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Current-induced anisotropic memory effect in $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ (y = 0.43) thin film

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Current-induced anisotropic memory effect in the $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ (y = 0.43) film has been investigated. It is found that large electrical currents result in a dramatic reduction in film resistivity, and only slightly rebounds after the removal of the current, giving rise to a memory effect. Even more, the change of resistivity, measured under small current after removing processing current, is considerably faster along the direction parallel than perpendicular to processing current, yielding an anisotropic memory effect. According to the dielectrophoresis model, conduction filaments in the two directions could be different, and they may be easily formed along the processing current, thus the change of resistivity is different in the two directions. The anisotropic memory effect can be erased by either magnetizing the sample to fully magnetic state or heating the sample to higher temperatures. *© 2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4832459]

I. INTRODUCTION

The manganese oxides of the formula $R_{1-x}A_xMnO_3$ (R = rare-earth, A = alkaline-earth) were originally noticed because of their colossal magnetoresistance (CMR).[1–3] However, subsequent study reveals a variety of fantastic properties such as the strong correlation between the charge, orbital, and spin degrees of freedom.[4] Among them, the dynamic phase separation associated with the order-disorder transition of the manganites is especially interesting.[5] The research shows that conductive phase and insulating phase can coexist in submicron-scale and, moreover, their fractions and distributions can be easily modulated by external stimulus such as magnetic field,[6–8] electric current/filed,[9–14] and laser radiation.[15,16] Under the influence of external magnetic field, the system experiences an insulating to metallic transition, and usually remains in an intermediate state, rather than returning to the initial state, after removing magnetic field. This phenomenon is called memory effect. The memory can be erased by increasing temperature or applying magnetic field.[17] Sometimes laser illumination can produce similar effect.[18]

It is believed that magnetic field and light illumination could change the fraction of the insulating and conductive phases, thus influence the resistivity of the sample. Different from magnetic field and light, electric field/current processing may force the conduction with a preferred direction. For example, Belova found that the growth of metallic inclusions within the charge-ordering (CO) host preferably align along electric field,[19] i.e., the current processing can induce anisotropic phase separation in the manganese oxides. Dong *et al.* theoretically analyzed the current effect of phase-separated manganites,[20] and concluded that the metallic clusters preferred to configure themselves into strips along the direction of electric field, undergoing a filamentous percolation, i.e., the resistivity of the sample is different in the directions parallel and perpendicular to electrical current.

Up to now, there are many reports on the current effect of the manganites, but nearly none of these works paid attention to the direction dependence of the memory effect. $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ (y = 0.43) (LPCMO) is an oxide with the typical features of phase separation. In the temperature region below CO transition, it decomposes into a mixture of ferromagnetic (FM) metallic and CO insulating domains, and both domains are sub-micrometers in size. It is therefore an appropriate object for the study of the anisotropic memory effect induced by electric current.

In this paper, we presented a systematic investigation on the memory effect induced by the electrical current for the LPCMO thin film. We paid special attention to the anisotropic conduction of the sample along the directions, respectively, parallel and perpendicular to applied current. We first processed the sample and recorded the resistivity with various electric currents that are large enough to modify the resistive state of the sample, then measured the resistance again with a small current after removing the processing current along two perpendicular directions. It is found that large electrical currents result in a dramatic reduction in film resistivity, and only slightly rebounds after the removal of the current, giving rise to a memory effect. Even more, the resistivity, measured under small current after removing processing current, is considerably lower along the direction parallel than perpendicular to processing current, yielding an anisotropic memory effect.

II. EXPERIMENTS

A ceramic target with the nominal composition of LPCMO was prepared by solid-state reaction procedure. A 240-nm-thick LPCMO films was grown on the (110) $SrTiO_3$ substrate of the dimension of 3×5 mm^2 by the pulsed laser

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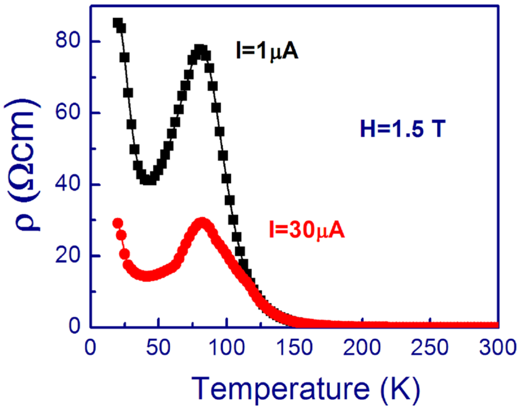

FIG. 1. Temperature dependence of the resistivity of $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ ($y = 0.43$) film, measured with two applied currents of 1 μA and 30 μA under an applied field of 1.5 T.

ablation technique. The temperature of the substrate was kept at 700 °C, and the oxygen pressure was at 45 Pa during the deposition. For anisotropic resistance measurement, the LPCMO film was then patterned into cross bar structure by the conventional photolithography and chemical etching technique (inset in Fig. 3). Silver pads (yellow area) were deposited on the four ends of cross-bar shaped sample for electric connection. The point contacts for electrical measurement were connected to the sample by ultrasonic Al-wire (20 μm in diameter) bonding. The standard four-probe technique was adopted for the resistance measurements along the x- and y-axes. A Keithley 2400 SourceMeter was used to provide processing and measurement currents and a 2182A Nanovoltmeter was used to collect electric signals. It is noteworthy that recording the resistance by applying processing current necessitated a two-probe through two internal lines of the four probes in order to guarantee the voltage within the limit of 2400 SourceMeter. In the process of electric measurements, a constant magnetic field of 1.5 T was applied to depress sample resistance to the range that the SourceMeter can provide enough high processing currents. All the measurements were carried in a Vibrating Sample Magnetometer (VSM) made by Quantum Design.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the resistivity of the sample as a function of temperature in the presence of a field of 1.5 T, measured with two different currents. It clearly demonstrates the influence of applied current on sample resistance in the temperature range below ~120 K. From 1 μA to 30 μA, the resistance is decreased by ~66% at 20 K, the lowest temperature of the present study.

Considering that the history memory usually occurs at low temperatures, we first cooled the sample to 20 K, then applied a magnetic field to depress sample resistance. For the present experiment we adopted a field of 1.5 T. It is a field depressing the sample resistance exactly to the range that it is available for the SourceMeter to output a current of the order of hundreds of micro-Amps. After these procedures, we processed the LPCMO film by applying a high current

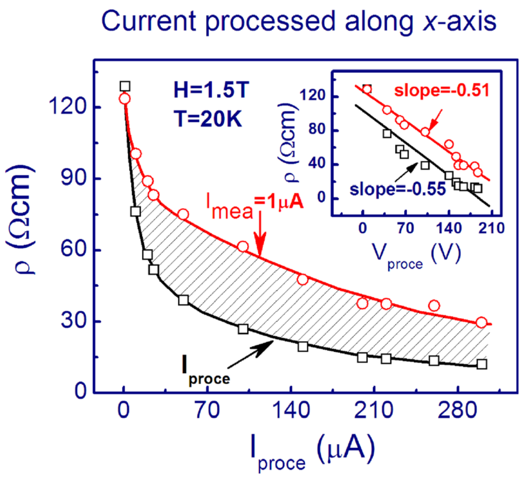

FIG. 2. Resistivity as a function of processing current (I_{proce}) measured at 20 K under the field of 1.5 T. The direction of processed current is along x-axis. The black symbols correspond to the resistivity measured with processing currents (I_{proce}), and the red symbols represent the resistivity measured with 1 μA ($I_{mea} = 1\mu$A) after the removal of processing current. The hatched area marks the volatile part of the memory effect. Inset: Resistivity as a function of processing voltage (V_{proce}). All lines are a guide to the eye.

along the x-axis through two internal lines of the four probes and simultaneously recorded the resistance, and then remove the processing current and measured the resistance again with a low applied current of 1 μA by the standard four probe technique. Repeating this process again and again after a successive increment in processing current after each cycle, two series resistance values can be obtained. Fig. 2 depicts the resistivity as a function of processing current I_{proce}. The resistivity exhibits a rapid decrease with applied current, especially in the initial stage. It is reduced by ~91.3% when the processing current grows from 1 to 300 μA, measured with the processing current, and by ~75.7% measured by a low current of 1 μA ($I_{mea} = 1$ μA) after removing the processing current. The hatched area marks the volatile part of the current effect. This result suggests that the processing current

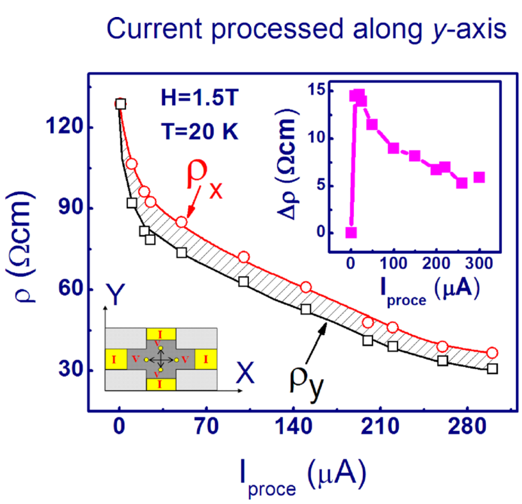

FIG. 3. Influence of processing current (I_{proce}) on resistivity, measured along the x- and y-axes with a current of 1 μA after the removal of the processing current along the y-axis. The upper right inset plot shows the difference of resistivities in two directions as a function of processing current (I_{proce}). The sketch shows the sample structure and electrode configuration.

depresses the resistivity, and this effect remains existed even removing the applied current. The inset of Fig. 2 shows a linear decrease of the resistivity with processing voltage V_{proce}, described by $R = aV + b$ with the slope of $a = -0.51$ and -0.55 for the resistances measured with low and high currents, respectively. No remarkable difference between the two slopes was observed, implying the similar reduction speed of the two resistivities.

It is an interesting question whether or not the processing direction affects the resistance. The inset sketch in Fig. 3 shows the cross-bar-structured sample for anisotropic resistance measurements. The experiments were conducted following the procedures below: The sample was first heated to a temperature above 300 K to erase possible history memory, and then cooled to 20 K. Afterwards a magnetic field of 1.5 T was applied, the processing current was applied in the *y*-axis and ramped from 0 to $I_{\max}$ and to 0 again ($I_{\max} = 300\ \mu$A), and then the resistance was measured along the *x*- and *y*-axes with an applied current of 1 μA. As shown in Fig. 3 and the inset plot, the resistivities along the two directions (ρ_x and ρ_y) are similar in the initial state, begin to deviate from each other at a low processing current of 10 μA, and the resistivity in processing direction is obviously smaller than that in perpendicular direction. The maximum difference of $\sim$15% is attained at $I_{\text{proce}} = 20\ \mu$A. Although a processing current above 20 μA further depresses ρ_x and ρ_y, the relative resistivity difference remains essentially unaffected (not shown). Also, the resistivity displays a linear decrease with processing voltage at a rate of $\sim$0.5 Ω cm/V.

This is the first observation for the current-induced resistive anisotropy. To verify the reliability of this observation, we changed the processing direction from *y*-axis to *x*-axis. Repeating the procedures described in the previous paragraph, we obtained the results presented in Fig. 4. The difference between ρ_x and ρ_y emerges at $\sim$10 μA, and reaches a maximal value of $\sim$−8.6% at $\sim$100 μA. In this case, ρ_x is smaller than ρ_y, exactly opposite to the results in Fig. 3. Once again, we observed considerably intensified depression of the resistivity in processing direction.

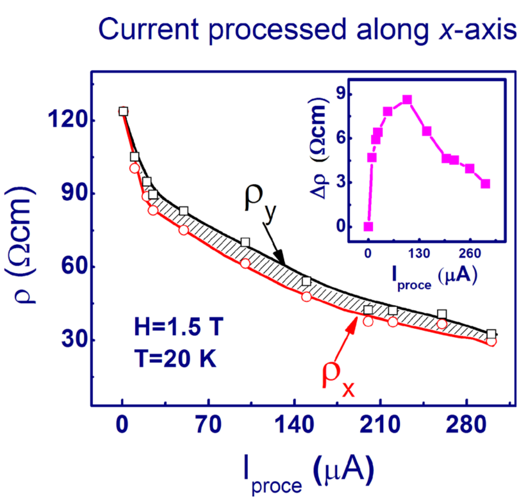

FIG. 4. Resistivity versus processing currents, measured along the *x*- and *y*-axes with a current of 1 μA after the removal of the processing current along the *x*-axis at 20 K under an applied field of 1.5 T. The inset plot displays the difference ($\Delta\rho$) of ρ_x and ρ_y as a function of processing current (I_{proce}).

LPCMO is typical phase-separated manganite. At low temperatures FM metallic domains and CO insulating domains coexist in the sample. As well established, the electric-field-resulted resistivity drop can be attributed to the collapse of the insulating CO phase, which is driven into metallic phase by electric field. From our experiment results in Fig. 2, it is plausible to believe that large current has converted parts of the insulating phase into metallic phase, yielding metallic filaments. Although the formed filamentary paths may be unstable since the resistance rebounds after the removal of the current, most of the effect is memorized. However, electric field affects not only the relative proportions of coexisted electric phases but also their spatial arrangements. According to the percolation model adopted in previous studies,[21,22] the metallic patches may nucleate in the form of threadlike metallic paths across insulating regions. As a result, the resistance can be greatly reduced while the magnetization is nearly unaffected. This is actually a signature of direction-preferential percolation. However, in previous work, only the electro-resistance along the direction of the processing current was studied, and no experimental effort has been devoted to the influence of current processing on the transport along the orthogonal direction. Theoretically, a dielectrophoresis model was proposed to explain the colossal electroresistance of phase-separated manganites.[20] It was supposed that electric field may affect the arrangement of different electric phases by assembling filamentous paths along its direction. This actually suggests an electric-processing-caused anisotropic transport in two orthogonal directions: The charge transport in the direction parallel to processing current could be easier than that along the orthogonal direction. Our experiment results could be a confirmation of anisotropic conduction in manganites. According to Figs. 3 and 4, the charge transport is obviously easily along the direction of processing current. In our experiment, the measurement current was applied through two external lines of the four probes, and the voltage signals were collected through the two inner probes. Therefore, the direction-dependent transport behavior could not be an effect of current-induced change of the electrode-sample contact. This phenomenon could not be an artifact either since it remains when exchanging processing axes. It could be an indication that the conduction paths burst by electric current has an inbuilt preferred direction.

The low processing current range in Figs. 3 and 4 deserve special attention. In this range, the resistance exhibits a sudden decrease with applied current. It is also in this range that ρ_x and ρ_y deviate from each other. Actually, the $\Delta\rho = |\rho_x - \rho_y|$ difference decreases when the processing current goes beyond this range, though either ρ_x or ρ_y keeps decreasing. In general, we expect that the conduction paths burst by processing current may show a dendritic structure, breaking through insulating region. It is possible that weakly linked points that act as bottlenecks impeding conduction but are susceptible to electric current were first broken by processing current. Since the current is low, its component in perpendicular direction has no obvious effect. As a result, the conduction filaments are considerably directional. The increase of the processing current will make the conduction paths more irregular in the meantime generating new paths and coarsening the already formed filaments. This, in turn, leads to the depression of the electric anisotropy, accompanying the improvement of the total conduction.

It is well known that the role of Joule heating needs to be considered in any electric-field- or current-driven effect. The temperature increase ΔT of the film at the temperature T induced by the Joule heating can be expressed as follows: $\Delta T(T,E) \approx 2P_l/k_{sub} = 2E^2 S[\rho(T+\Delta T)k_{sub}]^{-1}$, where P_l is the power dissipated per unit length of the sample, k_{sub} is the thermal conductivity of the substrate, S is the cross section of the film, andρ is the resistivity of the sample. According to our experimental values, the ΔT due to Joule heat is about 0.001 K at 20 K, which strongly suggests that Joule heating plays a negligible role in the whole measurements.

To explore the factors affecting the anisotropic memory effect observed in LPCMO, we further studied the influence of magnetic field. The experiment begins with the sample with the *x*-axis as easy conduction direction, obtained via the processing processes described in previous paragraphs. Fixing the temperature at 20 K and cycling magnetic field along the paths 1.5 T $\rightarrow$ H_{tr} $\rightarrow$ 1.5 T ($0 < H_{tr} < 4$ T), we obtained the resistivity-magnetic relations under a field of 1.5 T in Fig. 5. It is shown that the magnetic field below 1.5 T has no effect on resistivity, and the electric anisotropy keeps unaffected though the resistivity value grows slightly with the decrease of H_{tr}. This result indicates that a magnetic field lower than the presented field has no obvious effect on conduction paths. This is understandable since the FM fraction in the sample is not modified by H_{tr}. When H_{tr} exceeds 2 T, however, both the ρ_x and ρ_y exhibit a rapid decrease. From $H_{tr} = 2$ T to 4 T, the resistivity reduces by ~78%. It is a signature for the growth of the FM metallic fraction in the sample. Remarkably, the anisotropy of the resistivity was simultaneously depressed; the difference of resistivity varies from ~3 Ω cm to 0.4 Ω cm, reduced by ~87%. It implies that the growth of the FM domains has destroyed the original conduction paths and, therefore, depresses the differences of resistivity along two orthogonal directions.

Magnetic field enhances the growth of the FM phase, and thus deteriorates the unidirectional characters of the

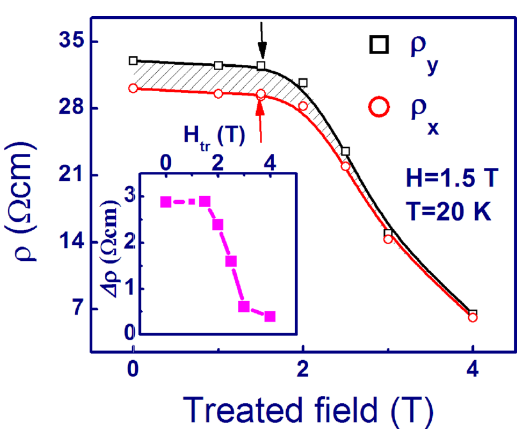

FIG. 5. Resistivity as a function of treated magnetic field for the current-processed sample (along x-axis), measured at 20 K under a field of 1.5 T. Inset: Difference ($\Delta\rho = |\rho_x - \rho_y|$) of the resistivities along the *x*- and *y*-axes, respectively. The arrows denote the initial resistivities.

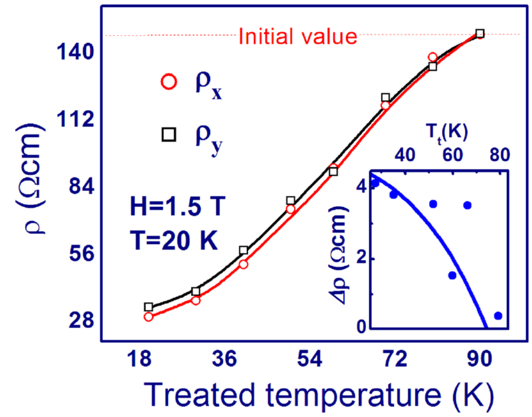

FIG. 6. Resistivity as a function of treated temperature for the current-processed sample (along x-axis), measured at 20 K under a field of 1.5 T. Inset: The blue symbols correspond to the difference ($\Delta\rho = |\rho_x - \rho_y|$) of the resistivities along the *x*- and *y*-axes, respectively. The line is a guide to the eye.

conduction paths. Heating the sample to higher temperatures, the thermal energy may also produce visible influence on the memory effect. The experiment also begins with the sample with the *x*-axis as easy conduction direction. After the thermal cycle from 20 K $\rightarrow$ T_{tr} $\rightarrow$ 20 K (20 K < T_{tr} < 100 K) in the absence of magnetic field, the magnetic field (1.5 T) was switched on and a pair of resistance data (ρ_x and ρ_y) corresponding to T_{tr} were collected. Lifting T_{tr} successively to different temperatures and then repeating the above procedure, the ρ-T_{tr} relation can be obtained. Here, we would like to emphasize that between two successive measurements the sample has always been warmed up to 300 K to erase possible memory effect. In Fig. 6 we show the results attained. A monotonic growth of the resistivity with T_{tr} can be clearly seen. This means that the memory effect of the sample has been destroyed by thermal energy, and the high resistance state has been gradually recovered as T_{tr} increases. To the temperature of 90 K, the original state is totally restored and, simultaneously, the anisotropy disappears completely. However, a careful analysis indicates that the resistance difference decreases at a much more rapid speed compared with that of the resistance growth. It means that the anisotropy is more sensitive to thermal energy. It is because the thermal energy can accelerate electron motion and partially counteract the energy derived from the electric field.

IV. SUMMARY

In summary, we have studied the current-induced effect on the $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ (y = 0.43) film by means of the dielectrophoresis model. With increasing current, the resistivity decreases. The resistivity drop induced by the applied current for manganite is suggested to originate from the appearance of percolation path. According to the measured resistivities in different directions after switching off the processing currents, it reveals that the change of resistivity in processing direction is obviously faster than that in perpendicular direction, which indicates that the metallic filaments grow easily along the direction of processing electric current, resulting in the anisotropic memory effect. It is also found that an application of strong magnetic field and high temperature can damage the anisotropic memory effect of the sample.

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