# Switching behavior of $La_{1-x}Ca_xMnO_3$ thin films grown on $SrTiO_3$ substrates

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We report the switching behavior of resistive states induced by current flowing in $La_{1-x}Ca_xMnO_3$ films. The transport behavior upon treatments by applying a large dc current has been investigated in the absence of magnetic field. A switching from low to high resistive state was found upon applying a large dc current for the both compositions. The critical currents for the switching are 10.5 mA (density of $2.3\times10^3$ A/cm$^2$) and 6.7 mA (density of $1.6\times10^3$ A/cm$^2$) for compositions $x=0.2$ and 0.3, respectively. Our repeated measurements indicate that the high resistive state appeared in the film $x=0.2$ is much stable. Exposing the sample in air and room temperature does not cause any change of the transport properties. However, for the film with $x=0.3$, the induced high resistive state behaves with unstable characteristics. Keeping the sample at a low temperature of 20 K, a relaxation to a higher resistive state takes place. The mechanism of phase separation was taken into account in the interpretation of the observed phenomena. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2831327]

Colossal magnetoresistance (CMR) effect in mixed-valence-doped manganese perovskites has triggered intense scientific activity in recent years. Phase separation together with concomitant percolation behavior have been supposed to be the core of CMR effect.[1,2] An increasing interest has been recently attracted to the influence of electric field/current to the transport in CMR materials.[3–10] Significant change of electric resistance induced by static electric field has been demonstrated in $La_{1-x}Ca_xMnO_3$ using field effect configurations. The observed electroresistance and CMR effects are found to be remarkably complementary and strongly favoring a percolative phase separation picture.[3] It has been also observed that an applied current could lead to a transition from the electrically insulating charge-ordered state to a ferromagnetic (FM) metallic state.[4]

Recently, we focused on the influence of electric field/current on the transport properties in films of mixed-valent manganites[7–10] and found that a current with a high density can significantly affect the balance of multiphase coexistence and cause a series of changes of transport properties. In the present paper, we report current-induced switching behavior in $La_{1-x}Ca_xMnO_3$ (LCMO) thin films. Upon applying a large current (several milliamperes, $10^3$ A/cm$^2$), a switching from low to high resistive state takes place. However, the appeared high resistive states for compositions $x=0.2$ and 0.3 show different stabilities.

The present $La_{1-x}Ca_xMnO_3$ ($x=0.2$ and 0.3) thin films were grown on single crystal substrates of $SrTiO_3$ with (100) orientation using pulsed laser deposition technique. The thicknesses of the films $x=0.2$ and 0.3 were about 90 and 80 nm, respectively, controlled by deposition time. The electric measurements were done by using the standard four-probe technique in a closed cycle cryostat. In order to apply a current with high density, the films were patterned into a microbridge with the width of 50 $\mu$m and length of 200 $\mu$m using lithography technique. Four silver contacting pads were then evaporated on the sample, and the current leads were connected to the silver pad using a MEI-907 supersonic wire bonder to obtain low Ohmic contacts. A constant current source with a high voltage limit (Sorensen DCS 300 V-3.5 A) was employed when a large current needs to be applied.

The Curie temperature $T_C$ of present $La_{1-x}Ca_xMnO_3$ ($x=0.2$ and 0.3) thin films is revealed at 259 and 246 K, respectively, based on magnetic measurements using commercial superconducting quantum interference device. We found that the $T_C$ slightly fluctuates from sample to sample and also weakly depends on thickness. For $La_{0.7}Ca_{0.3}MnO_3$, the $T_C$ is comparable to that of its bulk, but for $La_{0.8}Ca_{0.2}MnO_3$, the $T_C$ is much higher than that of the bulk material (~190 K).[8] The treatments of bias current on the samples were performed by applying a large dc current for a short duration at specific temperatures, 218 and 208 K for $x=0.2$ and 0.3, respectively, where $R$ starts to increase in $R$-$T$ curves (see Fig. 1). The specific temperatures were chosen based on the following considerations. Firstly, the resistance at the specific temperatures is still small and the direct impact caused by self-heating effect should be weak. Secondly, the strong competition between electron itinerancy and self-trapping at the start where metal-like phase transforms to insulatinglike phase[11] would be strong and the influence of bias current on magnetic, structural distortion, and transport properties would be enhanced.

Figure 1 displays temperature dependent resistivity on cooling and warming for the induced states. The excitation

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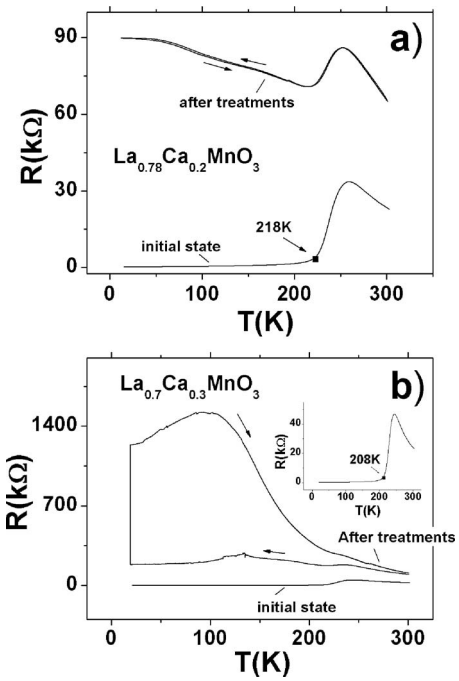


FIG. 1. The temperature dependence of resistivity of the induced high resistive state on cooling and warming processes for (a) $La_{0.8}Ca_{0.2}MO_3$ and (b) $La_{0.7}Ca_{0.3}MO_3$ thin films. For comparison, the *R-T* dependence of the initial state is presented for the corresponding samples. The measuring current is the same, 10 $\mu$A, for all curves. Inset of (b) shows the *R-T* details of the initial state for $La_{0.7}Ca_{0.3}MO_3$.

currents are 10.5 mA (density of $2.3\times10^3$ A/cm$^2$) and 6.7 mA (density of $1.6\times10^3$ A/cm$^2$) for $x=0.2$ and 0.3, respectively. The duration is the same, 5 min, for the both compositions. For comparison, the $R(T)$ curve of the initial state for the corresponding sample is also plotted. One can find that a switching from low to high resistive state appears for the both compositions upon applying the large dc currents. It is noteworthy that currents smaller than 10 mA for $x=0.2$ or lower than 6 mA for $x=0.3$ cannot induce the switching of resistive state. The induced state exhibits high resistivity in the temperature range from 10 to 300 K. A new resistive peak develops at low temperature, while the resistance anomaly around Curie temperature $T_C$ remains and the position of the resistance peak $T_p$ keeps nearly unchanged. An interesting feature is that the novel resistive state for the two compositions exhibits different stability. The high resistive state of sample $x=0.2$ is much stable. Exposing the sample in air and room temperature could not cause any change of the transport properties. The resistivity exhibits completely reversible behavior in temperature cycles [see Fig. 1(a)]. In contrast, sample $x=0.3$ shows unstable characteristics. After the sample was treated by a large current, cooling it to a low temperature of 20 K and keeping it for a short while, a relaxation to a higher resistive state takes place. Thus, a large resistive hysteresis on temperature is observed [see Fig. 1(b)].

The induced high resistive states exhibit high sensitivity to a weak current, such as 0.6–10.4 $\mu$A. Figure 2 shows the temperature dependences of the resistance for sample $x=0.2$ measured using different weak currents, 10.4, 5.1, 1.8, 0.9, and 0.6 $\mu$A, in cooling and warming processes. One can

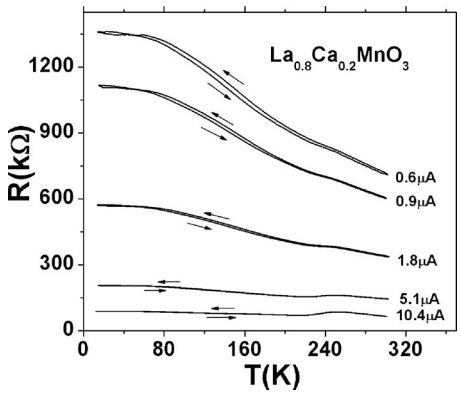


FIG. 2. The *R-T* dependences of $La_{0.8}Ca_{0.2}MO_3$ thin films with different weak currents, 10.4, 5.1, 1.8, 0.9, and 0.6 $\mu$A in cooling and warming processes. The resistive behavior of the high resistive state is independent of the sequence of the applied weak currents.

find that the enhanced resistance is highly sensitive to weak currents and fully reversible on temperature, indicating stable characteristics of the high resistive state. The resistive behavior is independent of the sequence of the applied weak currents. However, sample $x=0.3$ shows different behaviors. Figure 3 displays the *R-T* curves measured using small currents in a sequence of 10.1, 1.8, 0.6, and 10.1 $\mu$A in cooling and warming processes. Although the electric transport is also sensitive to weak currents, a large hysteresis still exists when the measuring current was tuned to 1.8 $\mu$A. In other words, a relaxation to a higher resistive state also occurs at 20 K for the case applying a current of 1.8 $\mu$A. However, when the measuring current is further tuned to a smaller value, 0.6 $\mu$A, the lowest current for the present experiments, the hysteresis almost disappears except for the noiselike switching at low temperatures. Our repeated measurements indicated that the noiselike resistivity appeared from 20 to 80 K is not caused by technical reasons but intrinsic. Shown in the inset of Fig. 3 are the repeatedly measured *R-T* curves using a same current of 0.6 $\mu$A. One can find that the noiselike behavior can be fully reproducible. At this stage, when we remeasured the resisvitity using a relative larger current, 10 $\mu$A, the resistance behavior almost returned to the previously relaxed high resistive state measured using the

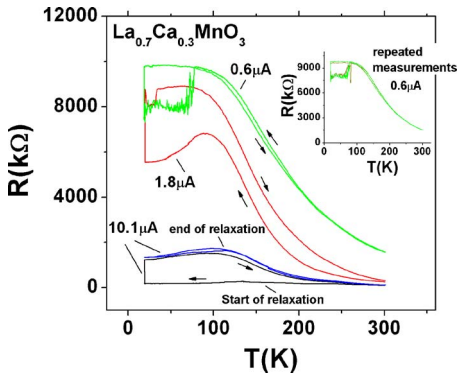


FIG. 3. (Color online) The *R-T* dependences of $La_{0.7}Ca_{0.3}MO_3$ thin films measured using small currents in a sequence of 10.1, 1.8, 0.6, and 10.1 $\mu$A in cooling and warming processes. Shown in the inset are the repeated *R-T* curves measured under the same current of 0.6 $\mu$A. The noiselike resistivity is fully reproducible.

same current of 10 $\mu$A. However, the resistive hysteresis nearly disappears. This phenomenon indicates that the sample is at a relatively stable state. We repeated the experiments on several samples and found that the developing trend of the resistive state for sample $x=0.3$ is nearly consistent with each other. Unlike the case of $x=0.2$, in which a stable high resistive state is immediately formed after flowing a large current, sample $x=0.3$ undergoes a relaxation process before a relative stable high resistive state is formed.

Self-heating effect is ineluctable upon flowing a large current. The application of a large excitation current is performed in a vacuum of $10^{-2}$ mbar since a proper vacuum is needed to maintain a thermal isolation. To make sure that the interesting observations are not caused by escape of oxygen due to the self-heating, various experiments were performed.[8,9] Simply annealing in a vacuum could only cause a shift of the position of peak resistance to lower temperature and make the resistance increase. No similar phenomena, as described in Fig. 3, were observed at all.

According to the phase diagram of $La_{1-x}Ca_xMnO_3$, the concentration $x=0.2$ is located at the border between inhomogenous antiferromagnetic and ferromagnetic ground states. The strong competition between different magnetic interactions would enhance the influence of bias current on transport and magnetic properties. Therefore, investigations on the influence of electric current on the transport properties were widely carried out for the critical composition $x=0.2$.[5,6,8,9,12] In contrast, the current effect on $x=0.3$ was seldom reported. One reason might be the current-induced unstable characteristics, which may be not easy to be detected. The different characteristics of phase separations for the two compositions may be responsible for the different switching behavior. Previous NMR (Ref. 13) and structural investigations[14] on $La_{0.8}Ca_{0.2}MnO_3$ systems have confirmed that the phase-separated ground state is constituted of two ferromagnetic phases with different orbital orders (OOs). In one phase, an antiferrodistorsive-type OO is favored by FM superexchange interactions, giving insulating characteristics, while in the other phase, the ferrodistorsive-type OO promotes double-exchange interactions, exhibiting metallic behavior. However, for $La_{0.7}Ca_{0.3}MnO_3$ system, a direct neutron scattering evidence indicated that the multiphase coexistence is between a conventional FM metal and a phase with strong short range charge ordering.[15] Thus, it is understandable that the two compositions may display different transport behaviors upon flowing a large current.

It was experimentally found that an electric field could directly affect the directional order of orbitals and, thus, alter the magnetic and conducting states.[16] When the applied current is high enough, the associated electric field in the phase space may be sufficiently high to contribute to the crystal fields,[17] thus strongly impacts the local distortion, orbital order, and charge order and enforces a thorough change in the topology of the phase coexistence. Because of the different local environments and crystal fields in different compositions, the interaction between electric field with orbital order or charge order should be different, and the caused results may be various. For $x=0.2$, the high resistive state is immediately formed when the current treatments are finished, but for $x=0.3$, a relaxation process is required before the resistive state is stabilized. The action of the strong current may be realized either by means of carrier injection or by direct interaction with elastic forces.[6,17] It can be understandable that the formation process of high resistive state could not be instantaneous and a relaxation process may be required, depending on the specific local environment of microstructures. Figure 3 manifests the relaxation process of the high resistive state for sample $x=0.3$. One can find that the sample always tends to relax to a higher resistive state before being stabilized. The intrinsic behavior of the noiselike resistance may reflect the instability of the induced distortion in the relaxation process.

In summary, current-induced switching of resistive state has been investigated in $La_{1-x}Ca_xMnO_3$ ($x=0.2$ and 0.3) thin films. A switching from low to high resistive state takes place upon applying a large dc current. However, the two compositions display different switching behaviors. For $x=0.2$, the high resistive state is immediately formed when the current treatments are finished, but for $x=0.3$, a relaxation process is required before the resistive state is stabilized. The different characteristics of phase separations for the two compositions are considered for understanding the different switching behaviors.

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