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Investigations on electroresistance effect in epitaxial manganite films using field effect configurations

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The influence of static electric field on the transport properties in $\mathrm{La_{0.7}}A_{0.3}\mathrm{MnO_3}$ (A=Ca, Ba) epitaxial thin films was investigated by using field effect configurations (FEC). A single layer manganite film was deposited on $\mathrm{LaAlO_3}$ (LAO) substrate by pulsed laser deposition technique, and then a simple FEC was formed on it using the lithography technique, in which the manganite film was used as a channel, and the LAO substrate as a gate. Surprising results were achieved by employing such a FEC. The transport resistance increases with a positive gate voltage but decreases with a negative bias, which means the electroresistance (ER) effect changes sign with the field direction. The observed reduction of resistivity for the $\mathrm{La_{0.7}Ca_{0.3}MnO_3}$ and $\mathrm{La_{0.7}Ba_{0.3}MnO_3}$ channels reaches ~32% and ~34% upon a bias of −80 and −300 V, respectively. The films could completely return to their pristine state after the bias was removed and the ER effect could be fully reproduced. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2189197]

The phenomenon of colossal magnetoresistance (CMR) in perovskite manganite oxides has stirred considerable excitement in recent years due to the related physics and potential applications. However, the mechanism of the CMR effect is still not fully understood. It is generally agreed that the double-exchange interaction via spin-polarized conduction electrons is the main cause of the colossal magnetoresistance. High external magnetic fields align the magnetic moments of Mn ions, thereby enhancing the double-exchange transfer integral and increasing the electron conductance. Additionally, there are other superexchange-type interactions among the Mn ions that account for the antiferromagnetic or ferromagnetic insulating phases. Strong competitions among the coexisting interactions lead to a phase-separated ground state for the doped manganites in a wide range of hole concentration.[1] It has been suggested that the largest MR is associated with spatial inhomogeneity related to multiphase coexistence, which generally 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 the technological potentials for these kinds of materials. Recently an increasing interest has been shown in the so-called "colossal electroresistance" (CER) effect.[2–10] Namely, their electronic conductance may be controlled by an electric current or a static electric field. 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 the $\mathrm{La_{0.8}Ca_{0.2}MnO_3}$ single crystal has also been reported.[3] Recently we focused on the influence of electric current on the transport properties in thin films of mixed-valent manganites.[7–10] Our 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.[8–10] However, in the case that a large current is applied, local heating due to highly filamentary conduction inevitably occurs and makes the schemes complex. If using field effect configuration (FEC) to investigate the electroresistance (ER) effect, there will be no concern for local heating effect.

In previous studies, the FEC was fabricated mainly by the multilayer technique. An early investigation using a dielectric gate revealed interesting ER phenomena,[4] and this was followed by the demonstration of a large field effect using a ferroelectric gate.[5] In these studies, the channel layer was the first layer grown on a substrate, followed by the gate layer. Subsequently, a great ER effect was reported using an inverted multilayer configuration,[6] in which the ferroelectric/dielectric gate layer was the first layer grown on a substrate and then followed by the channel layer. One knows that a rigorous process is usually needed to fabricate multilayer structures with high quality. Films with lower quality often lead to various problems, such as a poor interface and severe interdiffusion, which may influence the intrinsic characteristics of the investigated targets. In this letter, we present the results of systematic investigations on the ER effect using a very simple FEC, which is formed on a single layer film. The manganite film acts as a channel and the substrate acts as a gate. Surprising results were achieved. The field effect is found to be significant. The ER changes the sign with the bias direction, but the temperature T_P at which the resistance peaks keeps nearly unchanged upon any bias in any directions. The films could completely return to their pristine state after removing the bias and the ER effect could be fully reproduced.

The present $\mathrm{La_{0.7}Ca_{0.3}MnO_3}$ (LCMO) and $\mathrm{La_{0.7}Ba_{0.3}MnO_3}$ (LBMO) thin films were grown on single crystal substrates of $\mathrm{LaAlO_3}$ (LAO) with (100) orientation using the pulsed laser deposition technique. Disks of stoichiometric $\mathrm{La_{0.7}}A_{0.3}\mathrm{MnO_3}$ (A=Ca, Ba) were used as the targets. The deposition took place in 1 mbar of pure oxygen. The energy of the laser beam was ~200 mJ with a wavelength of 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 the

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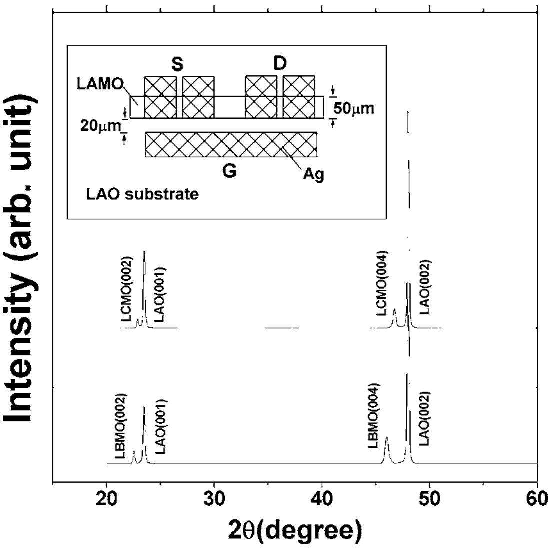

FIG. 1. X-ray diffraction spectra for the $La_{0.7}Ca_{0.3}MnO_3$ and $La_{0.7}Ba_{0.3}MnO_3$ epitaxial thin films grown on the $LaAlO_3$ substrates. The inset shows the top view of the device geometry.

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 the energy dispersive spectra (EDS) 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 (see Fig. 1). Besides the reflection from the LAO substrate and the (00l) peaks of the LCMO and LBMO films, no other peaks are visible, demonstrating that the grown films are of single phase. The LBMO lattice is bigger than that of the LCMO considering the radius of Ba^{2+} is larger than that of Ca^{2+} ions, which is also clearly indicated in Fig. 1. The resistance measurements were done by using the standard four-probe technique in a closed cycle cryostat. The Curie temperature T_C of the LCMO and LBMO films is determined to be ~278 and ~315 K, respectively, based on thermal magnetization measured under a low field of 100 Oe. In order to apply a static electric field to the film, a FEC was fabricated on a single layer film using the lithography technique, in which the LAMO film was used as a channel, and the LAO substrate as a gate. The inset in Fig. 1 shows the top view of the device geometry. The LAMO films grown on the LAO substrate were first patterned into long bridges with a width of 50 μm and then silver pads were evaporated on the bridge and the exposed LAO substrate, forming a FEC. The gate thickness is controlled by the positions of the evaporated silver pads. In this work, the gate thickness is 20 μm (see the inset of Fig. 1). Electrical leads were connected to the silver pads using a MEI-907 supersonic wire bonder to obtain low ohmic contacts. A voltage source with a limit of 300 V (Sorensen DCS 300 V−3.5 A) was employed to supply gate bias.

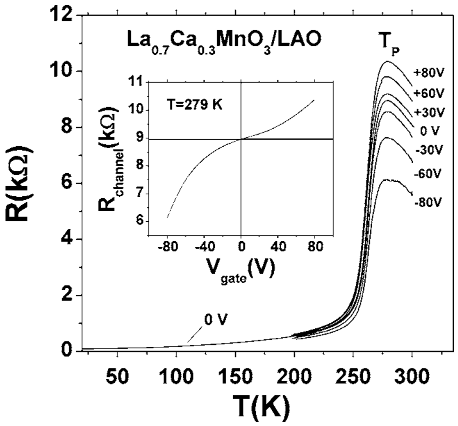

FIG. 2. Temperature dependent resistance of the $La_{0.7}Ca_{0.3}MnO_3$ channel for the field bias ranging from +80 to −80 V. The inset shows the channel resistance as a function of the gate voltage measured at 279 K.

The temperature dependent resistivity of a LCMO channel for a field bias ranging from −80 to +80 V is shown in Fig. 2. One can find that the field effect is significant. An important feature is that the ER effect changes its sign with the field direction. The resistance increases with a positive gate voltage but decreases with a negative gate voltage, which is consistent with the previous observations in using multilayer device configuration.[6] The similar results also indicate that we were measuring intrinsic properties of the LCMO films. The reduction of the resistance upon a bias of −80 V (-4×10^4 V/cm) reaches 32%. Such a magnitude of reduction is nearly the same as the resistive response observed in a magnetoresistance (MR) ratio under a magnetic field 2.5 T. Another attractive feature, which is indeed noteworthy, is that the position T_P of the insulating-metallic transition remains nearly unchanged upon applying any electric field in any direction. Such a characteristic is very different from the case of general MR. Generally, an application of magnetic field improves the spin alignment and affects the electric conduction in the sample. As a result, the resistive point T_P shifts to higher temperatures, yielding CMR. The inset in Fig. 2 presents the LCMO channel resistance as a function of the gate voltage measured at temperature T_P ~279 K. One can find the ER magnitude is asymmetric upon changing the bias direction. Also, nonlinear dependence and no saturation trend are shown for sweeping the gate voltage along both positive and negative directions. Our repeated measurements indicated that the films could completely return to their pristine state after the bias was removed, and the ER effect was fully reproducible.

We also investigated the ER effect for the LBMO channel using an identical FEC. Figure 3 presents the temperature dependent channel resistance upon bias of ±300 V for the LBMO channel from 10 to 318 K. (T_P is near 318 K. It is regretable that the resistance data at a temperature higher than 318 K cannot be measured due to the limitation of our apparatus.) The ER phenomena that appeared are very similar to the case of LCMO. The change of conductive resis-

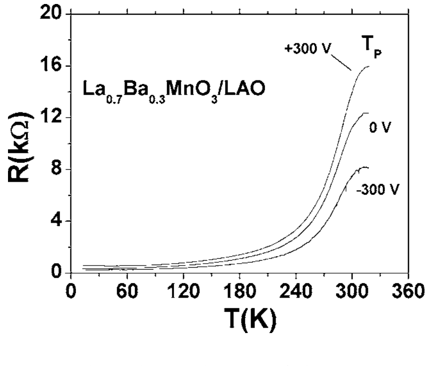

FIG. 3. Temperature dependent resistance of the $La_{0.7}Ba_{0.3}MnO_3$ channel under the field bias of ±300 V.

tance upon bias is also great for the LBMO channel, and the sign of the ER effect is also polarity dependent. The reduction of resistivity upon a bias of −300 V reaches 34%, which is nearly the same as that caused by −80 V in the LCMO channel. After the bias is removed, the LBMO channels could completely return to their initial state, and the ER effect could be fully reproduced. The ER effect of the LBMO upon the same bias is smaller than that of the LCMO, which is also consistent with the previous observations in using the multilayer device configuration.[6] However, in the previous studies,[6] the ER effect of the LBMO channel under bias does not show smooth dependence on temperature but fluctuates in the whole measured temperature range. Such fluctuations might be associated with the fabrications of multilayer structures. Using the present simple device configuration formed on a single layer film there are no such fluctuations at all for the LBMO channel.

Electric field effects have been intensively studied in high-T_C superconducting perovskites.[11–13] Electric fields modulate the conductive resistivity and considerably shift the superconducting transition temperature. The induced field effect is also dependent on the polarity of bias. All of these could be completely explained by field induced modulation of mobile carrier density.[13] However, in the case of the present manganites, the fact that an electric field causes no change for the peak position of resistivity seems not to support the charge transfer mechanism. If the carrier density in the channels was remarkably changed by electric fields, the Curie temperature T_C (the temperature of the peak resistance T_P is usually consistent with T_C in the $La_{0.7}Ca_{0.3}MnO_3$ and $La_{0.7}Ba_{0.3}MnO_3$ systems) should be changed. However, we did not detect an obvious shift of T_P. Furthermore, the fact that the nonlinear dependence of resistance for the sweeping gate voltage along both positive and negative directions is consistent with the previous results observed by using a multilayer configuration.[6] Such behavior (see the inset of Fig. 2) is also different from the linear dependence observed in the superconductor. Phase separation should be a key element for understanding the experimental observations. It has been a well established that metallic and insulating phases coexist in a broad range of phase spaces even for $La_{1-x}Ca_xMnO_3$.[14–16] In an inhomogeneous sample, part metallic and part insulating, an applied electric field may penetrate into the film and have a distribution in the phase space depending on the size, shape, and the distribution of the coexistent phases. When the electric field in the phase space is high enough and can be a fraction of crystal fields in the manganites, the Jahn-Teller distortion, which arises from crystal fields, is expected to be strongly impacted,[4] resulting in a significant change of the coexistent multiphase. It was supposed that the electric field in the phase space changes the relative volume fractions of the metallic and insulating phases by pushing the interface to move.[6] The field polarity controls the direction of interface movement. Applying a negative gate voltage pushes the interface into moving to insulating phase, leading to an increase of metallic fraction and thus a decrease of resistance. A positive voltage does the opposite, leading to an increase of resistance. After the bias is removed, the interface goes back to the initial position and the films return to the original state. The ER effect can be fully reproduced. For the present two systems LCMO and LBMO, the required bias is highly different to achieve the same magnitude of ER effect, which may be governed by the different characteristics of the coexistent phases in different systems. For a similar ER magnitude, a higher bias is needed for the LBMO channel compared to the LCMO channel. One knows that the details of crystal fields and Jahn-Teller distortion vary depending on local environments in different systems. It is understandable that an electric field, which is able to impact the distortion and thus move the interface of coexistent phases, should be different for the LCMO and the LBMO manganites.

In summary, the influence of the static electric field on the transport properties in $La_{0.7}A_{0.3}MnO_3$ (A=Ca, Ba) films was investigated by using a very simple FEC, which is formed on a single layer film. Such an easy manipulative technique avoids the possible problems that appeared in multilayer structures. Surprising results were achieved. The field effect was found to be significant. ER reaches 32% and 34% upon biases of −80 V (-4×10^4 V/cm) and −300 V (-1.5×10^5 V/cm) for the LCMO and LBMO channels, respectively. Furthermore, the sign of ER effect was found to be polarity dependent. Our primary analysis indicates that the modulation of the electric field on the coexistent multiphase is responsible for the observed ER effect. The present simple device configuration is easily applied to various materials. This may open a new avenue for exploring potential applications in fabricating field effect transistor based on epitaxial perovskite heterostructures.

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