Current-induced effect on the resistivity of epitaxial thin films of $La_{0.7}Ca_{0.3}MnO_3$ and $La_{0.85}Ba_{0.15}MnO_3$

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Electric-current-dependent resistance has been studied in epitaxial thin films of $La_{0.7}Ca_{0.3}MnO_3$ and $La_{0.85}Ba_{0.15}MnO_3$. Attention was focused at the influence of the applied dc current on the resistance of these epitaxial thin films in the absence of a magnetic field. A significant change in the ratio of the peak resistance at different currents or current resistance was found to be ~23%–26% with a current density up to 8×10^4 Acm^{-2}. For both $La_{0.7}Ca_{0.3}MnO_3$ and $La_{0.85}Ba_{0.15}MnO_3$ compounds, the dependence of the measured resistance on the current revealed a good linear relationship. Although the nature behind such an effect has not been well understood yet, the feature that the resistance in doped manganese oxides could be easily controlled by the electric current should be of interest for various applications such as field effect devices. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1587001]

Perovskite compounds $La_{1-x}A_xMnO_3$, where A stands for Ca, Ba, Sr, Pb, etc., have been the subject for intense studies due to their rich varieties of phenomena such as colossal magnetoresistance (CMR), ferromagnetism with metallic conduction, and charge/orbital ordering. The insulator–metal and paramagnetic–ferromagnetic phase transitions found at low temperatures lead to a dramatic reduction in their resistivity and subsequent CMR.[1] The point at which the transition takes place is referred as the Curie temperature T_C. The values of T_C could be modified by the application of magnetic fields, pressure, current bias, and illumination with light or *x* ray. It is believed that these phenomena are related to the strong coupling between spin, charge, and orbital degrees of freedom. The double exchange mechanism,[2] Jahn-Teller effects,[3] and phase separation[4] are commonly used to explain the experimental observations. Recent interests of research have been attracted to the influence of electric field or the electric current on the CMR materials for their potential applications.[5–9] Studies on the effect of the electric field in perovskite manganites included colossal electroresistance (ER) and strong current-induced abrupt resistivity jumps, many performed on charge-ordered samples $Pr_{1-x}Ca_xMnO_3$ involving current injection into highly conducting filamentary paths. The strong nonohmic conductivity in such systems leads to the collapse of the insulating charge-ordered state. A recent investigation reported by Wu *et al.*[6] showed that for a LCMO/$PbZr_{0.2}Ti_{0.8}O_3$ heterostructure, a biased electric field could also cause a remarkable decrease in the peak resistance. Wahl *et al.*[7] also found a remarkable depression in magnetization driven by a bias current in noncharge-ordered ferromagnetic manganese oxide $Pr_{0.8}Ca_{0.2}MnO_3$.[7] They attributed such a current driven magnetization decrease to the excitation of spin waves by the electric current, rather than the filamentary picture usually invoked for nonlinearity in manganese oxides. However, the physics behind such ER and CMR effects are far from being understood.

In this letter, we report the current effect (instead of electric-field effect) on the peak resistance of the ferromagnetic transition in epitaxial thin films of $La_{0.7}Ca_{0.3}MnO_3$ and $La_{0.85}Ba_{0.15}MnO_3$ grown on single crystal of $SrTiO_3$. We observed that the peak resistance in these thin films was depressed significantly by an increased dc current, even in the absence of an applied magnetic field. Although the nature of the observed effect in these epitaxial CMR thin films has not been clarified yet, such a feature with a strong drop in resistance under the application of an electric current might be of interest for fundamental physics and of great technological potential for CMR thin films.

Our samples were grown on single crystal substrates of $SrTiO_3$ using pulsed laser deposition. Sintered ceramic disks of $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) and $La_{0.85}Ba_{0.15}MnO_3$ (LBMO) with a diameter of 50 mm and thickness ~4 mm were used as the targets. The deposition temperature was 750 °C as measured by a *k*-type thermocouple in the heater block. The deposition takes place in a pure oxygen atmosphere of the pressure of 2×10^{-1} mbar. The energy of the laser beam is ~300 mJ, wavelength is 308 nm, and the pulse frequency is 6 Hz. The thickness of the film was about 100 nm, controlled by deposition time. A postannealing at 800 °C for 1 h was taken in air.

X-ray diffraction has been performed on all grown films to examine their phase structures. The diffraction spectra always reveal sharp peaks of the formed ABO_3 phase with the *c*-axis perpendicular to the substrate surface. Besides the reflection from substrate and the (00ℓ) peaks of the LCMO or LBMO, no other peaks are visible, demonstrating that the grown films are of single phase (Fig. 1). The inset of Fig. 1 gives a rocking curve of the (004) peak for LCMO. It shows a very small value of the full width at half maximum, typically smaller than 0.2°, implying a high epitaxy and a good crystallinity of our films.

The electric measurements were done by using the stan-

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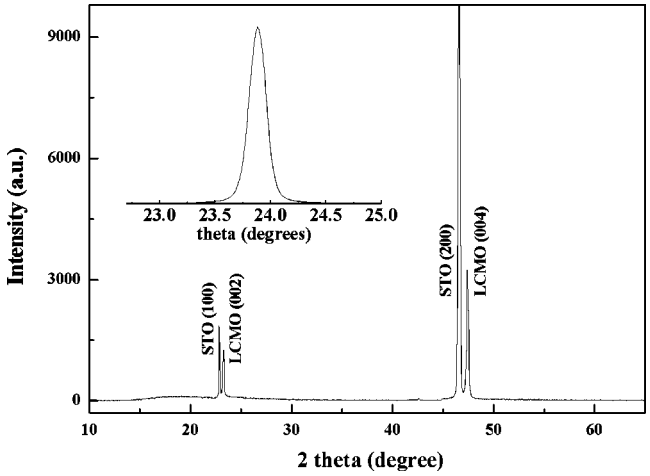

FIG. 1. Typical x-ray diffraction spectra for LCMO epitaxial thin films. The inset plot shows the rocking curve measured on (004) peak of LCMO.

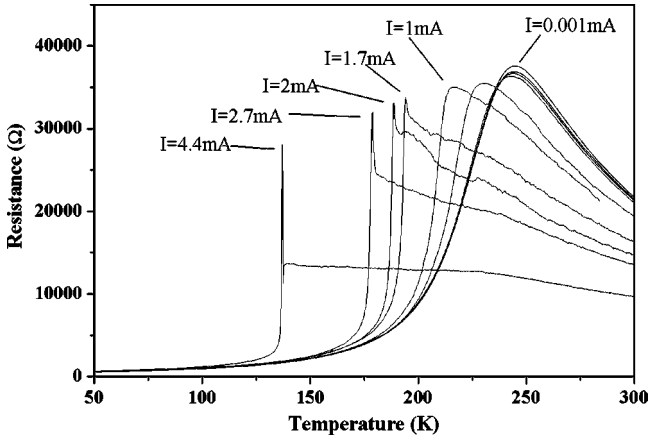

FIG. 3. The $R-T$ curves for a LBMO film with variety of bias currents. The applied currents from the highest ER peak to the lowest peak are 0.001 mA, 0.1 mA, 0.2 mA, 0.3 mA, 0.7 mA, 1 mA, 1.7 mA, 2 mA, 2.7 mA, and 4.4 mA, respectively.

dard four-probe technique in a closed-cycle cryostat. The films were patterned into a microbridge with a width of 50 μm and length of 200 μm using the photoresist and lithography techniques. 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 good ohmic electric contacts. As the resistance of the measured film samples is quite large, the current was provided by a constant current source with a high-voltage limit (Sorensen DCS 300 V-3.5 A).

In this experiment, we focused on the change in the ratio of the peak resistance at different currents, i.e., current resistance (CR), instead of the usual ER which is the change in the ratio at different electric fields. To ensure that the obtained results are reliable, the CR measurements were performed on a number of samples and all the results were quite consistent. To avoid the influence of self heating as much as possible, all measurements were done in a slow warming up process, i.e., the temperature was increased gradually from a predetermined low temperature to 300 K during the measurements. Figure 2 presents a typical dependence of resistance of the LCMO film on temperature under different currents from $I=1$ μA to 4 mA. The conduction is metallic in the low-temperature region and semiconductive when T exceeds $T_p=253$ K in the case of a small applied current (1 μA), where T_p is the temperature corresponding to the maximum resistance. With the increase of the applied current, the most striking observation is the significant decrease of the peak resistance (R_p). R_p takes $\sim$22.8 kΩ for $I=1$ μA and $\sim$17.4 kΩ for $I=4$ mA, the relative reduction is $\sim$23% as large.

In LCMO, the concentration of Mn^{4+} is near the optimum value ($\sim$0.33) for the ferromagnetic order and metallic conduction. A study of the current effects is also carried out for LBMO, in which the content of Mn^{4+} is 0.15, near the critical value for the long-range ferromagnetic order. Interestingly, similar behaviors were observed in LBMO though the maximum T_p/R_p is a little lower/higher compared with LCMO (Fig. 3). The CR is $\sim$25% under a current density of $\sim 9\times10^4$ Acm^{-2}.

The CR effect may be a universal feature of the manganites that show a metal–insulator transition. This point is even clear from Fig. 4, in which the peak resistance was presented as a function of applied current. It shows a linear decrease of R_p with current at a rate depending on the maxi-

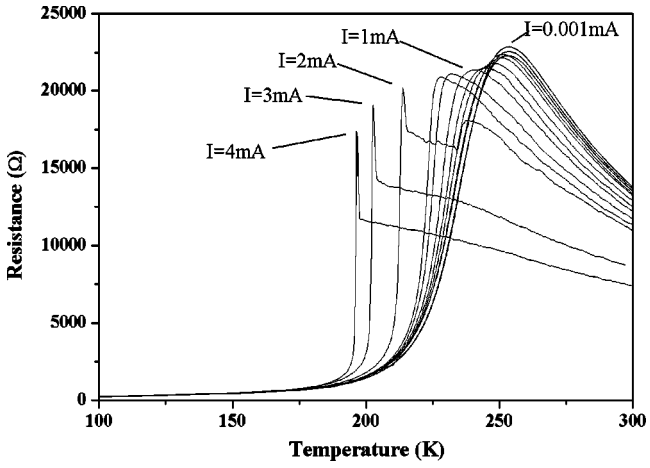

FIG. 2. The $R-T$ dependences for a LCMO film with different bias currents. The applied currents from the highest peak to the lowest peak are 0.001 mA, 0.1 mA, 0.2 mA, 0.3 mA, 0.4 mA, 0.6 mA, 0.8 mA, 1 mA, 1.2 mA, 1.4 mA, 2 mA, 3 mA, and 4 mA, respectively.

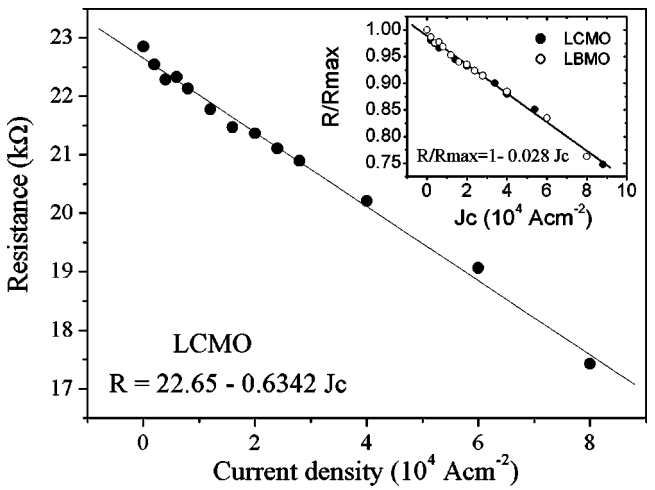

FIG. 4. The peak resistance as a function of current density for the LCMO film. A good linear dependence was found. The inset shows the normalized R_p-Jc relation for both LCMO and LBMO films.

mum resistance in the low current limit (R_{max}). The slope is ~0.63 for LCMO and ~1.01 for LBMO. However, when normalizing the peak resistance by R_{max} (it is ~22.65 kΩ for LCMO and ~37.14 kΩ for LBMO), we obtained the same linear relation for both films as shown in the inset of Fig. 4. This is a very surprising observation. We find that for two different samples, the relative resistance decreases with the current

$$R/R_{max} = 1.0 - 0.028Jc.$$

We have not tried to apply a higher current to avoid damaging the sample. However, we expect that the CR will increase with the increase of current density.

It is certain that such a reduction of the peak resistance could not be caused by Joule heating, which may influence the measurement of the location of T_p instead of the value of the maximum resistance. The small size of the sample also eliminated the effects associated with sample inhomogeneity, which may influence R_p.

From Figs. 2 and 3, it can be found that the metal–insulator transition becomes sharper and sharper with the increase of current. This could be a result of self-heating. With the increase of resistance in the warming process, Joule heat increases rapidly, which rapidly drives the system passing the resistive transition. However, the thermometer in the heater block cannot follow the rapid temperature change of the film, and it always displays a delayed temperature. As a result, the quick changed real temperature in the film leads to a sharper transition. The quicker the real film temperature varied, the shaper $R-T$ peak observed. Accompanying the reduction of the peak resistance, the resistive transition significantly shifts to lower temperatures as shown in Figs. 2 and 3. This effect could not be intrinsic. It is also a reflection of the effect of self-heating, which makes the real temperature of the film much higher than that detected by the thermometer.

The reduction of the peak resistance observed in this experiment may be understood qualitatively from the strong interaction between carrier spins and localized spins in Mn ions. It is caused by the electric current instead of the electric field in the previous experiments. Due to the electrostatic shielding, our observation could not be made if only a bias field applied. Considering a fact that the magnetic coupling between Mn ions is produced by the e_g electrons which prefer to hold their spin orientation when hopping among Mn ions. Therefore, the strength of the magnetic coupling is proportional to the hopping rate of the e_g electron as predicted in the double exchange theory for metallic ferromagnetism in doped manganites.[3] Based on such a picture, the strong Hund's rule coupling forces the spins of conduction electrons to be parallel to a localized spin on the same site. If the two localized spins on the nearest-neighbor sites are parallel, the conduction electron could easily move to the neighboring site. Otherwise, a higher energy is needed due to the Hund's rule coupling. When a current is applied, the conduction electrons are forced to move forward whether or not their spin is parallel to neighboring localized spin. However, the spin exchange coupling conserves the total spins, and the movement of the conduction electron will produce a tendency to force the localized spins to be parallel. Similar physics were discussed in the spin transfer theory for magnetic multilayers structures.[10,11] Ordering of localized spins may reduce the spin scattering of conduction electrons. Thus, a direct consequence of the increase of applied current is the ordering of localized spins, which may reduce the spin scattering of conduction electrons. In this picture, a high-temperature shift of T_p is expected, and, then the reduction of R_p with applied current is understandable.

On the other hand, the phase separation scenario seems a universal feature in CMR manganites.[4,12,13] 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. Previous researchers ascribed CMR, ER, or CR to the percolative mechanism of phase separation.[6,13,14] One might consider a state of equilibrium in which metallic ferromagnetic clusters, embedded in a insulate matrix, approach a state of percolation. An applied electric field or current perturbs the coexistence of different phases and sets up filamentary currents across the insulate region, inducing the further polarization of ferromagnetic regains and the reduction of resistance. The present large reduction of the peak resistance, induced by an electric current, may be ascribed to the aforementioned two aspects; one is the strong interaction between carrier spins and localized spins in Mn ions, the other is the percolative mechanism of phase separation.

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[1] J. M. D. Coey, M. Viret, and S. Von Molnar, Adv. Phys. **48**, 167 (1999), *Colossal Magnetoresistive Oxides*, edited by Y. Tokura (Gordon and Breach, New York, 2000).
[2] C. Zener, Phys. Rev. **82**, 403 (1951).
[3] A. J. Millis, P. B. Littlewood, B. I. Shraiman, Phys. Rev. Lett. **74**, 5144 (1995).
[4] A. Moreo, S. Yunoki, and E. Dagotto, Science **283**, 2034 (1999).
[5] S. B. Ogale, V. Talyansky, C. H. Chen, R. Ramesh, R. L. Greene, and T. Venkatesan, Phys. Rev. Lett. **77**, 1159 (1996).
[6] T. Wu, S. B. Ogale, J. E. Garrison, B. Nagaraj, A. Biswas, Z. Chen, R. L. Greene, R. Ramesh, T. Venkatesan, and A. J. Millis, Phys. Rev. Lett. **86**, 5998 (2001).
[7] A. Wahl, C. Simon, S. Mercone, D. Saurel, and C. Martin, cond-mat/0206073.
[8] S. Mercone, A. Wahl, C. Simon, and C. Martin, Phys. Rev. B **65**, 214428 (2002).
[9] Y. Yuzhelevski, V. Markovich, V. Dikovsky, E. Rozenberg, G. Gorodetsky, G. Jumg, D. A. Shulyatev, and Y. M. Mukovskii, Phys. Rev. B **64**, 224428 (2001).
[10] J. C. Slonczewski, J. Magn. Magn. Mater. **159**, L1 (1996); *ibid.* **195**, L261 (1999).
[11] L. Berger, Phys. Rev. B **54**, 9353 (1996); J. Appl. Phys. **89**, 5521 (2001).
[12] M. Uehara, S. Mori, C. H. Chen, and S.-W. Cheong, Nature (London) **399**, 560 (1999).
[13] M. Fath, S. Freisem, A. A. Menovsky, Y. Tomioka, J. Aarts, and J. A. Mydosh, Science **285**, 1540 (1999); C. P. Adams, J. W. Lynn, Y. M. Mukovaski, A. A. Arsenov, and D. A. Shulyatev, Phys. Rev. Lett. **85**, 3954 (2000).
[14] V. Markovich, E. Rozenberg, Y. Yuzhelevski, G. Jung, G. Gorodetsky, D. A. Shulyatev, and Y. M. Mukovskii, Appl. Phys. Lett. **78**, 3499 (2001).