Current-induced asymmetric transport in $La_{0.8}Ca_{0.2}MnO_3$ epitaxial thin films

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We investigated the influence of a dc current on the transport properties in $La_{0.8}Ca_{0.2}MnO_3$ epitaxial thin films grown on $SrTiO_3$ substrates. A prominent finding is the appearance of asymmetric transport properties, like the behavior of *p-n* junctions, in a wide temperature range from 10 K to 300 K after the films were applied by a dc current over a threshold value. The asymmetric behavior resulted from a relative weak excitation is unstable and even spontaneously disappears. By optimizing excitation condition, the rectifyinglike behavior turns to be more significant and stable. Applying a small ac current could make the asymmetric transport collapse, subsequently, conventional symmetric transport behaviors appear. Phase separation and the coexistent ferromagnetic phases with different orbital order are taken into account in the interpretation of the observed phenomena. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2093934]

Because of the strong coupling between spin, charge, and orbital degrees of freedom, colossal magnetoresistive (CMR) perovskite manganites typically exhibit a rich variety of electronic and magnetic properties. A growing interest has been recently attracted to the influence of electric field/current to the transport in CMR materials.[1–7] Significant change of electric resistance induced by static electric fields has been demonstrated in thin films of $La_{1-x}Ca_xMnO_3$ using field effect configurations.[1] An applied dc current could lead to a transition from the electrically insulating charge-ordered (CO) state to a ferromagnetic (FM) metallic state, even for $Y_{0.5}Ca_{0.5}MnO_3$ in which a large magnetic field (~40 T) has no effect on the charge-ordered state.[2] In our recent study, remarkable change of resistance induced by a dc current has been observed even in epitaxial thin films of $La_{1-x}Ca_xMnO_3$ and $La_{0.85}Ba_{0.15}MnO_3$.[5–7] Furthermore, several groups[8–10] investigated the influence of a pulsed ac current on the transport properties in perovskite oxides sandwiched between different metal electrodes. A reversible resistance switching can be induced in the sandwiched systems. Such a resistance switching was experimentally proven taking place at the interface between metal and oxides (not in the body of the oxides).[9]

Phase separation is a character of the strongly correlated electronic systems. Experiments on various manganites have proved that an electric field/current can destabilize or even completely destruct charge or orbitally ordered phase in the systems. However, it is unclear that how the systems respond to an extreme condition, such as the application of an extremely large electric field. Manganite perovskites $La_{1-x}Ca_xMnO_3$ are characterized by a complex phase diagram containing a rich variety of magnetic and electronic phases. Because of the subtle competition between charge-carrier motion and magnetic spin and orbital moments, phase-separated state becomes a stable ground state, especially for low-doped samples. In this study, we chose $La_{0.8}Ca_{0.2}MnO_3$ (LCMO) epitaxial thin films to investigate the influence of an extreme large current on the transport properties. Asymmetric transport appears after an excitation by applying a large dc current for a short duration. The observed asymmetric transport takes place in the body of the films and strongly depends on the duration of dc current excitation.

The present $La_{0.8}Ca_{0.2}MnO_3$ thin films, with thickness ~100 nm, were grown on $SrTiO_3$ (001) substrates. The details of film preparation and characterization can be found in our previous reports.[5,6] The electric transport properties in the body of the films were measured by using the standard 4-probe technique. In order to apply a current with a high density, the films were patterned into a microbridge of 50 μm in width and 200 μm in length. Silver contacting pads were then evaporated on the sample and the current leads were made using a MEI-907 supersonic wire bonder to obtain low Ohmic contacts.

Figure 1 presents the temperature dependent resistance without magnetic field and magnetization measured under 100 Oe. Inset is the schematic of the samples for applying dc current excitation and measuring electric transport. It is found that the Curie temperature T_C of $La_{0.8}Ca_{0.2}MnO_3$ film is at ~272 K, much higher than that of its bulk material (~190 K). Such a deviation is consistent with previous reports.[6,11] To investigate the current-induced effect, a dc current with high density was applied at a specific temperature (~228 K), where R starts to increase in the R-T curve (see Fig. 1), for a short duration δt. The specific temperature of ~228 K was chosen based on the following two considerations. First, the resistance at ~228 K is still small and the direct impact caused by the self-heating effect should be

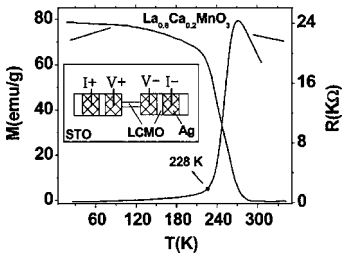

FIG. 1. The temperature dependent resistivity at zero field and magnetization measured under 100 Oe for a $La_{0.8}Ca_{0.2}MnO_3$ thin film. Inset is the schematic of the samples for applying dc current excitation and measuring electric transport.

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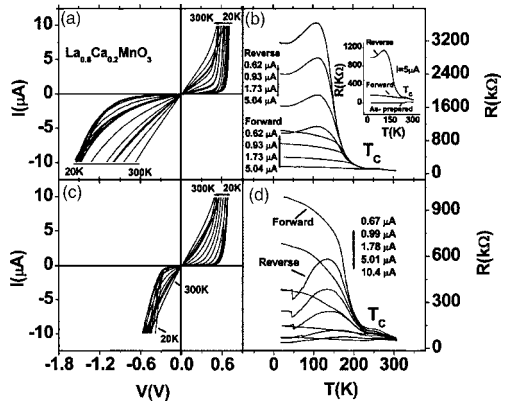

FIG. 2. For a $La_{0.8}Ca_{0.2}MnO_3$ film, after excited by a dc current of 9.8 mA with a duration of 8 min. (a) *I-V* curves from 20 K to 300 K with a step of 20 K; (b) *R-T* curves upon applying currents of 0.62, 0.93, 1.73, and 5.04 μA in both directions parallel and antiparallel to the excitation current; inset of (b) shows the *R-T* curves in both directions compared to that of the pristine state under a same current of 5 μA. For such an induced state, after applying a small ac current 0.01 mA about 200 cycles; (c) *I-V* curves from 20 K to 300 K with a step of 20 K; (d) *R-T* curves upon applying currents of 0.67, 0.99, 1.78, 5.01, and 10.4 μA in both directions for this stage.

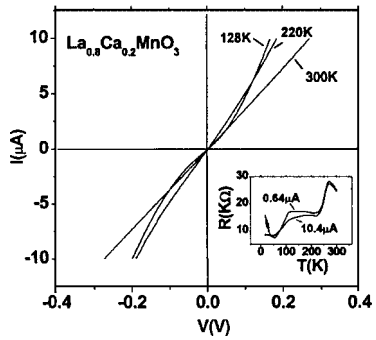

FIG. 3. *I-V* curves of the state induced by 9.8 mA with 8 min after the ac current treatment for further 300 cycles for a $La_{0.8}Ca_{0.2}MnO_3$ film. Inset is the *R-T* curves measured using different currents of 0.64 μA and 10.4 μA.

weak. Secondly, the strong competition between electron itineracy and self-trapping[12] at ~228 K, the start metallike phase transforms to the insulatinglike phase, would enhance the influence of bias current on transport and magnetic properties.

After the films were treated by such a manner, their *I-V* curves exhibit asymmetric characteristics, similar to the rectifying behaviors in conventional *p-n* junctions. It is found that the asymmetric transport resulted from a relative weak excitation, e.g. applying a current of 9.8 mA with 2 min duration, is unstable and sometimes spontaneously returns to its pristine state. Suitably prolonging the acting time or increasing the magnitude of excitation current could stabilize the state with asymmetric transport. Figure 2(a) presents the *I-V* curves of the film measured at different temperatures from 20 K to 300 K after the current-excitation. The dc current applied for excitation was 9.8 mA (the current density $\sim 2.2\times 10^5$ A cm^{-2}) and the duration δt was 8 min. One can notice the rectifyinglike properties at various temperatures. The forward direction is the same as the dc excitation current. A threshold of electric current exists for inducing such rectifyinglike behaviors. An applied current lower than 9 mA hardly causes a remarkable change of transport in these films. It is found that the asymmetric behavior is strong at low temperatures, and gradually becomes weak with increasing temperature. We know the transport properties in our samples were completely dominated by LCMO films since the STO substrates behave dielectric. For understanding the interesting observations, we measured the temperature dependent resistance (*R-T* curves) upon applying small currents in both directions parallel and antiparallel to the excitation currents [see Fig. 2(b)]. The inset of Fig. 2(b) presents the *R-T* curves in both directions compared to that of the pristine state under a same current of 5 μA. It is found that the electric resistance is not only significantly increased compared to the pristine state, but also becomes dependent on the direction of the applied current in the whole measured temperature range. However, the resistance anomaly around T_C always remains, and the position of the resistance peak T_P keeps nearly unchanged upon current excitations. For temperatures above 200 K, the resistance is weakly dependent on the value and direction of the applied current, but at lower temperatures, the remarkably enhanced resistance becomes highly sensitive to currents and strongly dependent on current direction. Such features are consistent with the evolution of the rectifyinglike behavior with temperature observed in Fig. 2(a). The induced asymmetric transport is rather stable. Exposing samples in air at room temperature for quite a long time (>2 weeks) did not cause any change in the rectifyinglike behavior. On the other hand, an application of a small ac current could strongly influence the asymmetric transport and even make it disappear in the whole temperature range from 20 K to 300 K.

Figure 2(c) exhibits the *I-V* curves of the film after the treatments by applying a small ac current (0.01 mA, 0.025 Hz) for about 200 cycles. The diffusion voltage and the shape of *I-V* curves in the forward direction remain nearly unchanged compared to the freshly induced state. However, the breakdown voltage significantly decreased and the shape of *I-V* curves in the reverse direction changed considerably. The breakdown voltage even becomes smaller than the diffusion voltage. Figure 2(d) shows the temperature dependent resistance upon applying different small currents in both directions for this stage. The resistance with current along forward direction remains almost unchanged compared to the freshly induced state [see Figs. 2(b) and 2(d), noting the different scales of the *R* axis]. However, the resistance behavior with current along reverse direction has been significantly changed and even becomes smaller than the forward resistance. These features are consistent with the observations in Fig. 2(c). The appeared step-change in reverse resistance at low temperatures is intrinsic and may reflect the metastable characteristics. More treatments by such an ac current would make the induced resistive state collapse, and the asymmetric behavior disappears. Figure 3 and its inset display the *I-V* and *R-T* curves of the film after ac current treatments for further 300 cycles. The *I-V* curves become nearly linear and symmetric. Simultaneously, the resistance reduces remarkably [see Fig. 2(b) and the inset of Fig. 3, noting the different scale of the *R* axis] and becomes independent of the direction and also nearly insensitive to the magnitude of the small applied current. One thing should be noticed is that during the whole modulation process upon ac current treatments the resistance anomaly around T_C always remains, and the position of the resistance peak T_P keeps nearly unchanged.

The rectifyinglike behavior could become more asymmetric and stable by optimizing excitation condition, such as suitably extending the duration of current excitation or in-

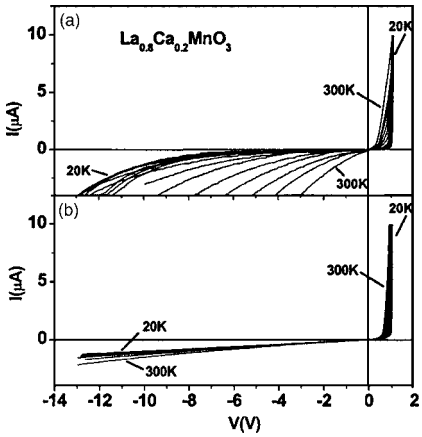

FIG. 4. *I-V* curves measured from 20 K to 300 K with step of 20 K for the states induced by (a) 9.8 mA with 11 min, and (b) 11.2 mA with 5 min.

creasing the current. Figures 4(a) and 4(b) present the *I-V* curves of the films measured at different temperatures from 20 to 300 K after different excitations, 9.8 mA with 11 min, and 11.2 mA with 5 min, respectively. The *I-V* curves become more asymmetric in the whole temperature range. For the case of 11.2 mA with 5 min, the all *I-V* curves in the reverse direction almost coincide with each other, indicating the transport is nearly independent of temperature. For such a case the obtained rectifyinglike behavior is more stable. Applying ac current can hardly destroy it. One reason might be that the reverse resistance is so high that a sufficient current for destroying the asymmetric transport can be hardly applied.

Such observed effects cannot be attributed to the escape of oxygen due to the self-heating effect. We studied vacuum annealing effects for an as-prepared thin film of the present composition with identical thickness. The results are very similar to a previous report.[13] Simply annealing in a vacuum could only cause a shift of the position of peak resistance to lower temperature and make the resistance increase. No asymmetric transport was observed. We also verified that the asymmetric transport is not due to the Schottky potential between LCMO and silver electrodes. We redeposited four silver electrodes on other positions or even replace them by silver paste after current excitation. No change of the asymmetric transport was found.

Recent NMR (Ref. 14) and structural investigations[15] on low-doped LCMO system have confirmed that its phase-separated (PS) ground state is constituted by two ferromagnetic phases with different orbital order (OO). In one phase an antiferrodistorsive-type OO is favored by FM superexchange interactions, behaving insulating characteristics, while in another phase the ferrodistorsive-type OO promote double exchange interactions, exhibiting metal behavior. An electric field may directly affect the directional order of orbitals and thus alter the magnetic and conducting states.[16] In the system with phase separation, partially metallic and partially insulating, an applied electric field would have a distribution in the broad range of phase space depending on the size, shape, and the phase distribution. When the applied current is high enough, the associated electric field distributed in the phase space may strongly impact the orbital order of the phases and enforce a thorough change in the topology of the phase coexistence. Eventually a new state with new coexistence of the phases may be formed. After removing the large excitation current, an asymmetric barrier, likely the depletion region in *p-n* junctions, might be remained in the phase space. Asymmetric transports appear.

The action of the strong current on the topology of the phase coexistence or the rearrangements of the orbital domains could be realized either by means of carrier injection or by direct interaction with elastic forces. Therefore, it can be understandable that the process is not instantaneous, and a sufficient magnitude of the excitation current and a duration of acting time is required. A weak excitation (relative small current or short acting time) cannot thoroughly disturb the PS state and the induced state is unstable and even spontaneously return to its pristine state. Suitably prolonging the acting time or increasing magnitude of the current for excitation can further improve the induced orbital distortion, resulting in a strong increase of electric resistance in the whole temperature range from 20 K to 300 K. The local elastic forces existed in the microstructure stabilize the orbital distortion and make it robust. It can not spontaneously return to its initial state without external perturbations. An application of an ac current may destabilize the induced orbital distortion in the system[3,4] and make the sample gradually change towards its initial state. The formation of the asymmetric potential might be related to the corresponding change of local microstructure caused by the strong interaction between large current with the PS state. To completely understand the nature of the asymmetric barrier and the evolution process upon an application of a small ac current, further detailed investigations are still required.

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