Effect of current processing on the transport property of the $La_{0.67}Ca_{0.33}MnO_3$ film

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Current effect on the transport property of the $La_{0.67}Ca_{0.33}MnO_3$ film has been studied as a function of temperature and magnetic field. The two-lead resistance of the sample shows a strongly asymmetric behavior with respect to current direction after a treatment under a current of the density of $\sim 1.6 \times 10^5$ A/cm^2, and the resistance is much lower in forward direction, the direction of the processing current, than in backward direction. A definite change in current–voltage slope at small but finite forward voltages is observed when alternating the electric field from backward to forward, which is indicative of the presence of two different resistive states corresponding to the two current directions and an energy barrier between these states. The magnetoresistance effect of the two resistive states is significantly different, and it is much stronger for the backward current than for the forward one. This has been proved a combined effect of the inclining of the current–voltage slope and the depression of intermediate energy barrier under magnetic field. The present work suggests a possibility to get a different magnetoresistance via interfacial engineering. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1944894]

Effect of electric field on the transport behavior of manganites has attracted wide attention in recent years due to its rich physics and great potential in developing electronic devices.[1–6] It was found that the resistance of manganites could experience a great reduction when electric field exceeded a critical value, and a significantly nonlinear current–voltage relation is resulted.[1,2] This effect is especially strong in phase-separated manganites, for which competitive and energetically degenerated electronic states coexist and small external stimulus can result in dramatic electroresistance (ER).[5] A generally accepted explanation to the ER effect is based on the assumption of the formation of filamentary conducting paths in manganites due to the melting of charge-ordered phase under electric field.[1,2,4]

A recent research revealed the presence of an interesting feature of the ER effect in manganite-based devices. It was found by Liu *et al.*[7] that the two-lead resistance of a $Pr_{0.7}Ca_{0.3}MnO_3$ layer sandwiched by a Ag top electrode and a $YBa_2Cu_3O_7$ bottom electrode can be modified drastically by electric pulses, and the resulting ER effect is strongly pulse direction dependent: The forward electric pulse depresses while the backward one enhances the resistance. Explanations to these observations are still in dispute. Liu and co-authors postulated the presence of conducting clusters in $Pr_{0.7}Ca_{0.3}MnO_3$ even at room temperature and the directional arrangement of these clusters under electric field.[7] On the contrary, Baikalov *et al.* believed that these behaviors cannot be simply ascribed to manganites alone, and proposed an electric pulse-induced oscillation of the contacting resistance between a low and a high values.[8,9]

We noted that most of the previous studies were performed at room temperature without magnetic field. Considering the fact that a distinctive property of the manganites is the strong magnetic-resistive interplay, in this letter we will present a systematic investigation on the combined effects of temperature, current and magnetic field for the typical manganite $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) to get a thorough understanding of the current effect.

A LCMO film of the thickness of $\sim$120 nm was grown on the (001) $SrTiO_3$ substrate by the pulsed laser ablation technique. The temperature of the substrate was kept at 720 °C and the oxygen pressure at $\sim$80 Pa during the deposition. X-ray diffraction analysis shows that the film is single phase and epitaxially grown. The film was patterned by the conventional lithographic method into a thin line of the width of 0.2 mm, and two rectangular silver pads were subsequently deposited on it to get a good electric contact. The Ag-LCMO interface is 0.2×2 mm^2 in size for each electrode.

Resistance of the sample was measured as a function of temperature, and a typical transport behavior of bulk LCMO was observed with the metal-to-insulator transition (MIT) occurring at $\sim$235 K. To perform current processing, the sample was first cooled down to 5 K without current, subsequently warmed up to room temperature with a constant current between 10 and 40 mA being applied. This process produced a strong effect on the resistive property of the sample particularly when the applied current exceeded 30 mA. In this letter, we will focus our attention on the sample treated by a current of 40 mA (current density $\sim 1.6 \times 10^5$ A/cm^2).

One of the most striking observations of the present work is the anisotropic resistive behavior produced by current processing.[10] The resistance of the sample to the electric current of different direction is different, and it is much lower when the current is applied in the same direction as the processing current (called hereafter as forward current). Figure 1 exemplifies the temperature dependence of the resistances recorded under the measuring current of −0.5 and 0.5 mA, respectively. It shows that the resistive discrepancy appears at $\sim$150 K and develops with the increase of temperature until T=235 K, where the maximum resistance difference occurs. The backward resistance can be $\sim$3 times as

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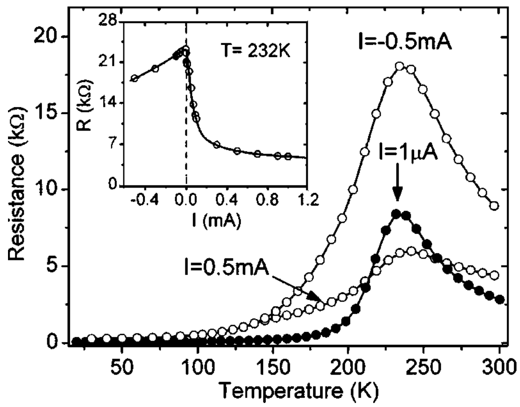

FIG. 1. Temperature-dependent resistance of current-processed LCMO film measured under the applied current of −0.5 and 0.5 mA. Results without current processing are also presented for comparison (black symbols). Inset is a plot showing the current dependence of the resistance at a fixed temperature 232 K.

large as the forward one. The resistive data collected at a fixed temperature but different currents clearly demonstrate the current effect. As shown by the inset of Fig. 1, the resistance undergoes a sharp drop with the increase of the forward current, whereas a rather slow decay with opposite current. The resistance is ~23 300 Ω for $I=0$ and ~3000 Ω for $I=5$ mA, giving rise to an ER $[1-\rho(0)/\rho(I)]$ of ~680%. Despite of the great resistance variance, fascinatingly, the MIT takes place at an essentially the same temperature for different currents.

To get a further knowledge about current effect, we measured the current–voltage (I–V) relation of the sample in the presence of a magnetic field between 0 and 4.5 T, and the typical results are presented in Fig. 2. As expected, the I–V curve shows a strong anisotropy with respect to the direction of current. The *I–V* dependence deviates significantly from a linear relation in the forward direction due to the rapid increase of current with voltage, while keeps practically linear in opposite direction. This feature is visible in all of the *I–V* curves obtained at different temperatures, either below or above the MIT of the LCMO film, and magnetic fields. Magnetic field affects significantly the I–V slope, leading to a colossal magnetoresistance (MR). A close view of Fig. 2 reveals the presence of two different *I–V* slopes separated by

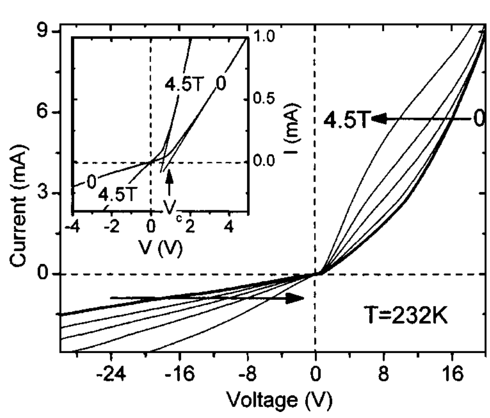

FIG. 2. Selected current–voltage characteristics of LCMO measured at 232 K under the magnetic fields of 0 and 4.5 T. Data for $H=0$ are plotted as a thick line for clarity. Inset plot demonstrates the presence of critical voltages for the current-driven resistive transition and the magnetically tenable feature of the *I–V* curves.

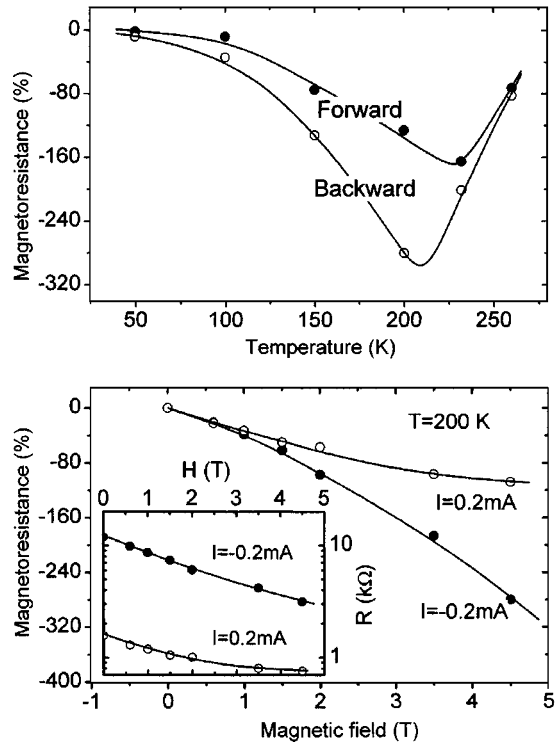

FIG. 3. Magnetoresistance as a function of temperature (top panel) and magnetic field (bottom panel). Inset plot in the bottom panel shows the resistance corresponding to $I=-0.2$ and 0.2 mA. Solid lines are guides for the eye.

a visible kink in the *I–V* curve, which is a sign of two resistive states (inset in Fig. 2). It is interesting that the magnetic field effect is different for these two states. As shown in Fig. 3, the MR, defined as the relative variation of the *I–V* slope under magnetic field $[1-\rho(0)/\rho(H)]$, is considerably larger for backward current than for forward current. Furthermore, the MR maximum occurs at two very different temperatures of ~210 and ~232 K in the two directions. The former is obviously lower than whereas the latter coincides pretty well with the MIT temperature of LCMO. This is a feature in sharp contrast to that of the ordinary manganites, for which the MR maximum locates near the MIT temperature, and indicates a completely different mechanism for the backward MR. The maximum MR difference appears at ~200 K and a simple calculation shows that the MR is ~−280% and ~−100% under a field of 4.5 T when alternating current direction. The MR difference reduces and, finally, vanishes as the temperature approaches 320 K, though the giant gap remains in the resistance of different direction (inset in Fig. 3).

There are two apparent possible consequences for current processing. The first one is the variation of the resistance of LCMO and the second one is the depravation of the Ag-LCMO contacting that contributes an ohmic resistance. It has been argued that filamentary conducting paths could be formed in the film under the impulsion of strong electric pulses. In that case, a resistance drop and a transport behavior independent of current-direction were resulted.[1,2] This is obviously not the only process in our sample. The contacting resistance may not be a determinative reason either for the increase of the backward resistance noting that the resistive enhancement was also observed even when four-point technique was used for the study of the transport property.[4] The strongly asymmetric resistive behavior of LCMO reminds us of *p*-*n* or Schottky junctions. A careful analysis shows that the change of the *I*–*V* slope takes place only above a critical voltage (V_c) (inset in Fig. 2). This suggests the presence of an energy barrier between the high and low resistive states. The great increase of the resistance of the Ag-LCMO system also proposes the occurrence of interfacial energy barriers. A simple calculation shows that the resistance peak at ~235 K, measured under the applied current of 1 μA, increases by ~2 times after current processing (~23 300 Ω vs ~8400 Ω as shown in Fig. 1). The resistance enhancement could not be ascribed to LCMO alone. The unchanged MIT temperature implies that the effect of current processing on LCMO may not be as strong as believed. In fact, it is possible for the Schottky barrier to be formed at the Ag-LCMO interface due to, for example, the electrochemical reaction resulted by current processing. The Schottky barriers could be different at the two Ag-LCMO interfaces because of the different directions of the electric field, which explains the asymmetry of the *I*–*V* relations. These analyses mean that the backward resistance could be jointly determined by the interfacial energy barriers and the LCMO film, and the former may be more important regarding the much large resistance compared with the original LCMO film (Fig. 1). The V_c–T relation is further studied, and it indeed reproduces the main feature of the *R*-*T* relation with the V_c maximum locating at ~232 K (top panel of Fig. 4). In contrast, the forward resistance measured under a large current is more likely an intrinsic property of current-processed LCMO than energy barrier, and the low resistance could be a sign of the occurrence of filamentary conduction paths in LCMO.

Based on these analyses, the anisotropic MR behavior of LCMO can also be understood. It is obvious that the depression of the resistance of LCMO implies a modification of magnetic field to interfacial energy barrier. Significant variations of V_c with magnetic field are indeed observed especially near the MIT temperature of LCMO. A simple estimate indicates that a field of 4.5 T can lower V_c from 0.75 V to 0.65 V for $T=150$ K and from 0.96 V to 0.74 V for $T=232$ K (bottom panel of Fig. 4). Therefore, the backward MR is a combined result of that of the Schottky barrier and the LCMO film, which explains why the MR maximum deviates from the MIT temperature of LCMO. These results suggest a possibility to get a completely different MR via interfacial engineering.

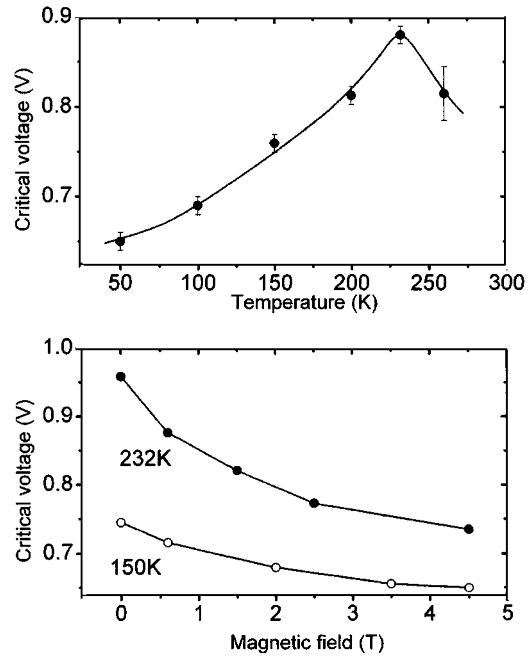

FIG. 4. Variation of the critical voltage for the current-driven resistive transition as functions of temperature (top panel) and magnetic field (bottom panel). Solid lines are guides for the eye.

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