Anisotropic conduction induced by current processing in the $La_{0.8}Ca_{0.2}MnO_3$ film

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Effects of current processing have been experimentally studied for the $La_{0.8}Ca_{0.2}MnO_3$ film. An anisotropic conduction is observed when the film is processed by a current of the density of $\sim 2.4\times 10^5$ A/cm^2. Difference between the resistances in the direction of the processing current and the reverse direction can be as high as 10 000%. Different from the original film, which exhibits a metal-to-insulator transition at ~214 K, the sample becomes insulating below 300 K under low voltage bias and undergoes a metallic transition at very different temperatures in the two directions when the voltage bias is high enough. These features remain after reordering the electrodes, which reveals the intrinsic origin of the anisotropy, instead of interfacial barrier due to current processing. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2369844]

A distinctive feature of hole-doped manganites is the strong magnetic-resistive interplay: The resistivity of the manganite undergoes a great reduction under magnetic field, which is called as colossal magnetoresistance.[1] In fact, in addition to magnetic field, electric field/current,[2–5] light illumination,[6] and high pressure[7] also have a strong impact on the transport property of the manganite. Compared with others, the most dramatic effects may be those produced by electric current. There are reports that the applied current can result in a great resistivity reduction in the charge/orbital-ordering manganites, which has been attributed to either the collapse of the charge/orbital ordering[2] or the effect of heat.[8,9] Strong current effects have also been observed in the manganites without significant charge ordering such as $La_{0.8}Ca_{0.2}MnO_3$ and $La_{0.67}Ca_{0.33}MnO_3$,[3–5] the latter is a sample with an optimal hole concentration for the colossal magnetoresistance effects. Taking $La_{0.67}Ca_{0.33}MnO_3$ as an example, its resistance decreases from ~4.1 kΩ to ~1.7 kΩ when the electric current increases from 0.001 to 6 mA, though the critical temperature for the resistive transition remains unaffected.[5] The underlying physics for this effect is still unclear. Coexistence of multiphases and the influence of applied current are believed to be possible origins of this effect.

In the work mentioned above, electrical current affected only the magnitude of the resistivity. Recently, Sun *et al.*[10] observed a completely different current effect characterized by a significantly anisotropic conduction. The authors first processed the $La_{0.67}Ca_{0.33}MnO_3$ film by applying a strong current, then measured its resistance using a low current. It is surprising that the resistance strongly depends on the direction of the measuring current. It is much smaller in the direction of the processing current than in the opposite direction. The maximum difference can be as high as ~400%. Similar phenomena were subsequently observed by Hu and Gao in a $La_{0.8}Ca_{0.2}MnO_3$ film.[11]

In the work of Sun *et al.* a two-probe configuration was utilized. In this case, current processing and resistance measurement used the same electrodes. The conventional four-probe technique was adopted by Hu and Gao. Although the outer two electrodes and the inner two electrodes were, respectively, used for sample processing and subsequent resistance measurement, the possibility that the processing current passed through the conductive inner electrodes, via the underlying manganite film, cannot be ruled out. Therefore, a possible explanation for this effect is that current processing causes the formation of an interfacial energy barrier between the metallic electrode and the oxide sample, and it is the interfacial potential that causes the anisotropic conduction. In fact, interfacial potential produced resistive switching was indeed observed before in many systems.[12,13] However, the work presented in this letter suggests an alternative possibility: Current processing can result in an intrinsic anisotropy of the manganite film.

A $La_{0.8}Ca_{0.2}MnO_3$ (LCMO) film was epitaxially grown on a (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 ~100 Pa during the deposition. The film thickness was ~100 nm, controlled by deposition time.

The film was patterned by the conventional lithography technique into a geometry of a 50×50 μm^2 microbridge clamped by two rectangular blocks of the size of ~1 $\times 2$ mm^2 for each. The latter are reserved for the preparation of electric contacts. Two electrodes for current processing were made by silver paste, covering the outer four-fifths of the LCMO block. Subsequent measurements reveal a typical resistive behavior of the hole-doped manganite, characterized by a metallic conduction at low temperatures and an insulating conduction at high temperatures (Fig. 1). A magnetic field of 5 T shifts the insulator-to-metal transition from $T_{MIT}\approx 214$ K to ~255 K, resulting in a huge magnetoresistance (~96%).

The sample was then mounted on an aluminum radiator using silver paste and directly dipped into liquid nitrogen. A constant current of 12 mA, supplied by a Keithley 2400 source meter, was applied. This process lasted for ~5 min. Current-voltage characteristics (*I-V*) of the sample were subsequently measured to check whether the expected effect ap-

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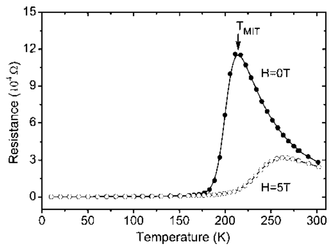

FIG. 1. Temperature dependence of resistance of the LCMO film measured under the fields of $H=0$ and 5 T. A metal-to-insulator transition occurs at $T_{MIT}\approx 214$ K.

pears. Instead of the linear increase of current with voltage observed before current processing, the *I-V* curve is strongly nonlinear and asymmetric against the polarity of the electric bias, like a horizontal line upwards bended at ~ 0.7 V [Fig. 2(a)]. This result, which is similar to that previously reported by Sun *et al.*[10] and Hu and Gao,[11] confirms the occurrence of dramatic current effect.

The silver paste was then washed away and another two rectangular Ag pads were deposited, by the laser ablation technique, on the whole LCMO blocks. The *I-V* relations of the sample were subsequently measured at selected temperatures between 10 and 300 K under the fields of $H=0$ and 5 T. It is interesting that the *I-V* relations remain nonlinear and anisotropic. This feature is especially strong in the low-temperature range. The current grows steeply with voltage in the direction of the processing current when the voltage bias exceeds ~ 1.2 V, while it keeps pretty small in the opposite direction up to the electric bias of ~ 10 V [Fig. 2(b)]. The resistance in the direction against processing current can be 100 times as large as that in the opposite direction at $T=10$ K. An increase in temperature leads to a gradual opening up of the bended curve; however, asymmetry remains significant even for $T=300$ K. Application of magnetic field causes a counterclockwise rotation of the *I-V* curve around its turning point in addition to slightly enhancing the electric anisotropy [Fig. 2(c)].

Compared with the results obtained before electrode rearrangement, the rigid turning in the *I-V* curve becomes considerably soft. This could be a consequence of the relaxation

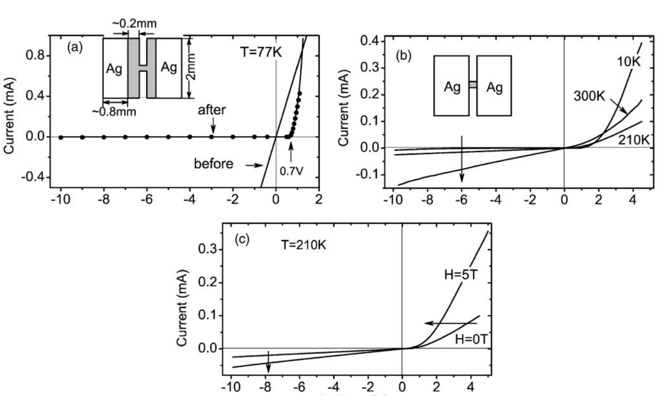

FIG. 2. (a) Current-voltage characteristics of the LCMO film measured before and after current processing with the electrode setting shown in the inset. The data measured immediately after the rearrangement of the electrodes are also shown for comparison (symbols). The positive direction is the direction of the processing current. (b) Current-voltage characteristics measured with the electrode setting shown in the inset. (c) Effect of magnetic field on the current-voltage relation at $T=210$ K, measured with the electrode setting of (b).

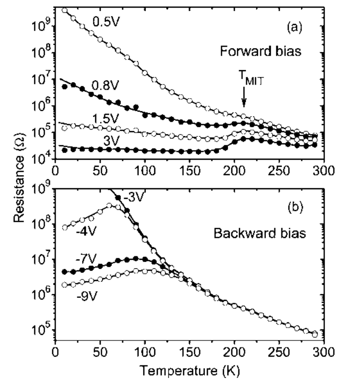

FIG. 3. Temperature dependence of the resistance of the current-processed LCMO film measured under different voltage biases.

of the current effect: The transport measurements were performed ~ 100 h later after current processing. In fact, an immediate measurement after rearranging electrodes gives an *I-V* curve coinciding exactly with the original one within experimental error [symbols in Fig. 2(a)].

These results actually imply a strong direction dependence of the electroresistance and magnetoresistance effects. This is confirmed by the data presented in Figs. 3 and 4. Figure 3 demonstrates the temperature-dependent resistance of the LCMO film recorded at selected voltage biases. Although both the forward and backward resistances are remarkably bias dependent, differences between them are obvious. The sample is completely insulating in the forward direction when the voltage bias is below ~ 0.5 V, with only a minor resistive anomaly at T_{MIT}, where the metal-to-insulator transition happens for the original LCMO film. An increase of voltage bias depresses the resistance by enhancing the metallic character of the conduction. A simple calculation indicates that the resistance at a fixed temperature $T=10$ K decreases from $\sim 5\times 10^9\ \Omega$ to $\sim 2\times 10^4\ \Omega$ as the voltage bias increases from 0.5 to 3 V, which implies an electroresistance of $\mathrm{ER}=1-R(3\ \mathrm{V})/R(0.5\ \mathrm{V})\approx 99.999\%$. It is interesting to note that although the resistance reduces greatly, the temperature for the resistive transition remains unaffected. This result reveals the different effects of magnetic field and current. The former depresses the resistance in the meantime shifting the resistive transition to high temperatures. In the reverse direction, in contrast, an insulator-to-metal transition takes place at a much low temperature when the voltage bias exceeds ~ 4 V. Different from the forward resistance, in this case the resistive transition is significantly bias dependent, growing from ~ 62 K for $|V|=4$ V to ~ 106 K for $|V|=10$ V. This feature is indicative of a substantially different mechanism for this transition.

Different responses of the forward and backward resistances to magnetic field are also observed. The forward resistance undergoes a great reduction under magnetic field, accompanying with a high-temperature shift of the resistive transition, a feature similar to that of the original LCMO. The maximum magnetoresistance is $\sim 66\%$ under the field of 5 T, occurring near T_{MIT}. On the contrary, magnetic field pushes the resistive transition to a low temperature when a reverse voltage bias is applied. As shown in Fig. 4(b), a field of 5 T drives the resistive transition from ~ 76 K to ~ 63 K under a bias voltage of $|V|=5$ V. This is a behavior significantly different from that of the ordinary manganite.

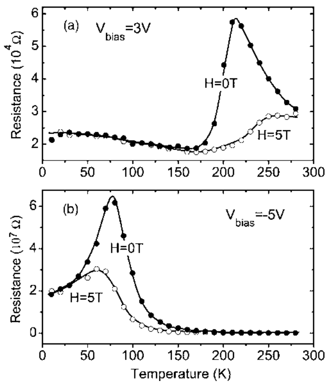

FIG. 4. Temperature dependence of resistance of the current-processed LCMO film measured under the fields of $H=0$ and 5 T.

The experiments were repeated several times for different samples with the same composition but different electrode settings, and essentially the same results are obtained. There are suggestions for the presence of a Schottky barrier at the interface of metallic electrode and insulating sample,[13,14] which is believed to be responsible for the direction-dependent transport behavior. Interfacial barrier at the metal-manganite interface could be negligibly small, as confirmed by the appearance of the linear *I-V* relation. However, there are signatures for the formation of interfacial barrier when a strong current passes through the metal-manganite interface. For example, for the $Ti/Pr_{0.7}Ca_{0.3}MnO_3$ system, a strongly asymmetric transport behavior emerges and develops after repeated *I-V* measurements.[13]

The rearranged electrode covers the whole LCMO block, including the unsullied part of the LCMO block against the two ends of the bridge. A direct calculation indicates that the density of the processing current is $\sim 2.4\times10^5$ A/cm^2 in the bridge while $\sim 6\times10^3$ A/cm^2 in the LCMO block. The latter is much smaller than the reported critical current for the occurrence of irreversible current effect ($\sim 1.2\times10^5$ A/cm^2).[10,11,15] This implies that, unlike the bridge, the LCMO block will return to its initial state after the removal of the processing current.[5] Therefore, the contact between Ag and the fresh LCMO should be an Ohmic contact with a low contacting resistance, as that observed before current processing. Furthermore, if an interfacial potential exists between the remaining LCMO block and the Ag pad, its effect will be shunted noting the fact that the interfacial barrier will exhibit a great contacting resistance, especially in the reverse direction (Fig. 2). These analyses suggest that the asymmetric transport behavior could be an intrinsic property of the current-processed LCMO film.

The depression of resistance by current has been observed before in different manganites.[2–5] An interesting question is how anisotropic conduction occurs. This effect, which cannot be understood based on the known theories on the manganite, reminds us of the Schottky junction or *p-n* junction. In fact, direction dependence of the transport behavior may be indicative of the formation of junctionlike structure in the film during current processing. A morphology analysis by atomic force microscope indicates the presence of fine grains in the LCMO film, and the lateral grain size is ~ 1 μm. It is obvious that the resistance across the boundary would be larger than that of the grain itself. With this in mind, inhomogeneous local heating due to the application of a strong current will occur. The strongest self-heating may take place near grain boundaries, on which lattice defects concentrated. In addition to this, a directional migration of oxygen across the grain boundaries is also possible due to, for example, an electrochemistric reaction. All these may result in junctionlike grain boundaries. In this picture, the transport behavior of the LCMO film will be determined by the conduction of the grain and the interfacial structure, and the strongly bias-dependent property may mainly originate from the latter. When the resistance of the grain boundaries is depressed by a high electric bias, the behavior of the LCMO grains emerges. This explains the appearance of a resistive anomaly around T_{MIT} with the increase of electric bias and the independence of T_{MIT} of electric bias in Fig. 3(a): Electric bias affects mainly the grain boundaries. This is obviously a primary explanation, and a further study is required to get a deep insight into the mechanism for the anisotropic conduction in the LCMO film.

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