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The abnormal electroresistance behavior observed in epitaxial La_{0.8}Ca_{0.2}MnO₃ thin films

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Abstract

La_{0.8}Ca_{0.2}MnO₃ epitaxial thin films were fabricated on SrTiO₃ (100) substrates by using pulsed-laser deposition (PLD) technique. The transport behavior upon treatments by applying a large dc current has been investigated. It is found that when the applied dc current exceeds a threshold value the electric resistivity in these films could be significantly enhanced in the whole measured temperature range from 10 to 300 K. Simultaneously, abnormal electroresistance behavior appears. The enhanced resistance turns out to be sensitive to a weak current. Even a very small dc current could remarkably depress the high resistance, showing an unusual colossal ER effect. ER reaches ~1175% at temperatures lower than ~50 K, and ~705% at 300 K for a current changing from 0.7 to 11 µA. Structure measurements on micro-area by micron-XRD technique were carried out. No change of the structure and lattice parameter was found upon current treatments.

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1. Introduction

The phenomenon of colossal magnetoresistance (CMR) in mixed-valent manganites has attracted considerable attention in recent years due to the related physics and potential applications. A large number of experimental and theoretical studies on these mixed-valent systems have brought out rich variety of phenomena. The closeness of the free energies of various competing electronic, magnetic, and orbital states causes multiphase coexistence in these systems. The coexistent multiphase behave sensitively to external fields and radiation. Application of magnetic fields, high electric fields, or irradiation with X-rays may destroy insulating phases and lead to a conducting state. The possibility to use wide variety of perturbations to realize the reduction of resistance increases the technological potential considerably for this kind of materials. Since miniaturization is a key requirement in all technologies, it is desirable to have these materials in the form of thin films. Furthermore, the high-electric-field/current effects can be easily realized in thin-film samples if a suitable construction or heterostructure geometry is used.

Recently, more and more attentions have been attracted to the electric-current/field induced colossal electroresistance (CER) effect [1-6]. It has been observed [1] that an applied current could lead to a transition from the electrically insulating chargeordered (CO) state to a ferromagnetic (FM) metallic state, even for $Y_{0.5}Ca_{0.5}MnO_3$ in which a large magnetic field (~40 T) has no effect on the charge-ordered state. Considering the strong effect of external perturbations on the balance of multiphase coexistence, it can be expected that a sufficiently large current flowing and the possible produced magnetic fields may thoroughly disturb the subtle balance of multiphase coexistence and then might induce a new equilibrium state of coexistence, in which various novel CMR characteristics might appear. In comparing with bulk samples, a current with a very high density could be easily applied to a micro-bridge of CMR thin films thus the influence of high current density on the multiphase coexistence state can be studied.

Manganite perovskites $La_{1-r}Ca_rMnO_3$ are characterized by a complex phase diagram containing a rich variety of magnetic and electronic phases. Because of the subtle competition between charge-carrier motion, magnetic spin and orbital moments, phase-separated state becomes a stable ground state, especially for the low-doped samples. In this paper, we report an observation of an abnormal colossal electroresistance effect

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introduced by applying a large dc current in $La_{0.8}Ca_{0.2}MnO_3$ epitaxial thin films.

2. Experimental procedure

The present La_{0.8}Ca_{0.2}MnO₃ (LCMO) thin films were grown on single crystal substrates of SrTiO₃ (STO) with (100) orientation using pulsed laser deposition technique. A disk of stoichiometric La_{0.8}Ca_{0.2}MnO₃ was used as the target. The deposition took place in pure oxygen of 1 mbar. The energy of the laser beam was ~200 mJ, 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 film 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 composition of the film determined by energy dispersive X-ray analysis (EDAX) was very close to the stated composition.

The electric measurements were done by using the standard 4-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 μ m and length of 200 μ 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 flow needs to be applied. Magnetic measurements were performed using a Superconducting Quantum Interference Device (SQUID) magnetometer.

3. Results and discussion

The experiments of X-ray diffraction reveal sharp peaks of the formed ABO₃ phase with the *c*-axis perpendicular to the substrate surface. Besides the reflection from substrate and the (00l) peaks of the LCMO, no other peaks are visible, demonstrating that the grown films are of single phase and highly epitaxial.

The temperature dependent magnetization measured at 100 Oe reveals the Curie temperature $T_{\rm C}$ of the present La_{0.8}Ca_{0.2} MnO₃ film is ~260 K, which is higher than that of its bulk material (~190 K) [7]. Such a deviation is consistent with a previous report [8], in which one found that the unit cell volume of a La_{0.8}Ca_{0.2}MnO₃ film is much smaller than that of its bulk. The reduction in the unit cell volume due to strain effect would enhance the transfer integral of electron hopping between Mn³⁺ and Mn⁴⁺ and thus increase the $T_{\rm C}$. The value of $T_{\rm C}$ also depends on the fabrication process. With different growth, the $T_{\rm C}$ might slightly vary in a certain range around 260 K.

An interesting observation was the current induced ER. Namely, the samples were first excited by a large current with a current density $\sim 10^4$ A cm⁻² and then measured with a weak current. The high exciting current induced a metalstable state which turned out to be extremely sensitive to the applied currents. Even a very weak current $\sim 10^0 \mu A$ (density $\sim 10^1 A \text{ cm}^{-2}$) can

cause remarkable reduction in resistivity, leading to a giant ER. To study the current induced instability, dc currents with high density were applied to these samples at a specific temperature (~218 K), where the resistance starts to increase in R-T curve, for a short duration δt . We chose this specific temperature because that the resistance at 218 K is still small hence the direct impact caused by self-heating effect should be weak. In addition, at such a point, where the metal-like state starts to transfer to insulating-like state, enhanced influence of the current on transport and magnetic properties is expected owing to the strong competition between different magnetic interactions. We found that an application of a suitable large dc current at such a manner would result in a strong increase of electric resistance in the whole temperature range from 10 to 300 K and, more important, the enhanced resistance is extremely sensitive to weak currents.

In this research all samples were first excited by a large current for a duration δt , then their transport behavior were measured using small currents. Fig. 1 displays the temperature dependent resistance measured using a small current of 0.01 mA for the states excited by dc currents of 4.7, 7.9 and 10.6 mA, respectively. The excitation currents are applied at 218 K for a same duration of 5 min. For comparison the R-T curve of the asprepared state is also plotted. The state evolution with increasing the excitation current is manifested. The excitation by a current of 4.7 mA caused no change in the transport properties. Increasing the excitation current from 4.7 to 7.9 mA resulted in a slight increase of resistivity, meanwhile, a small cusp at ~ 140 K was developed. The enhanced resistance by 7.9 mA was found not sensitive to weak currents. Further increasing the excitation current to 10.6 mA (the current density $\sim 2.1 \times 10^5$ A cm⁻²) lead to a remarkable increase of resistance in the whole temperature range from 10 to 300 K. The residual resistance was significantly enlarged, but the resistance anomaly near $T_{\rm C}$ still remained and the position of the resistance peak $T_{\rm P}$ kept unchanged. More attractive finding was that the enhanced resistance in this case turned out being extremely sensitive to weak currents. Even a very small dc current could depress the high



Fig. 1. The temperature dependent resistance of a $La_{0.8}Ca_{0.2}MnO_3$ thin film measured using a same small current of 0.01 mA for the states induced by dc currents of 4.7, 7.9 and 10.6 mA, respectively. For comparison, the *R*-*T* curve of the as-prepared state is also plotted.

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