Colossal electroresistance and low-field colossal magnetoresistance in the single-crystal bilayered manganite $La_{1.2}Sr_{1.8}Mn_2O_7$

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We report the observation of current-induced abrupt transition from a metallic to an insulating state in a bilayered manganite $La_{1.2}Sr_{1.8}Mn_2O_7$ single crystal. A colossal electroresistance effect $[\rho(50\text{ mA})-\rho(1\text{ mA})]/\rho(50\text{ mA})=93\%$ is found at 120 K. Moreover, the current-induced transition is very sensitive to external magnetic fields. At the measuring current of 50 mA a low-field colossal magnetoresistance effect, $[\rho(H)-\rho(0)]/\rho(0)=-86\%$, occurs at $H=500$ Oe. This low-field magnetoresistance effect can be repeatedly modulated by external magnetic field. We interpret these phenomena as the consequence of the percolative conduction and local Joule thermal effect.

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Perovskite manganites have drawn a lot of research attention since the discovery of the colossal magnetoresistance (CMR) effect,[1] i.e., the huge change of resistance induced by a moderate magnetic field. In spite of intensive efforts in this field, producing the CMR effect in low magnetic fields is still a challenging goal toward practical applications. Apart from the CMR effect, a variety of interesting physical phenomena have been observed in manganites. Recently, it was found that the resistivity of some perovskite manganites can experience a great change when the applied electrical current/voltage exceeds a critical value.[2–5] This electrical-field-induced resistance change is usually termed as the electroresistance (ER). The large ER effect in manganites was first discovered in the charge ordered $Pr_{0.7}Ca_{0.3}MnO_3$ crystal.[2] It was found that a large enough electrical current or voltage is able to switch the insulating charge-ordered state to a metallic ferromagnetic state, and consequently, causes a huge negative ER. Later, a different type of ER effect was observed in some phase-separated manganites that show percolative conduction with a first-order metal-insulator (M-I) transition.[6–8] Wu *et al.* first reported a colossal positive ER in $La_{0.7}Ca_{0.3}MnO_3$ and attributed it to the presence of electronic phase separation (PS).[6] More recently, Tokunaga *et al.* studied the ER effect in phase-separated $(La_{0.3}Pr_{0.7})_{0.7}Ca_{0.3}MnO_3$ and $Nd_{0.5}Ca_{0.5}Mn_{0.97}Cr_{0.03}O_3$ crystals.[7,8] By using a magneto-optical imaging technique to visualize the inhomogeneous magnetization and conduction paths they demonstrated that the application of a large amount of current can lead to the collapse of percolative conduction paths through a local Joule heating. As a result, a large amount of current drives the system from a metallic state to an insulating state, which causes a large positive ER.

At present the study of ER effect in manganites mainly focuses on the cubic ABO_3 system, but little attention has been put on the layered manganites. Bilayered manganites, such as $La_{2-2x}Sr_{1+2x}Mn_2O_7$, consist of bilayers of MnO_2 sheets separated by insulating $(La,Sr)_2-O_2$ blocks. The anisotropic structure leads to the quasi-two-dimensional characters in magnetic and transport properties.[9,10] Formation of mixed phases and occurrence of percolation in bilayered manganites, especially in $La_{1.2}Sr_{1.8}Mn_2O_7$ which has a nominal hole density of $x=0.4$, have been confirmed by many experiments.[11–14] The phase separation and percolative conduction in $La_{1.2}Sr_{1.8}Mn_2O_7$ also leads to a steep M-I transition. Therefore, it would be worthwhile to investigate the influence of electrical field on the transport and magnetotransport properties in this phase-separated bilayered manganite. In this work, we have performed such a study and discovered a colossal ER effect as well as a current-assisted CMR effect under quite low magnetic fields (~500 Oe) in a $La_{1.2}Sr_{1.8}Mn_2O_7$ crystal.

Single crystals of $La_{1.2}Sr_{1.8}Mn_2O_7$ were grown by the floating-zone method in an optical image four-mirror furnace. The composition of the crystal is checked by inductively coupled plasma atomic emission spectroscopy. X-ray diffraction measurement on powder shows no trace of any secondary phase. Back-reflection Laue X-ray diffraction method was carried out to determine the crystallographic direction. A rectangular piece of sample was cut to take magnetization and resistivity measurements. Transport measurements were performed using the standard four-probe method. The applied magnetic field and electrical current are parallel to each other, both in the *ab* plane of the crystal. During the measurement of resistivity, pulsed currents were applied in all the experiments. The thermometer is set close to the sample in order to monitor the sample's temperature precisely.

Figure 1(a) shows the in-plane magnetization and resistivity as a function of temperature of the crystal. Resistivity is measured at a constant dc pulsed current of 1 mA and magnetization is measured in an applied field of 1 kOe. A metal-insulator transition shows up around 126 K. The temperature dependence of in-plane resistivity (ρ-T) curve exhibits a small hysteresis in the M-I transition region, suggesting that the transition has some first-order-like characteristics. In order to further clarify the nature of the phase transition, we also measured the isothermal magnetization in the vicinity of the critical point and drew the isothermal Arrott plots of H/M versus M^2, as shown in Fig. 1(b). According to the Banerjee criterion,[15] a negative slope in a relatively small M^2

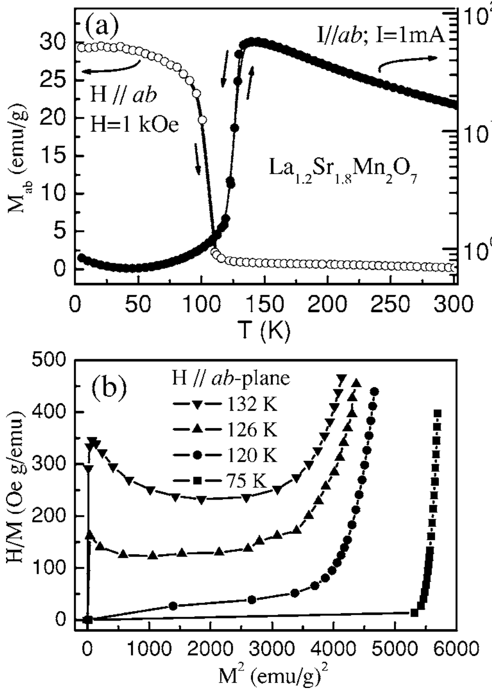

FIG. 1. (a) Temperature dependence of in-plane magnetization and resistivity in heating or cooling process of $La_{1.2}Sr_{1.8}Mn_2O_7$ single crystal. (b) The Arrott plots in the vicinity of critical points of the metal-insulator transition.

region at 126 and 132 K indicates a first-order character of phase transition in $La_{1.2}Sr_{1.8}Mn_2O_7$ single crystal.

Figure 2 shows the current dependence of resistivity (ρ-I curve) at various temperatures from 5 to 130 K. Below 100 K, the resistivity changes gently with increasing current and it always retains a low value as a metallic state. However, in the M-I transition region ($110\ \mathrm{K} < T < 130\ \mathrm{K}$), the ρ-I curves exhibit a peculiar behavior. With increasing current, the resistivity exhibits an abrupt switch from the low-resistive state to a high-resistive state at a threshold current (I_{th}^{up}) of 80, 45, and 10 mA at temperatures of 115, 120, and 125 K, respectively. Above these threshold currents, the resistivity remains a high value with further increasing current up to 100 mA. The successive decrease in current causes a change to the low-resistive state again but at a lower threshold current (I_{th}^{down}), i.e., the ρ-I curve shows hysteresis. A subsequent sweep to negative currents produces an antisymmetric ρ-I curve with respect to the origin. This fact confirms the reproducibility of the ρ-I characteristics. These results demonstrate a current-induced M-I transition in $La_{1.2}Sr_{1.8}Mn_2O_7$. At 130 K which is above the M-I transition temperature, there is no current-induced switch in resistivity. Based on the data in Fig. 2, we obtained the ER ratio, defined as $ER = [\rho(I) - \rho(I_0)]/\rho(I) \times 100\%$ with $I_0 = 1$ mA, as shown in the inset of Fig. 2. The maximum of ER ratio is as high as 93% and 95% at 120 and 115 K, respectively. Such a colossal positive ER effect was rarely reported before.

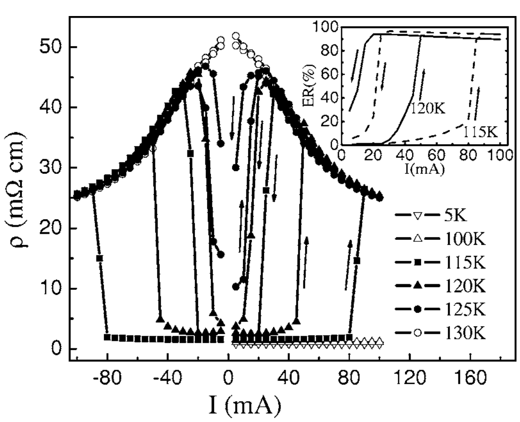

FIG. 2. ρ-I curves of $La_{1.2}Sr_{1.8}Mn_2O_7$ at various temperatures. The inset: ER vs current at 115 and 120 K.

In order to correctly understand the current-induced M-I transition in $La_{1.2}Sr_{1.8}Mn_2O_7$, we first tested the possibility of self-heating effect due to applied currents. According to the first law of thermodynamics, the thermal balance of the sample can be expressed as

$$W - Q = -\Delta U = nC_p\Delta T, \tag{1}$$

where ΔU is the change of inner energy of the sample, W is the Joule heat caused by applied current, Q is the thermal energy conducting from the sample to the chamber, C_p is the specific heat, and n is the mole number of the sample. Assuming that there is no thermal exchange between the sample and the chamber, Eq. (1) reduces to:

$$W = nC_p\Delta T. \tag{2}$$

The specific heat of $La_{1.2}Sr_{1.8}Mn_2O_7$ single crystal at 120 K is about $150\ \mathrm{J\ mol^{-1}\ K^{-1}}$.[16] The Joule heat, $W = \sum_{i=0}^{m} I_i V_i t_i$, where I_i is the applied current, V_i is the measured voltage, t_i is the width (~ 0.02 s) of the current pulse, and m is the number of current pulse, can be calculated using our experimental data. The result is $W = 0.24$ mJ when the applied current reaches the threshold current 50 mA. For a 4.4 mg sample used in the experiments, this heating would cause a raise of the sample's temperature for 0.2 K, i.e., from 120 to 120.2 K. If considering the thermal exchange between the sample and the chamber, the actual temperature increase should be smaller. Since the M-I transition temperature is 126 K, the Joule heating by pulsed currents should not be able to heat the whole sample to pass the M-I transition point. Therefore, the current-induced M-I transition is not due to self-heating effect.

We argue that this phenomenon should be attributed to the percolative conduction in the phase-separated state in $La_{1.2}Sr_{1.8}Mn_2O_7$. The role of applied pulsed currents is to heat locally the percolative path rather than the whole sample. In the M-I transition region, there are two competing states: FM metallic state and PM insulating state. The insulating regions have a high resistance and the transport current should be concentrated to metallic channels. The application of a large current leads to local heating of the metallic channels and drives some regions from metallic to insulating state. The closure of part of metallic channels forces the current flow into the remaining channels, and thus accelerates local heating to close the remaining metallic channels. As all the percolative conduction paths are closed, the crystal becomes insulating immediately.

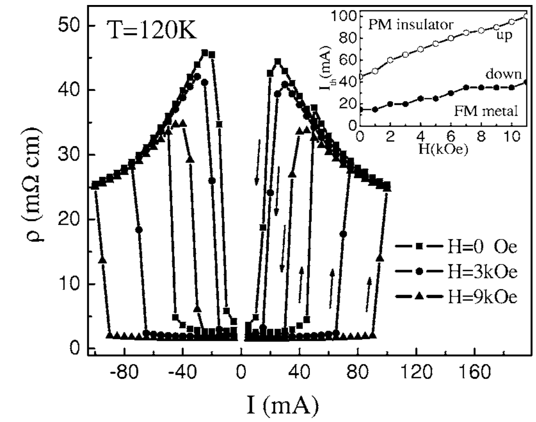

FIG. 3. ρ-I curves at 120 K in different magnetic fields of $La_{1.2}Sr_{1.8}Mn_2O_7$. The inset shows the field dependence of the threshold current with increasing current (up) and decreasing current (down) process.

We then studied the influence of magnetic field on this current-induced resistive transition. Figure 3 shows the ρ-I curves in different magnetic fields at 120 K. It is apparent that the current-induced switch of resistive states is very sensitive to external magnetic fields. With the increase of external magnetic field, the switch from the low resistive state to the high resistive state occurs at higher threshold currents. The inset shows the magnetic field dependence of the threshold current (I_{th}) in both the increasing current (up) and decreasing current (down) processes. I_{th} increases monotonically with increasing magnetic field. These results suggest that in a magnetic field more currents are required to switch the resistive state. This is consistent with the growth of FM metallic regions with increasing magnetic field.

The shift of resistive state switch in external magnetic field leads to a low-field CMR effect. Figure 4 shows the resistivity as a function of magnetic field at various currents at 120 K. When the current is small ($I \leqslant 40$ mA), the system is still in the metallic state and the resistivity changes slightly with magnetic field. When the current is more than 50 mA, the system has been switched into the insulating state in zero

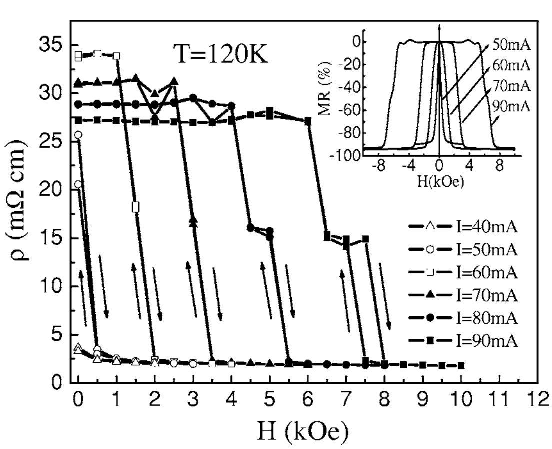

FIG. 4. Magnetic-field dependence of resistivity measured with various currents at 120 K. The inset shows MR as a function of magnetic field at 120 K.

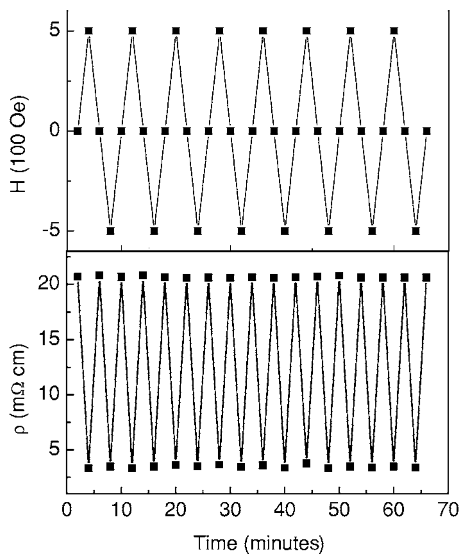

FIG. 5. Repeatable modulation of resistivity by external magnetic field. Resistivity is measured at 50 mA current.

magnetic field. However, as the magnetic field increases and exceeds a critical value, the resistivity suddenly switches back to the low-resistive state because I_{th} in this field has exceeded the applied current. When the magnetic field decreases, the resistivity returns to the high resistive state almost at the same critical fields. The switch from high-resistive state to low-resistive one induced by a small magnetic field results in a low-field CMR effect. The inset of Fig. 4 plots the magnetic field dependence of the MR ratio at 120 K. Here, MR is defined as $MR=[\rho(H)-\rho(H=0)]/\rho(H=0)\times 100\%$. The MR ratio is as high as -86% in 500 Oe field at 50 mA current, and -93% in 2000 Oe at 60 mA current. The CMR effect in such low magnetic fields is striking and could benefit practical applications.

This current-assisted low-field CMR in $La_{1.2}Sr_{1.8}Mn_2O_7$ reflects the competing role played by electrical current and magnetic field. While a large amount of electrical current can drive the metallic regions into insulating states through local Joule heating and consequently block the percolative conduction paths, a magnetic field, on the other hand, promotes the growth of FM metallic regions. At the vicinity of the percolation threshold, there must be a high sensitivity to external magnetic field since a tiny change in the metallic fraction induced by a small magnetic field could connect the conduction paths and lead to a large change in conductivity.

It is well-known that a low-field CMR is crucial for many applications in magnetic recording and magnetic sensors. The current-assisted low field CMR in $La_{1.2}Sr_{1.8}Mn_2O_7$ crystal could open a new way to this end. In practical applications, the low field CMR response should be repeatable without any decay. We have examined the CMR response with altering magnetic field. Figure 5 shows the repeatable modulation of the resistivity by periodical change of magnetic field. In zero magnetic field, the crystal is in the high-resistive state ($\sim$20 m$\Omega\cdot$cm) at a 50 mA measuring current. When a 500 Oe magnetic field is applied, the resistivity becomes a low-resistive state ($\sim$3 m$\Omega\cdot$cm). When the magnetic field is removed, the resistivity restores the high-resistive state. When a negative 500 Oe fields is applied, the same effect happens. Therefore, the switch between the high-resistive and low-resistive state, i.e., the CMR response, can be repeatedly modulated by choosing the optimal current and magnetic field.

In summary, we have observed current-induced abrupt switch of resistivity from metallic to insulating states in a bilayed $La_{1.2}Sr_{1.8}Mn_2O_7$ crystal. This abrupt change leads to a CER effect. Meanwhile, a small magnetic field can shift the current-induced transition and leads to a low-field CMR effect. These effects are discussed in terms of the adjustment of percolative conduction by electrical current and magnetic field. This current-assisted low-field CMR is constantly repeatable and thus could benefit applications in magnetic sensors and magnetic recording.

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