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Dependence of negative differential resistance on electronic phase separation in unpatterned manganite films

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Here, we report that small but well-defined negative differential resistance (NDR) steps can be observed at critical voltages in unpatterned millimeter-scale manganite films of $\text{Pr}_{0.65}(\text{Ca}_{0.75}\text{Sr}_{0.25})_{0.35}\text{MnO}_3$. We systematically investigate the magneto-transport properties of strained thin films grown on LaAlO_3 and SrTiO_3 substrates and map out their phase diagrams which show temperature- and magnetic-field-dependent electronic phase separation (EPS). Our data suggest that the onset of NDR only occurs “deep” within the regions of EPS, underscoring the subtle nature of filamentary transport in manganite thin films with competing phases. © 2012 American Institute of Physics. [doi:10.1063/1.3684806]

In some mixed-valence manganites, the coexistence of multiple magnetic phases, a phenomenon known as electronic phase separation (EPS), leads to the discovery of fascinating physics related to complex energy landscape.^{1–5} Direct microscopy evidences have been achieved, showing mesoscopic ferromagnetic (FM) clusters inside an antiferromagnetic (AFM) matrix.^{4,6} Importantly, the percolative transport as a result of the coexisting charge-ordered insulator (COI) and the ferromagnetic metal (FMM) phases amplifies the magnetic field effect, giving rise to colossal magnetoresistance (CMR).³ Furthermore, quenched disorder and inhomogeneous strain distribution can cause micrometer-scale EPS.^{7,8} Earlier studies have reported that the external factors like temperature, magnetic field, and substrate strain can modulate the fraction, shape, and stability of the “soft” electronic phases, leading to rich physics.^{9–12}

One of the most notable transport phenomena in phase-separated manganites is the abrupt resistance steps at threshold electric fields, which has stimulated lots of interest.^{13–21} These observations can be categorized into two groups: in one group, the sample resistance abruptly decreases under a critical electric bias, which can be attributed to the electric field-induced melting of COI, similar to the CMR effect.^{13–15} In the other group, rupture of filamentary conducting channels as a result of local metal-to-insulator transition gives rise to the effect of abrupt resistance increase at critical voltages, i.e., negative differential resistance (NDR).^{17,18,20,21} Since the conducting filaments have nano or micro scale dimensions, the clear observation of abrupt resistance steps is often believed to entail the lithographic patterning of bridge-like microscale (nanoscale) structures. However, such processing steps may affect the characteristics of EPS as a result of chemical contamination and the rough edges of patterned structures. Furthermore, the heating effect may be more severe and localized in the narrow bridges, causing spurious effects. Finally, the investigated manganite films in previous experiments were deposited on a single type of sub-

strate, which is insufficient to reveal the complex physics related to the strain-dependent EPS.

Here, we show that NDR steps can be observed even in unpatterned $\text{Pr}_{0.65}(\text{Ca}_{0.75}\text{Sr}_{0.25})_{0.35}\text{MnO}_3$ (PCSMO) films with a macroscopic (millimeter) size. This helps to reduce the heating effect compared to the patterned structures, which is further confirmed by examining the effect of measurement speed. Our systematic studies of strained films deposited on different substrates indicate that the onset of NDR steps is closely related to the percolative transport in manganite films. Interestingly, we found that NDR steps do not show up across the whole EPS region, but only within the “deep” part instead, suggesting the intricate role of filamentary conduction.

PCSMO films were grown on (001) SrTiO_3 (STO, cubic with $a = 3.905 \text{ \AA}$) and LaAlO_3 (LAO, pseudocubic with $a = 3.793 \text{ \AA}$) substrates using pulsed laser deposition. The area of the films is $5 \times 5 \text{ mm}^2$ and the thickness is about 200 nm. The substrate was held at 750°C during growth followed by annealing at the same temperature for 60 min and then slowly cooled to room temperature. The oxygen pressure was 20 Pa during deposition and 1000 Pa during annealing and cooling. The current versus voltage (I - V) curves were collected using a pulsed (0.1 s) voltage mode with 5 s interval to avoid any heating effect. In the magnetization and transport measurements, the magnetic fields were applied along the sample plane.

The x-ray diffraction (XRD) patterns of the PCSMO films on LAO and STO substrates in Figs. 1(a) and 1(b) show good crystallinity and c -axis orientation. Reciprocal space mapping (RSM) data around the (033) reflections are shown in Figs. 1(c) and 1(d), and the extracted lattice parameters are $a_{S1} = 3.79 \text{ \AA}$, $c_{S1} = 3.92 \text{ \AA}$ and $a_{S2} = 3.91 \text{ \AA}$, $c_{S2} = 3.80 \text{ \AA}$, for the PCSMO films on LAO and STO substrates, respectively. The corresponding unit cell volumes are $V_{S1} = 56.3 \text{ \AA}^3$ for PCSMO/LAO and $V_{S2} = 58.1 \text{ \AA}^3$ for PCSMO/STO. Since the lattice parameters of bulk orthorhombic ($Pnma$) PCSMO are $a = 3.851 \text{ \AA}$, $b = 3.840 \text{ \AA}$, and $c = 3.848 \text{ \AA}$, the LAO and the STO substrate supply coherent in-plane compressive and tensile strain, respectively.

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FIG. 1. (Color online) XRD θ - 2θ spectra measured on (a) the PCSMO/LAO and (b) the PCSMO/STO samples. The corresponding RSM data collected around the 033 reflections are shown in (c) and (d), respectively.

Let us focus first on the PCSMO film grown on LAO. Figure 2(a) shows the I - V curves taken under a 6 T magnetic field at various temperatures from 20 to 150 K. At 20 K, the I - V curve shows a step-like decrease in the current at a threshold voltage (V_C) of 18.5 V. Although the magnitude of the NDR step is small, its existence is unambiguous. The threshold voltage monotonously reduces as the measurement temperature increases, and more than one NDR steps were often observed during a single voltage scan. However, the NDR step disappears at temperatures above 50 K. In Fig. 2(b), for the I - V curves under different magnetic fields at 30 K, NDR step first appears at 5 T and then V_C decreases with increasing magnetic fields. The NDR step vanishes for mag-

netic fields above 8 T. Overall, the NDR steps in the I - V curves have a systematic dependence on temperature and magnetic field.

We can draw some immediate conclusions based on the above observations. First, the NDR steps observed here in unpatterned PCSMO films are much smaller than those in previously reported patterned microscale devices,^{10,17,18,20} which is apparently the result of the large sample size. Nevertheless, our observation of NDR in lithography-free samples significantly simplifies the experimental procedures and motivates future efforts. Moreover, the current density at the NDR step in our sample (~ 2 A/mm²) is four orders of magnitude smaller than the values in previously reported

FIG. 2. (Color online) I - V curves of the PCSMO/LAO samples measured at varied conditions of (a) temperature, (b) magnetic field, and (c) time interval between voltage pulses. The solid arrows mark the NDR steps. The dashed arrows in (c) indicate the directions of voltage sweeping. (d) Temperature dependencies of the magnetization at 0.5 T and resistance under magnetic fields varying from 0 T to 12 T with 3 T steps for the same sample. (e) Magnetic field dependencies of the resistance measured at different temperatures. (f) Corresponding phase diagram of the same sample. The solid squares are extracted from (e), and the hollow triangles are from the RT curves under different magnetic fields in (d). The circles indicate where I - V curves were collected, among which the solid ones represent the field and temperature conditions where the NDR steps were observed.

patterned films.^{10,17} The smaller current density in the unpatterned PCSMO films implies much weaker Joule heating and self-induced magnetic field. On the other hand, the much larger current density used to trigger the onset of NDR steps in patterned manganite bridges seems to suggest stronger “pinning” of the conducting filaments, which may be a result of the artifact of edges roughness or contamination as a result of lithography and chemical processing.

Second, the NDR steps observed here cannot be explained by the electric-field-induced melting of COI or depinning of charge density waves which presents an irreversible step-like decrease of the resistance.^{13–15,22} Instead, the abrupt increase of resistance at the NDR steps in Fig. 2 suggests a transition from the FMM to the COI phase.

Finally, the NDR steps in PCSMO/LAO cannot be due to the current driven Joule heating effect which causes an increase of the whole sample temperature and triggers a metal-insulator transition.²³ In our experiments, only pulsed voltage was used, and a thermal couple directly in contact with the film surface did not detect any significant temperature change during the I - V measurements. To confirm this, we studied the influence of interval time on the characteristics of I - V curves, and the data are shown in Fig. 2(c). With the shortest interval time of 0.5 s between voltage pulses, the I - V curve shows a convex profile with obvious hysteresis, indicating a nonlinear effect of Joule heating. As the interval time increases, the thermal dissipation catches up with the Joule heating, and the I - V curves become more linear, but the NDR steps are retained, which suggests that they cannot be attributed to the Joule heating effect.

To explore the origin of NDR steps in Figs. 2(a) and 2(b), we systematically studied the magneto-transport properties of the PCSMO/LAO sample. Figure 2(d) shows a typical zero-field cooled (ZFC) and the field cooled (FC) magnetization versus temperature curves measured under a magnetic field of 0.5 T. The upturn of the magnetization

below 175 K indicates the growth of the FM phase. The RT curve in Fig. 2(d) for $H = 0$ shows no insulator-metal transition within the instrumental limit, while a resistance peak appears at 83 K under 3 T magnetic field and shifts to higher temperature with increasing magnetic field. The thermal hysteresis in the RT curve is a characteristic of EPS in the vicinity of first-order phase transition.^{24,25} In Fig. 2(e), the field dependent resistance at different temperatures was measured to establish the phase diagram. Each curve was collected after zero-field-cooling the sample from 300 K to the predetermined temperature. Based on these data, we mapped out the phase diagram in Fig. 2(f).²⁶ The temperature and field values where the I - V curves were measured are marked as circles, while solid circles represent the places where NDR steps were observed. It is noteworthy that the NDR steps appear only “deep” within the region of EPS, suggesting that EPS is the prerequisite but not sufficient for their onset.

We also studied the transport properties of PCSMO film grown on STO substrate. Comparing to the insulating behaviour of PCSMO/LAO in Fig. 2(e), the RT curve of PCSMO/STO in Fig. 3(b) shows a insulator-metal transition at 90 K and an obvious thermal hysteresis under zero magnetic field, which suggests a pronounced EPS. The temperature dependence of magnetization collected under a 0.5 T magnetic field indicates the onset of FM phase at about 160 K. Coinciding with the EPS suggested by the RT curve under zero magnetic field, multiple NDR steps were observed in the I - V curves below 60 K in Fig. 3(a). Similar to the LAO case, the phase diagram for the PCSMO/STO sample (Fig. 3(d)) derived from the RT curves in Fig. 3(b) and the relative humidity (RH) curves in Fig. 3(c) further confirms the relationship between EPS and NDR steps. Furthermore, we can see that at low temperatures, a magnetic field is more effective to induce the FM phase in PCSMO/STO than in PCSMO/LAO, although the magnitudes of strain are almost the same: 1.584% compressive in PCSMO/LAO and 1.532% tensile in

FIG. 3. (Color online) (a) I - V curves at different temperatures for the same sample without external magnetic field. The arrows indicate the direction of the temperature sweep. (b) Temperature dependencies of the magnetization at 0.5 T and resistance under magnetic fields varying from 0 T to 12 T with 3 T steps of PCSMO/STO. (c) Magnetic field dependencies of the resistance measured at different temperatures. (d) Corresponding phase diagram of the same sample. The solid squares are extracted from (c), and the hollow triangles are from the RT curves under different magnetic fields in (b). The circles indicate where I - V curves were collected, among which the solid ones represent the field and temperature conditions where the NDR steps were observed.

PCSMO/STO. Nevertheless, the onset of NDR steps can only occur after the conducting filaments in the PCSMO films develop into a “mature” state in the regions of EPS, and the associated local phase transition is responsible for the NDR steps.

In summary, we found that the I - V curves of unpatterned PCSMO films can show NDR steps which were previously reported only in the mesoscopic structures. These small but sharp steps are uncommon in electronic materials and represent the most drastic nonlinear characteristics of percolative transport in a strongly correlated electron system. The dependence of NDR steps on temperature and magnetic field in PCSMO films deposited on two different substrates suggests that EPS is the prerequisite but not sufficient for the onset of NDR steps, which put constraint on the theoretical models.

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