Electroresistance and current-induced metastable states in the thin film of half-doped manganite $Pr_{0.5}Sr_{0.5}MnO_3$

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Electroresistance (ER) effects and current-induced metastable states in the thin film of $Pr_{0.5}Sr_{0.5}MnO_3$ were investigated. The initial state was insensitive to weak currents and susceptible to high-density currents. As the current density reached a certain value, metastable states, which were very sensitive to weak currents at low temperatures, were excited. It was found that as the excited current increased, the induced metastable state showed a larger electroresistance in a wider temperature range. Interfacial effects related to electrodes could be ruled out. The observed effects might be related to the coexistence and instability of the multiphases in manganites. © *2013 AIP Publishing LLC* [http://dx.doi.org/10.1063/1.4800841]

Manganites with perovskite-like structures belong to the family of strongly correlated systems. They came to the eyes of physicists as early as 1950s and have been a continuing focus since the early 1990s,[1–5] when the unexpected sensitivity of resistance to applied magnetic fields, namely "colossal magnetoresistance (CMR)," was discovered. In these materials, the complex couplings between different degrees of freedoms (spin, charge, orbital, and lattice) give rise to rich electronic phases and many tantalizing electromagnetic properties. It is found that these various phases in manganites have close energies and may coexist in a single chemical phase. The intricate balance between competing phases is readily susceptible to external stimuli,[6–15] which can cause drastic effects. This feature actually not only offers an arena to understand the physics of strong correlation but also provides an opportunity to develop practical devices.

Electric fields/currents are frequently used to tune the properties of manganites. An early demonstration of electric fields/currents effects in manganites was reported in single crystals of $Pr_{0.7}Ca_{0.3}MnO_3$ by Asamitsu *et al.*[6] They showed that the electrical insulating charge-ordered state was melt by the electric fields/currents, which lead to a resistance drop of several orders. Currents of high densities can also excite highly resistive metastable states, which shows pronounced electroresistance (ER), from ferromagnetic metallic states.[7] More intriguingly, asymmetric conduction, resistance steps, negative differential resistance, and low-temperature persistent conductivity can also be induced by electric fields/currents.[8–10] Accumulating studies pointed out that phase separation/competition is a crucial ingredient for understanding these unusual phenomena. Half-doped manganite $Pr_{0.5}Sr_{0.5}MnO_3$ (PSMO) is a typical example of phase competition.[16–19] Upon cooling, it exhibits a high-temperature paramagnetic insulator-ferromagnetic metal transition and a low-temperature ferromagnetic metal-antiferromagnetic insulator transition. There have been studies on how PSMO responses to magnetic field, hydrostatic pressure, and light.[16,20,21] However, no investigation on current-induced effects in this interesting material has been carried out. In this paper, we report electroresistance and current-induced metastable states in the thin film of PSMO.

The thin film of PSMO was grown on (001)-oriented $SrTiO_3$ (STO) by using pulsed laser ablation.[22] The deposition was carried out in flowing oxygen and the pressure of chamber was kept at 30 Pa. Throughout the growth, the substrate holder was maintained at $\sim$800 °C, as monitored by the k-type thermal couple. To avoid possible oxygen deficiency, the sample was annealed subsequent to deposition in 0.5 bar oxygen at grown temperature for 30 min. As determined with a Dektak stylus profiler, the film thickness is $\sim$60 nm. Microbridges (see the inset of Fig. 1) were made by photolithography. The ohmic contacts were achieved by using silver electrodes, which were deposited through thermal evaporation. The transport properties were measured in a four-probe configuration. The magnetization was measured in a field of 20 Oe parallel to film surface by a Superconducting Quantum Interference Device (SQUID).

The structural properties were assessed by x-ray diffraction (XRD). Figure 1 displays the XRD pattern (θ-2θ scan) for PSMO on STO. Only reflection peaks from (00l) planes of PSMO (for PSMO, pseudocubic index is used) and STO are visible, indicating the grown layer is c-axis oriented. The Φ scans of the (113) reflection (see Ref. 22) demonstrate the cubic-on-cubic epitaxy. The out-of-plane lattice constant of the PSMO film, calculated from the reflection peak, is $c \sim 3.77$ Å. For bulk PSMO, the pseudocubic lattice constant a_p is $\sim$3.84 Å. The reduced value of out-of-plane lattice constant suggests the compressive out-of-plane strain $[\varepsilon_{zz} = (c - a_p)/a_p = -1.8\%]$. The relation between the in-plane strain ε_{xx} and out-of-plane strain ε_{zz} is $\varepsilon_{xx} = -\varepsilon_{zz}(1-\nu)/(2\nu)$, where ν is the Poisson ratio. Typical reported ν for manganites ranges from 0.3 to 0.5. Thus, a negative ε_{zz} (compressive strain) means a positive ε_{xx} (tensile strain).

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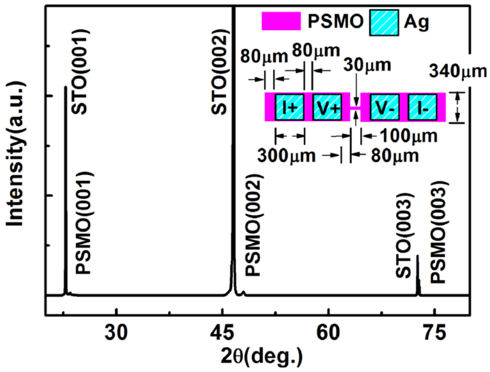

FIG. 1. XRD pattern (θ-2θ scan) of PSMO/STO. Shown in the inset is the layout of the microbridge and the four-probe configuration used to measure all R-T curves in Figs. 3 and 4.

Shown in Fig. 2 are the temperature (T) dependences of resistance (R) and magnetization (M) for our PSMO film. Upon cooling, the conduction first changes from insulating/semiconducting behavior to metallic one at $T_P \sim 263$ K. Further lowering the temperature leads to a second transition at $T_{MIN} \sim 208$ K, from metallic conduction to insulating conduction. There are also two transitions in the M-T curve when the temperature decreases. Around T_P, there is a steep increase in magnetization. At $T \sim 150$ K, magnetization begins to decrease with decreasing temperature very slowly. Similar features were also reported by Wagner *et al.*[23] In single crystalline PSMO, there is a structural change accompanying the FMM-AFI transition.[17] The structures are tetragonal and monoclinic in the FM and AFI states,[18] respectively. When PSMO is deposited on STO, the substrate-imposed tetragonal symmetry makes such a structural transition difficult.[22] The appearance of low-temperature metal-insulator transition in thin PSMO film on STO may be owing to the enhanced Jahn-Teller distortion as a result of large strain.[22]

To study the electric current-induced effects, the samples were patterned to microbridges by using standard photolithography technique. R-T curves for all microbridges are essentially the same, with slight fluctuation from one bridge to another. This indicates that our film is uniform. Figure 3(a) displays the typical temperature dependence of resistance for patterned PSMO measured with currents ranging from 1 to 2000 μA. Changing the current from 1 μA to 50 μA has no appreciable influence on the R-T curve. In other words, the as-prepared PSMO film is not sensitive to low-

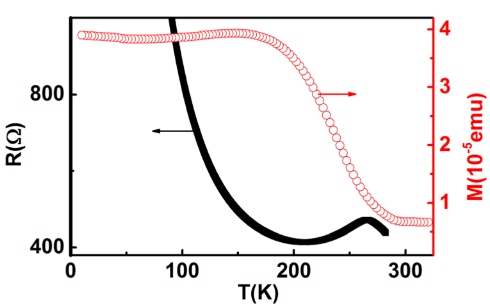

FIG. 2. Temperature dependences of resistance (left) and magnetization (right) for the PSMO film. The applied field of the dc magnetic measurement is 20 Oe.

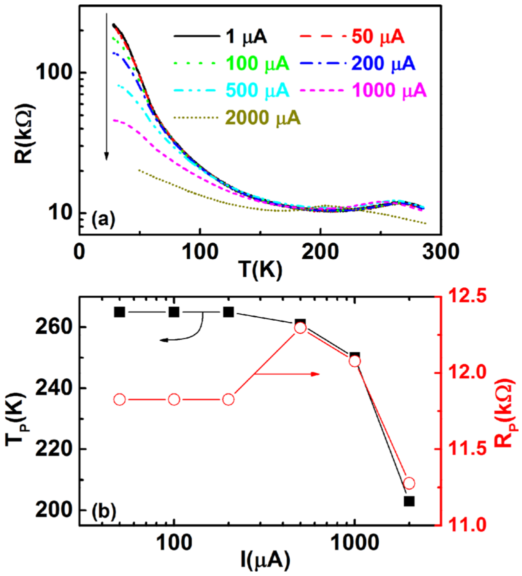

FIG. 3. (a) Temperature dependences of resistance measured with different currents (I = 1, 50, 100, 200, 500, 1000, and 2000 μA) for the initial state; (b) T_P and R_P as a function of bias current I.

density current. As the current increases from 50 to 200 μA, while high-temperature resistance keeps unchanged, the low-temperature resistance is suppressed substantially. When the current is further increased, the reduction of low-temperature resistance becomes significant. At the same time, there are considerable changes at high-temperature regions. The evolutions of T_P and peak resistance R_P are summarized in Fig. 3(b). As the current reaches a certain value, T_P becomes lower with the increase of current density. This is similar to previous reports.[24] Such a phenomenon is probably related to the self Joule heating, which can cause a temperature gradient between sample surface and sample holder. For the current induced suppression of low-temperature resistance, Joule heating should have a non-ignorable contribution. Nevertheless, such a heating effect cannot influence the value of peak resistance R_P.[24] It should be emphasized that after each large current, R-T curve was re-measured with a bias current of 1 μA. Even after the application of 2000 μA, there was no observable change. Thus, the current dependence of R_P should be an intrinsic property of PSMO and not related to the current excited states.[7,9] In hole doped $La_{0.7}Ca_{0.3}MnO_3$ and $La_{0.85}Ba_{0.15}MnO_3$, it was found that R_P decreased monotonously with the increase of current density. For our PSMO film, with the increase of current density, R_P first keeps unchanged, then increases, and finally decreases. The origin of the unusual relation between R_P and I is not very clear.

For PSMO, metastable states could also be excited by large currents. Current processing [electrodes I+ and I− were connected to the current source] was conducted at room temperature and the duration was fixed to be 20 min. After each processing, a R-T curve was measured under a bias current of 1 μA. Detectable changes appeared as the processing current became larger than ~4 mA. Shown in Fig. 4(a) are the metastable states induced by different currents. After current processing, the resistance becomes

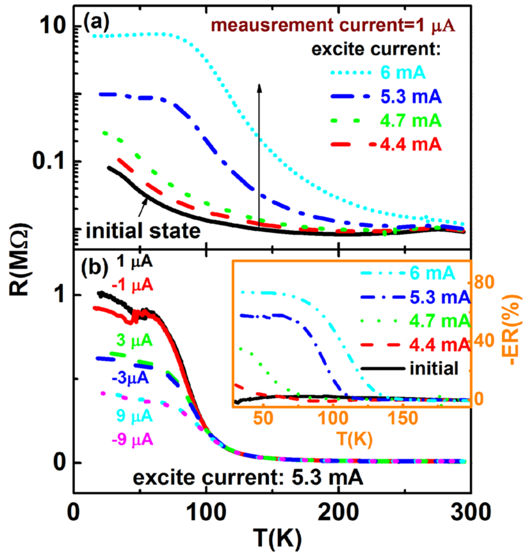

FIG. 4. (a) Metastable states induced by currents of different magnitudes. The measured current is fixed to be 1 μA; (b) temperature dependences of resistance measured with different currents (± 1, ± 3, and $\pm 9\,\mu$A) for the state induced by a current of 5.3 μA. The inset summarizes the temperature of $ER = 100\% \times [R(9\,\mu A) - R(1\,\mu A)]/R(1\,\mu A)$ for metastable states induced by different currents.

higher in the whole temperature range. The increase of resistance is much larger at low temperatures. It is found that the larger the excite current, the more dramatic the resistance change. With the increase of excite current, the temperature range of metallic state becomes smaller. As the processing current reaches 6 mA, the excited state is insulating in the whole temperature range. To study the properties of these current-induced metastable states, R-T curves were measured under small currents of different polarities [see Fig. 4(b)]. The magnitude of resistance is essentially independent of current direction. At low temperatures, the resistance is greatly reduced by weak currents. With the definition $ER = 100\% \times [R(9\,\mu A) - R(1\,\mu A)]/R(1\,\mu A)$, temperature dependences of ER for different metastable states were calculated and illustrated in the inset of Fig. 4(b). For each state, ER is negligible at high temperatures and becomes significant at low temperatures. As the excited current increases, the induced metastable state shows a larger ER in a wider temperature range.

The induced highly resistive metastable states are intrinsic properties rather than interfacial effects.[7–10] There were no substantial changes in the resistance measured between electrodes I+ and V+ (also between V− and I−, see the inset of Fig. 1) before and after current processing. The conduction between I+ and V+ (or V− and I−) kept linear and symmetric in the whole temperature range. Thus, the main changes should took place in the narrow part ($30\,\mu m \times 100\,\mu m$) of the bridge, where the current density was one order larger than that between I+ and V+ (and between V− and I−). Current-induced metastable states were observed in other manganite films, such as $Pr_{0.7}Sr_{0.3}MnO_3$,[10] $Nd_{0.7}Sr_{0.3}MnO_3$,[8] and $La_{1-x}Ca_xMnO_3$ ($x = 0.2$ and 0.3).[7] To understand such metastable states and the enhanced sensitivity to weak currents, it would be helpful to note that manganites are electronically soft.[4] There are cross couplings between different degrees of freedom (spin, charge, lattice, and orbit), which give rise to diverse electronic ground states with very close free energy. Application of large electric fields/currents may drive it from one state to another in local areas, shifting the balance of competing phases. It is known that phase competition is at the heart of many intriguing properties of manganites. A shifted balance may induce new properties, such as the enhanced sensitivity to weak currents. To get beyond this qualitative schematic picture and discover the exact mechanism, more further theoretical and experimental efforts should be made.

Current-induced effects in microbridges of PSMO were studied. Upon cooling, PSMO exhibited a high-temperature insulator-metal transition and a low-temperature metal-insulator transition. As-prepared film had a weak sensitivity to low-density currents. With the increase of current density, the low-temperature resistance was reduced significantly. The peak resistance showed an unusual dependence on current density. Metastable states were excited by currents with higher densities. With the increase of excite current, the metastable state had a larger resistance and an enhanced electroresistance. The observed phenomena were discussed in a phase separation/competition scheme.

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