Vol 16 No 10, October 2007
1009-1963/2007/16(10)/3120-05
Chinese Physics

Rotation of ferromagnetic clusters induced magnetoresistance in the junction composed of $La_{0.9}Ca_{0.1}MnO_{3+\delta}$ and 1 wt.% Nb-doped $SrTiO_3^*$

Xie Yan-Wu(谢燕武)†, Wang Deng-Jing (王登京),
Shen Bao-Gen(沈保根), and Sun Ji-Rong(孙继荣)

Beijing National Laboratory for Condensed Matter Physics, Institute of Physics,
Chinese Academy of Sciences, Beijing 100080, China

(Received 13 March 2007; revised manuscript received 11 April 2007)

A junction composed of ultrathin $La_{0.9}Ca_{0.1}MnO_{3+\delta}$ (LCMO) film and 1 wt.% Nb-doped $SrTiO_3$ was fabricated and its magnetoresistance (MR) was studied and compared with LCMO film. It was found that the resistance of the junction has a similar dependence on magnetic field as that of the LCMO film: the curvature of R–H curves is upward above Curie temperature (T_C) and downward below T_C. These behaviours strongly suggest that the rotation of ferromagnetic clusters in manganite also causes MR in the corresponding junction. This MR can be qualitatively understood by the change of the width of the barrier induced by the rotation of ferromagnetic clusters. These results suggest a possibility to obtain junctions with large low-field MR.

Keywords: manganite, magnetoresistance, manganite junction, ferromagnetic clusters
PACC: 7530V, 7220M, 7475

1. Introduction

P-n junctions based on colossal magnetoresistive (CMR) manganites, $R_{1-x}A_x\mathrm{MnO}_3$ (R is a trivalent rare-earth ion and A is a divalent dopant), have attracted much attention because of their potential applications in spintronics and other devices.[1−5] A prominent feature of these junctions is the simultaneous control by an electrical bias and a magnetic field. The figure of merit of these junctions for many applications is the magnetoresistance (MR $= R_\mathrm{H}/R_0 - 1$), where R_H and R_0 are the resistances of the junction with and without a magnetic field, respectively. For good lattice match, the junctions are generally constructed by growing manganite films on Nb-doped $SrTiO_3$, which is insensitive to magnetic field. Therefore, the MR of the junctions is mainly determined by the corresponding manganites.

In the previous researches in studying the MR of the manganite junctions most efforts have been focused on the modulation of local spins of manganite. The MR is explained from a picture that a magnetic field can vary the height of the interfacial barrier potential by varying the band structure of the manganites[1−5] or another picture that a magnetic field may tune the width of the barrier by releasing carriers in the manganites. In generic CMR manganites, the field needed to exhibit significant modulation of local spins is typically on the order of several Tesla,[6] which is too large for practical applications. Actually, in ferromagnetic state manganite films usually contain ferromagnetic clusters. Besides the modulation of local spins, the rotation of ferromagnetic clusters can also contribute to the MR of manganites,[6] which needs much smaller field and thus seems to be more important for applications. However, up to now there is no report of whether the rotation of ferromagnetic clusters in manganite will or will not cause MR in the corresponding junction.

In this paper, we study by contrast the magnetic-field dependence of the resistances of a $La_{0.9}Ca_{0.1}MnO_{3+\delta}$ (LCMO) film and a junction based on LCMO film. Our results showed that the resistance of the junction has a similar dependence on magnetic field as that of the LCMO film, strongly suggesting that the rotation of ferromagnetic clusters in manganite also causes MR in the corresponding junction. This behaviour can be qualitatively understood by assuming the change of the width of the barrier.

*Project supported by the National Natural Science Foundation of China (Grant Nos 10474133 and 10674169).
†E-mail: xieyanwu@g203.iphy.ac.cn

2. Experiment

LCMO was chosen because of its small lattice mismatch with $SrTiO_3$ (STO).[7] The stoichiometric $La_{0.9}Ca_{0.1}MnO_3$ is an insulator.[6] To increase the conduction and MR of $La_{0.9}Ca_{0.1}MnO_3$, excess oxygen should be introduced. To directly compare the properties of the LCMO film and the junction, an ultra-thin LCMO film was used. The junction was fabricated by growing LCMO film of 10 nm in thickness on 1 wt.% Nb-doped (100) $SrTiO_3$ (STON) substrate by the pulsed laser ablation technique. During the deposition, the temperature was kept at 700 °C, and the oxygen pressure was kept at 100 Pa. The thickness was controlled by deposition time. To absorb oxygen, the junction was annealed in air at 550 °C for 10 min. For comparison, we also prepared LCMO films on (100) STO and (100) $LaAlO_3$ (LAO) substrates under the same conditions.

A Rigaku x-ray diffractometer with a rotating anode and Cu Kα radiation was used for the characterization of the LCMO film. A SQUID magnetometer (MPMS-7) equipped with electrical measurement system was used to measure transport properties. The in-plane resistance of the LCMO film was measured by the standard four-probe technique. Size of the junction area is $1 \times 1\,mm^2$, fabricated by the conventional photolithography and chemical etching technique. The two-probe technique was used for the measurements of the current-voltage characteristics to avoid the effects of current distribution in the junction.[8] The contact resistance is smaller than 10 Ω and 50 Ω between Ag and STON and between Ag and manganite films through the whole temperature range (10–350 K), respectively, evaluated by comparing the results of four- and two-probe measurements.[9] It is small compared with the resistance of the junction, which varies from $\sim 10^8\,\Omega$ to $\sim 10^3\,\Omega$ for low to high bias, and therefore, would not affect the quantitative analysis of MR. In the MR measurements of both the film and the junction, magnetic field was applied along the film (in-plane direction).

3. Results and discussion

3.1. The LCMO film

Figure 1 shows the results of x-ray diffraction of LCMO films. For LCMO film grown on STO, only (l00) peaks of STO were observed. It is because the pseudo-cubic lattice constant of LCMO ($\sim$ 0.389 nm) is very close to that of STO ($\sim$ 0.3905 nm).[7] Thus the peaks of LCMO were overlapped with the peaks of STO. For LCMO film grown on LAO, besides the (l00) peaks of LAO, the (l00) peaks of LCMO appear, and no other peaks are visible. These results confirm that LCMO has small lattice mismatch with STO, and demonstrate that the LCMO films are of single phase and highly textured with the c-axis perpendicular to the film plane.

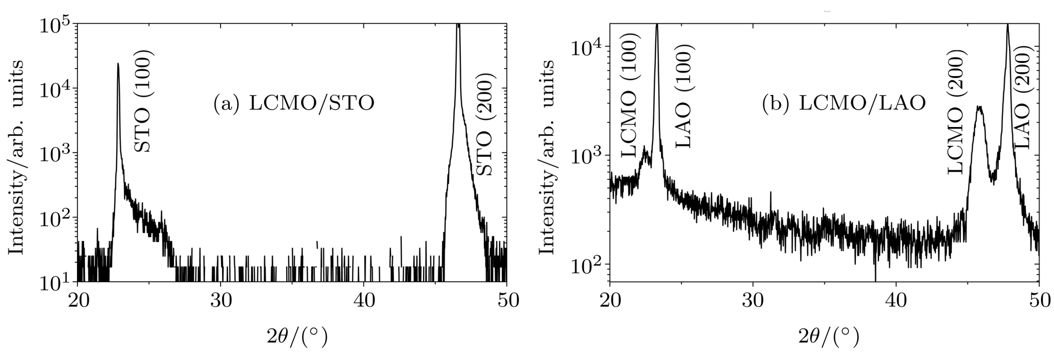

Fig.1. Typical x-ray diffraction pattern for LCMO films grown on (a) (100) $SrTiO_3$ and (b) (100) $LaAlO_3$ substrates.

Figure 2 shows the in-plane resistivity of the LCMO film (10 nm thick, grown on STO) measured under the fields of $H = 0$ and 5 T. Results for a thicker film (100 nm) are also presented for comparison. The LCMO film (100 nm) exhibits metal-to-insulator transition (MIT) at $\sim$ 220 K and a field of 5 T pushes the transition to $\sim$ 260 K, accompanied by the decrease of the resistivity. These characteristics are consistent with previous report[10] and are typical for CMR manganites. The MIT is associated with the ferromagnetic ordering at Curie temperature T_C. An insulating behaviour is observed in the whole temperature range studied for the LCMO (10 nm) film, and the resistivity is much larger than that of the thicker film. The maximum MR is $\sim -56\%$, occurring at ~ 140 K. The degeneration of the conductive property of the thin film has been ascribed to thickness effects.[11,12]

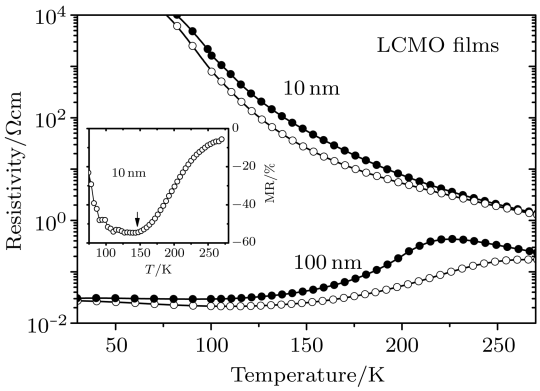

Fig.2. Temperature dependence of in-plane resistivity of LCMO films under 0 (dots) and 5 T (circles) magnetic fields. The inset is the MR (ρ(5T)/ρ(0T)-1) of the LCMO (10 nm) film.

As is known, the field dependence of resistance of manganite film is generally not linear.[6] Above T_C, the resistance decreases slowly at low field and relatively quickly at high field, and thus the curvature of R-H curve is upward. In contrast, below T_C the resistance decreases quickly at low field and relatively slowly at high field, and thus the curvature of R-H curve is downward. These field dependences can be simply explained as the following. A magnetic field can either rotate the ferromagnetic clusters (low field) or align the local spins (high field). The former depresses resistivity by reducing the magnetic scattering while the latter improves the double exchange transport. The rotation of ferromagnetic clusters is only important in the low-temperature ferromagnetic phase and leads to a rapid decrease of resistivity. In Fig.3 we present the field dependence of the normalized resistance of LCMO (10 nm) at various temperatures. It is interesting that similar field dependences as the typical manganites were found, though the LCMO (10 nm) film shows no MIT. The curvature of R-H curve is downward below 140 K and upward above 140 K. The critical temperature is well consistent with the temperature where the maximum MR occurs (Fig.2), indicating their coherent origin. These behaviours are not strange, and can be well understood in a phase separation scenario. Spatial inhomogeneity is even confirmed in $La_{0.7}Ca_{0.3}MnO_3$,[13] at such a doping concentration, a homogeneous low-temperature state might be expected. For LCMO (10 nm) film, although it is insulating, there are still lots of ferromagnetic phase, probably in the form of clusters, existing in the insulating matrix. Below T_C local spins in a cluster is aligned orderly. The critical temperature probably corresponds to the T_C of these clusters.

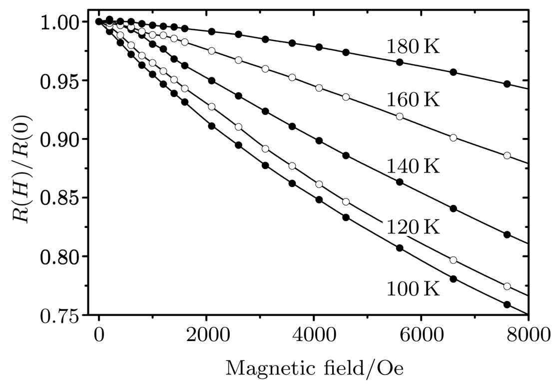

Fig.3. Magnetic field dependence of the normalized resistance $[R(H)/R(0)]$ of the LCMO film at different temperatures.

3.2. The LCMO/STON junction

Figure 4 presents the current-voltage (I-V) relations of the junction measured by tuning bias voltage. Fairly good rectifying behaviours characterized by strongly asymmetric I-V curves are observed. The current remains small up to the reverse bias of 0.6 V, but grows rapidly with voltage in the forward direction. Decrease in temperature has no obvious effects except for an expansion of the I–V curve along the V-axis. These behaviours are typical for the manganite junctions though the LCMO film here is only 10 nm in thickness.

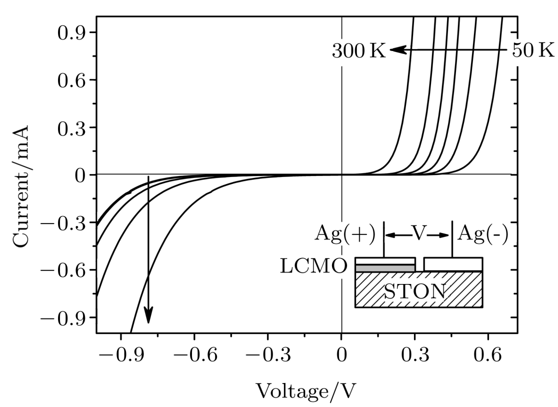

Fig.4. Selected current-voltage curves of the LCMO/STON junction. The interval is 50 K. The inset is the schematic view of the junction.

The junction shows prominent MR. In Fig.5 we present the temperature-dependent MR at two different biases. The MR changes with temperature in a similar trend to that of the LCMO film (inset, Fig.2). The maximum MR under 5 T field is $\sim -40\%$, occurring at ~ 120 K. This temperature is slightly lower than that of LCMO film (inset, Fig.2). Noting that T_C of LCMO decreases with thickness,[11,12] we consider it is reasonable. The MR of the junction is mainly determined by the LCMO layer near the interface, while the MR of the LCMO film is significantly influenced by the LCMO layer away from the interface. The similarity between the MR-T curves of the junction and the film confirms that the MR of the junction is induced by the MR of the film.

As shown in Fig.3, the rotation of ferromagnetic clusters contributes to the MR of the LCMO film, causing different field dependences of resistance above and below T_C. Therefore, it is expected that the rotation of ferromagnetic clusters will also bring about MR in the LCMO/STON junction. Accordingly, the field dependence of resistance of the junction should have different field dependences above and below 120 K. In Fig.6 we present the field dependence of normalized resistance ($R(H)/R(0)$) of the junction, at 100 K and 140 K. The curvature of R-H curve is downward at 100 K, and upward at 140 K, despite the bias. These results well consist with the expectation, strongly suggesting that the rotation of ferromagnetic clusters in LCMO also causes MR in the corresponding junction.

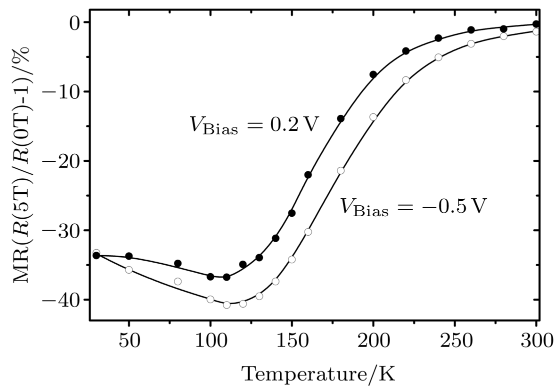

Fig.5. Temperature dependence of MR (R(5T)/R(0T)-1) of the LCMO/STON junction at two different biases.

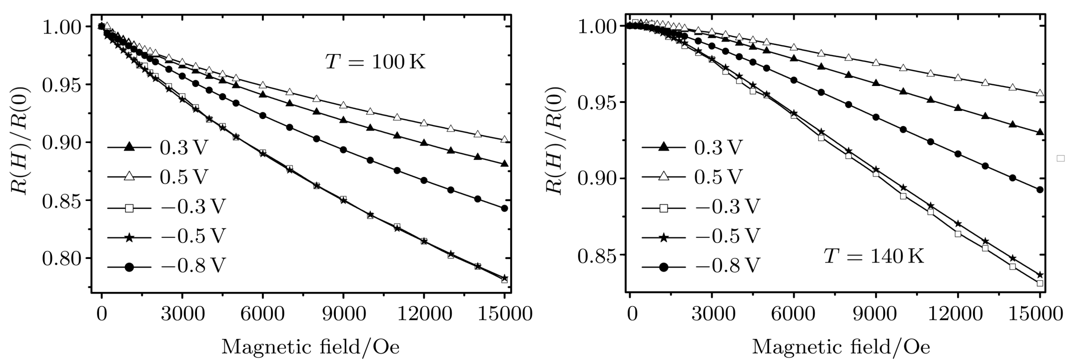

Fig.6. Magnetic field dependence of the normalized resistance ($R(H)/R(0)$) of the LCMO/STON junction at different biases. Upper panel: $T = 100$ K; lower panel: $T = 140$ K.

At 100 K, the MR under 1000 Oe (1 Oe $\approx$ 80 A/m) for LCMO and LCMO/STON are $\sim -4\%$ and $\sim -2.6\%$ (-0.5 V), respectively. These values seem not to be very attractive. However, it should be pointed out that in polycrystalline films, because of the grain boundary effect, the MR coming from the rotation of ferromagnetic clusters can be very large.[14,15] Accordingly, large low-field MR can be expected in the junctions composed of polycrystalline films and heavily doped STON.

Why the rotation of ferromagnetic clusters in manganite can produce MR in the junction? The band structure of manganite is determined by the double exchange, Jahn–Teller effect and other intrinsic effects.[6] The rotation of ferromagnetic clusters will not induce obvious change in the band structure. Therefore, the observed MR produced by the rotation of ferromagnetic clusters cannot be explained from the band view. In fact, tunnelling has been found to be the dominant mechanism for the LCMO/STON junction.[16] Because both LCMO and STON are heavily doped, the depletion layer of the junction is thin. A simple estimation based on conventional semiconductor theory showed that the depletion layer is in the order of several nm. It is easier for the charge carriers to tunnel through than to surmount the interfacial barrier. The rotation of ferromagnetic clusters can enhance the conduction of LCMO near the interface, which is equivalent to decrease the thickness of the barrier, and thus decrease the resistance of junction.

4. Summary

We have fabricated a junction composed of LCMO (10 nm) film and STON and studied its MR by comparison with LCMO film. We found that the resistance of the junction has a similar dependence on magnetic field as that of the LCMO film: the *R*-*H* curve is upward above T_C and downward below T_C. These behaviours strongly suggest that the rotation of ferromagnetic clusters in LCMO also causes MR in the corresponding junction. This MR can be qualitatively understood by the change of the width of the barrier induced by the rotation of ferromagnetic clusters. Moreover, considering that the rotation of ferromagnetic clusters induced MR can be very large for many mangnite films, our results suggest a possibility to obtain junctions with large low-field MR.

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