Abnormal electroresistance effect induced by electric currents in La$_{0.9}$Ba$_{0.1}$MnO$_3$ thin films

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Abstract

In this work, we report an abnormal electroresistance (ER) effect induced by electric current in La$_{0.9}$Ba$_{0.1}$MnO$_3$ thin films. La$_{0.9}$Ba$_{0.1}$MnO$_3$ thin films with thickness $\sim$100 nm were prepared by using pulsed laser deposition technique. The transport behaviors were investigated under various applied currents in the absence of magnetic field. Electric resistance peaks at insulation-metal transition temperature. It was found that the peak value of the resistance gradually decreases with increasing current when the applied current is not too large. At this stage, the films can completely revert to their initial state after the applied current is removed. However, when the current reaches a critical value, a novel state appears, in which a remarkable resistive peak appears at temperatures well below the Curie temperature. An intriguing feature is that the appeared resistance peak at low temperatures turns out to be extremely sensitive to a weak current. Even a very small current could greatly depress the height of the peak, abnormal ER effect appears. Physics related to the appearance of the novel state and the abnormal ER effect is discussed.

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Keywords: Abnormal electroresistance; La$_{0.9}$Ba$_{0.1}$MnO$_3$ thin film

1. Introduction

Colossal magnetoresistance (CMR) effect in mixed-valent manganese oxides has triggered intense scientific activity in recent years. Lots of experiments on polycrystals, single crystals, and thin films have been carried out to investigate the CMR behavior and its intrinsic physics. It has been suggested that the largest MR is associated with spatial inhomogeneity related to multiphase coexistence, which generically causes a sensitivity of physical properties to external perturbations, such as application of magnetic fields, pressure, current bias, or light illumination. The possibility of using a wide range of perturbations to influence the transport properties also increases their technological potential. An increasing interest has been recently attracted to the so-called colossal electroresistance (ER) effect in CMR manganites [1–9]. Namely, their electronic conductance may be controlled by an electric current or a static electric field. Significant change of electric resistance induced by static electric field has been demonstrated using field effect configurations [1]. It has been observed that an electric current could trigger the transformation of the electrically insulating charge-ordered state to a ferromagnetic metallic state [2]. Current-induced switching of resistive states in La$_{0.8}$Ca$_{0.2}$MnO$_3$ single crystal has been also reported [3]. Furthermore, several groups [8,9] investigated the influence of a pulsed AC current on the transport properties in perovskite oxides sandwiched between different metal electrodes. A reversible resistance switching can be induced in the sandwiched systems.

Recently, we focused on the influence of electric field/current on the transport properties in films of mixed-valent manganites [4–7]. Remarkable ER effect induced by electric current/static electric field has been observed in thin films of various manganites. Further studies revealed 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 this paper, we report an abnormal electroresistance effect introduced by electric current in La$_{0.9}$Ba$_{0.1}$MnO$_3$ thin films. It was found that the transport resistance gradually decreases with increasing current when the applied current is not too large. At this stage, the films can completely revert to their initial state after the applied current is removed. However, when the current reaches a critical value, a novel state appears, in which a remarkable resistive peak appears at temperatures well below the Curie temperature.

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doi:10.1016/j.mseb.2007.07.101
More intriguing is that the appeared resistance peak turns out to be extremely sensitive to a weak current. Even a very small current could greatly depress the height of the peak, abnormal ER effect appears.

2. Experimental

The present La$_{0.9}$Ba$_{0.1}$MnO$_3$ (LBMO) thin films were grown on single crystal substrates of SrTiO$_3$ with (1 0 0) orientation using pulsed laser deposition (PLD) technique. Disks of stoichiometric La$_{0.9}$Ba$_{0.1}$MnO$_3$ were used as the targets. The deposition took place in a pure oxygen of 100 Pa. The energy of the laser beam was ∼0.2 J, wavelength was 308 nm, and the pulse frequency was 6 Hz, respectively. The substrate temperature was 750 °C as measured by a k-type thermocouple inserted into the heater block. The thickness of the films was about 100 nm controlled by deposition time. A post-annealing at 800 °C for 1 h was made in air in order to avoid oxygen deficiency. The compositions of the films determined by energy dispersive X-ray analysis (EDAX) were very close to the stated compositions.

The experiments of X-ray diffraction 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 (0 0 l) peaks of the LBMO, no other peaks are visible, demonstrating that the grown films are of single phase and well oriented. The electric measurements were done by using the standard four-probe technique in a closed cycle cryostat. In order to apply a current with a high density, the films were patterned into a micro-bridge with the width of 50 μm and length of 200 μm using lithography technique (see inset of Fig. 1). Four silver contacting pads were then evaporated on the sample and the contacting pads 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 flow needs to be applied. Magnetic measurements were performed using a Superconducting Quantum Interference Device (SQUID) magnetometer.

3. Results and discussion

The measurements of thermal magnetization under a magnetic field of 100 Oe reveals that the Curie temperature $T_C$ of La$_{0.9}$Ba$_{0.1}$MnO$_3$ film is at ∼300 K (see Fig. 1), which is much higher than that of its bulk material (the $T_C$ of the bulk La$_{0.9}$Ba$_{0.1}$MnO$_3$ is ∼185 K). Many properties in strained CMR thin films significantly differ from that of bulk compounds. The bulk LBMO could be considered as a pseudocubic structure. As being deposited on STO, the in-plane lattice tends to adopt the same structure as STO substrate, and the out-of-plane parameter changes correspondingly to maintain unit cell volume. The out-of-plane lattice parameter of LBMO thin films derived from X-ray diffraction data is found smaller than the bulk parameter, indicating a tensile strain effect. In most cases, tensile strain suppresses ferromagnetism and reduces ferromagnetic Curie temperature $T_C$ in CMR thin films, which is generally interpreted by considering a strain-induced distortion of MnO$_6$ octahedra. However, some anomalous results have also been reported, showing ferromagnetism enhanced by tensile strain. It was suggested [10] that the tensile strain in La$_{0.9}$Ba$_{0.1}$MnO$_3$ films makes the energy level of the $d_{x^2-y^2}$ orbital along the in-plane direction become more stable than that of the $d_{3z^2-r^2}$ orbital, inducing the increase of conductive electrons having larger transfer integrals which leads to the increase of $T_C$.

Fig. 2 displays the temperature dependent resistance of a LBMO film measured under different currents from $I=10$ μA to 8.6 mA. All measurements were carried out in a slow cooling process. Starting temperature is 300 K, and arriving temperature is about 10 K. The cooling rate is ∼3 K/min. The measurements are instantly performed when the applied current is tuned to a desired value. A striking observation is the significant decrease of the peak resistance ($R_p$) with increasing current. The relative reduction of $R_p$ reaches ER ∼27% for current increase from $I=10$ μA to 6.5 mA. We call it normal ER effect. The self-heating effect is serious when a large current is applied. However, it does not affect the measured value of the peak resistance, although it may shift the location of the ferro-paramagnetic
transition [7]. The widening of the resistance peak with the increase of current recorded in cooling process is associated with self-heating effect. This can be understandable considering the asymmetry of the resistance peak [6]. Apart from the main transitions at \( T_C \) a small jump of resistance occurs with increasing current flow. Such a similar feature was also observed in Ca-doped manganites.

We can also find, from Fig. 2, when the initially applied current is lower than 6.5 mA the residual resistance remain unchanged. The films can completely revert to their initial state after the applied current is removed. However, as long as the applied current exceeds 6.5 mA the residual resistance abruptly increases. At this stage, the films cannot return to the original state after removing the applied current. Instead, a novel state appears.

Fig. 3 presents the re-measured \( R-T \) curves in both cooling and warming process under a small dc current of \( I = 0.05 \) mA after a series of \( R-T \) measurements using different currents. The currents were applied in a sequence of 0.05, 4.0, 6.5, 7.5 and 8.6 mA. The cooling rate is the same as the warming rate, \( \sim 3 \) K/min. All the \( R-T \) curves are found almost reversible in the temperature cycles. The state developing with the increase of applied current is clearly manifested. When the applied current exceeds a critical value 6.5 mA, an additional peak at low temperature \( T_L \) is developed with the resistance peak at \( T_C \) remaining. With increasing current from 7.5 to 8.6 mA, the additional peak at \( T_L \) increases significantly and the residual resistance abruptly increases. A most intriguing feature is that such an induced resistance peak at \( T_L \) is extremely sensitive to an applied current while the resistance peak at \( T_C \) keeps insensitive to small currents. Fig. 4 displays the \( R-T \) curves measured on cooling using small dc currents for the state introduced by 8.6 mA. One can find that even a rather weak current can strongly influence the magnitude of the resistance peak at \( T_L \). The relative reduction of the resistance peak \( (R_{\text{small-current}} - R_{\text{big-current}})/R_{\text{big-current}} \) at \( T_L \) reaches \( \sim 1740\% \) upon current changing from 0.0002 to 0.08 mA. The ER effect of the novel state is extremely large compared to the normal ER mentioned above. We call it abnormal ER effect. The novel state with high resistivity induced by an electric current or a static electric field seems a universal phenomenon in manganites with electronic phase separation. Similar behaviors were observed not only in films but also in single crystals of mixed-valent manganites. The appearance of the novel state is a natural effect of the coexistent multiphase upon an application of a large dc current or a high electric field.

For the electroresistance effect caused by current excitation in mixed-valent manganate oxides, one tended to explain it by considering several possible mechanisms, percolative phase separation [1,2,6,7], strong interaction between carrier spins and localized spins in Mn ions [7], as well as the phonon-assisted delocalization [11]. For understanding the novel state introduced by a large current and the abnormal ER effect in the present system, the mechanism of percolative phase separation should be a key element. It has been a well established fact that metallic and insulating phases coexist in a broad range of phase space especially for the low-doped manganites. When the applied current is high enough, the associated electric field distributed in the phase space may strongly impact the orbital order of the phases and enforce a thorough change in the topology of the phase coexistence. Eventually a new state with new coexistence of the phases may appear. Actually, the developing of \( R-T \) curve shown in Fig. 2 indicates the modulation process of the applied large current on the coexistence and reflects the evolution of the film state with the increase of current. The reduction of the conductivity with decreasing temperature below \( T_C \) can be a result of the volume increase of the insulating phase and the metallic-like behavior below \( T_L \) might suggest percolative conduction through metallic regions embedded in insulating regions. An abundance of insulating domains would lead to poor connection of minority metallic domains and make residual resistivity increase. The introduced state with high resistivity is instable, behaving more sensitive to the external perturbations compared to the as-prepared state. A small current could strongly influence
the formed coexistent multiphase, resulting in an abnormal ER effect.

4. Conclusion

We have investigated the influence of applied current on the transport properties in La$_{0.9}$Ba$_{0.1}$MnO$_3$ thin films. A novel state with high resistivity can be introduced by a large dc current. The novel state behaves metastable characteristics. A small current could strongly influence the transport properties, resulting in an extremely large ER effect. The possibility to control the transport resistance by an electric current instead of a magnetic field provides new methods for controlling electric/magnetic phases in perovskite manganese oxides, accordingly, novel functional magnetic, and electric devices can be expected.

Acknowledgements

This work has been supported by a grant of the Research Grant Council of Hong Kong (Project No. HKU 7024/04P), the CRCG of the University of Hong Kong, the National Natural Science Foundation of China, and the Beijing Natural Science Foundation of China with Grant No. 2072005.

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