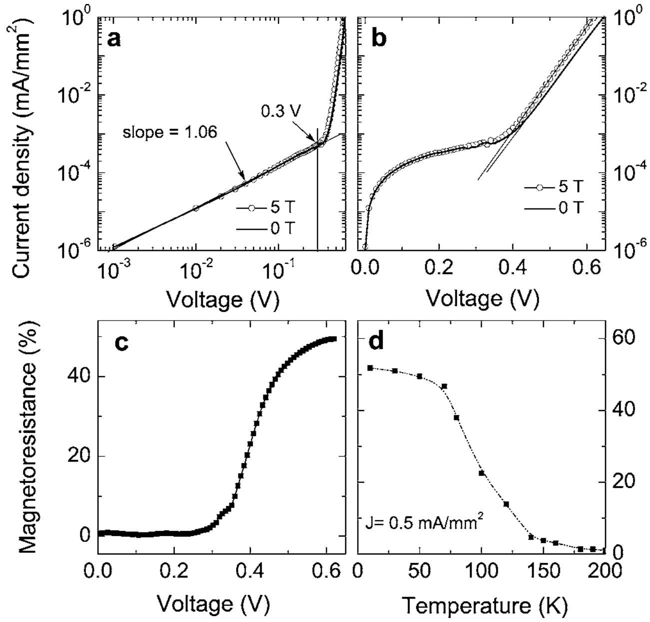

FIG. 3. (a) *I-V* characteristics of LSMO/1% STON with and without 5 T magnetic field at 50 K, drawn in double-log coordinates. (b) Redrawing (a) in semilog coordinates. (c) MR as a function of bias at 50 K. (d) MR as a function of temperature at $J=0.5$ mA/mm^2. Thin lines are guide for eyes.

Figure 2 presents the current-voltage (*I-V*) relations for the two junctions in the temperature range from 10 to 300 K. The junction LSMO/0.05% STON shows excellent rectification [see Fig. 2(a)]. The junction LSMO/1% STON also shows fairly good rectification, but its backward current density is much larger than the former [see Fig. 2(b)]. These features are similar to their thick counterparts.[4,5,7,8,13] In Figs. 2(c) and 2(d) are semilog plots of the *I-V* curves. For the junction LSMO/0.05% STON, log *I* and *V* are in good linear relation in wide temperature and bias ranges. The saturation of current below $\sim 10^{-7}$ mA/mm^2 is due to the limitation of amperemeter, which can only detect the current above 10^{-9} A. Here, thermal process is identified to be the dominant transport mechanism, just as in thick junctions.[5] For thermal process, to pass through the junction charge carriers have to surmount an energy barrier. Based on an analysis similar to that by Sawa *et al.*,[5] the barrier is estimated to be ~0.64 eV [inset of Fig. 2(a)], which is very close to that of the thick junction $La_{0.7}Sr_{0.3}MnO_3/SrTi_{0.9998}Nb_{0.0002}O_3$ (~0.65 eV).

The junction LSMO/1% STON demonstrates different log *I-V* relations. At low temperatures the log *I-V* curves split into two parts: at high bias range the curve is linear and at low bias range the curve is upwards bended—the current density is larger than that expected from linear relation. This detached trend becomes weak at high temperatures and disappears at ~220 K. Although the temperature varies from 10 to 220 K, the current density corresponding to the split is always around 10^{-3} mA/mm^2. The slope of the linear part only varies slightly from 10 to 300 K (the largest variation is ~40% for LSMO/1% STON, while ~160% for LSMO/0.05% STON), indicating that tunneling process is the main mechanism in this part.[8] To distinguish the mechanism of the bended low bias part, we draw the *I-V* curve in double-log coordinates [Fig. 3(a), 50 K]. It is interesting to note that in this case the low bias *I-V* relation is linear with a slope of ~1.06, strongly suggesting an Ohmic origin. This result indicates that the charge leaking dominants the electronic process in the low bias range. The leakage may be caused by interfacial defects, as demonstrated in the study of Wang *et al.*[14]

Actually, in a practical junction thermal current, tunneling current, and leakage current could coexist, though their contributions may vary with temperature and electric bias. For the junction LSMO/1% STON, the depletion region is thin due to the high carrier density in STON.[8,15,16] So it may be easier for the charge carriers to tunnel trough than surmount over the energy barrier. For tunneling, the forward current density J_f can be empirically expressed by Newman equation $J_f \propto \exp(\alpha T)\exp(\beta V)$ (α and β are constants), growing exponentially with temperature and bias (much faster than leakage).[8] Therefore, although the leaking current may generally exist, it only dominates in the low temperature and bias ranges.

Undoubtedly, the most attractive feature of the manganite junctions is the MR. In the following, we discuss the MR, $[R_{\mathrm{junction}}(0\ \mathrm{T})-R_{\mathrm{junction}}(5\ \mathrm{T})]/R_{\mathrm{junction}}(0\ \mathrm{T})$, of the junction LSMO/1% STON. In Fig. 3(b), we present the *I-V* curves at 50 K with and without magnetic field, respectively. It can be clearly seen that the magnetic field effect mainly occurs in the bias range dominated by tunneling process: magnetic field increases the slope of log *I-V* curve. The increase of the slope can be a consequence of magnetic field induced decrease of the maximum interfacial barrier.[17] The leaking current is almost not influenced by magnetic field. As seen in Fig. 3(c), below 0.3 V, the MR is near zero. Above 0.3 V, with the increase of bias, MR increases sharply and tends to saturation at ~0.6 V. The maximum MR is ~60%. In Fig. 3(d), we display the temperature-dependent MR at a constant current density 0.5 mA/mm^2. The shape of the MR-T curve is like a reverse "S." The MR decreases monotonously with temperature and vanishes at ~200 K.

Since STON is not sensitive with magnetic field, it is natural to ascribe MR to the LSMO layer. It has been sug-

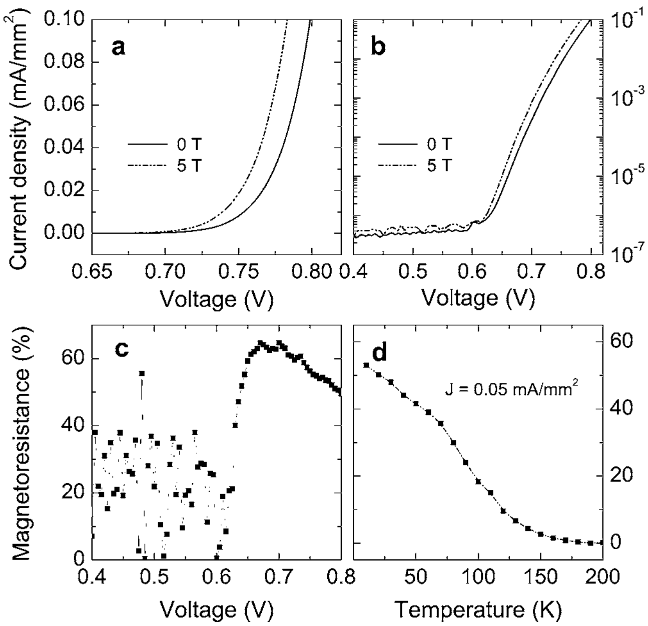

FIG. 4. *I-V* characteristics of LSMO/0.05% STON with and without 5 T magnetic field at 10 K. (b) Redrawing (a) in semilog coordinates. (c) MR as a function of bias at 10 K. (d) MR as a function of temperature at $J = 0.05\ \mathrm{mA/mm^2}$.

gested that in heavily doped junctions magnetic field depresses junction resistance by reducing depletion width, and thus increasing tunneling.[8,13] This case may still be true for the junction LSMO/1% STON. Applying magnetic field will have two consequences—releasing carriers localized in LSMO and decreasing maximum interfacial barrier—both of them leading to the reduction of depletion width. A question is that, in this junction, leakage contributes little to MR, while in a junction composed of a $La_{0.67}Ca_{0.33}MnO_3$ film and a 0.05% STON substrate, the leakage contributes the major MR.[14] Reason for this question is not clear yet. It is possible that the LSMO film is well grown while in the later the $La_{0.67}Ca_{0.33}MnO_3$ layer near the interface is marginally unstable, and there are plenty of defects and clusters whose spatial distribution can be influenced by magnetic field.

The junction LSMO/0.05% STON also has large magnetic field effect. Figure 4(a) shows the *I-V* curves at 10 K under 0 and 5 T. The field distinctly pushes the curve to low bias, which is similar to the results of the oxygen deficient junction $La_{0.7}Sr_{0.3}MnO_{3-\delta}/0.01$ wt % STON.[4] As stated above, when bias is small, the junction resistance is too large to be measured, and thus it is impossible to get reliable MR. The junction resistance decreases with increasing bias. The magnetic field effect is remarkable in all measurable bias range [>0.63 V, Fig. 4(b)]. The largest MR is $\sim 64\%$, appearing at ~ 0.67 V [Fig. 4(c)]. In Fig. 4(d), we display the temperature-dependent MR at a constant current density $0.05\ \mathrm{mA/mm^2}$. The MR decreases monotonously with temperature and vanishes at ~ 200 K.

The MR of the LSMO/0.05% STON could also originate from the field-induced change of tunneling current. Previous studies revealed that the thermal process is not sensitive to magnetic field.[4,14] Actually, the thermal process is strictly dominated only above 190 K, the signature of which is the deviation from linearity of the $\log J_0/T \sim 1/T$ curve below 190 K [inset of Fig. 2(a)].[5,16] As we know, tunneling is weakly temperature dependent while thermal process is strongly temperature dependent.[15] In the low temperature range, tunneling will have more contributions. Especially, at relatively high bias, the depletion region will be thin,[15,16] which significantly facilitates tunneling. That the MR mainly originates from tunneling process is supported by the following facts. First, the MR is distinct only at low temperature and high bias range in which tunneling process is believed to be important. Second, the maximum MR of this junction is close to that of the junction LSMO/1% STON and the MR of both junctions change with temperature in a similar manner. In addition, the magnetic field effect of this junction is similar to that of the oxygen-deficient junction,[4] in which magnetic field-induced reduction of depletion width has been experimentally observed. This speculation needs to be verified by further study.

In summary, we have experimentally studied the *I-V* characteristics and MR of 2 nm LSMO film based junctions, showing that these junctions capture main features of their thick counterparts and have remarkable MR. The main transport mechanism of junctions LSMO/0.05% STON and LSMO/1% STON are identified to be the thermal process and the tunneling process, respectively. The present study is of major interest for the further understanding of manganite based MR devices.

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