Stabilization of the metallic state by electric currents in phase-separated manganite thin films

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We present the evidence that electric current stabilizes the metallic phase in a metal or insulator phase-separated manganite film. Simultaneous IR transmission measurement, which reflects the average metal or insulator ratio, reveals that the observed resistance near the metal to insulator transition is governed by a filamentary path occupying an insignificant fraction of the total volume. This allowed us to access the behavior of a single mesoscopic metallic path under a varying current strength via macroscopic resistance measurement.

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I. INTRODUCTION

Manganese oxides have been extensively studied in the past decade particularly because of the drastic change in the transport properties under external stimuli:[1] sharp transition between conducting and insulating states has been often associated with the percolating behavior in the phase-separated electronic states.[2] In this scenario, the magnetically induced transition [colossal magnetoresistance (CMR)[3]] and photoinduced transition[4] can be easily understood in terms of the increase in the metallic fraction in the insulating background, since the external field favors the ferromagnetic metallic state in the former case and destroys the charge-ordered insulating state in the latter case.

Also known is the electric-field-induced insulator-to-metal transition,[5] in which it is presumed that the electric field forces the sliding of the charge-ordered state.[6] In an inhomogeneous system (as is the case in a phase-separated sample), however, it is difficult to estimate the critical field strength because the location of the actual insulating gap that supports the applied potential drop is not clear. In order to avoid this ambiguity, constricted geometry has been employed recently, in which the closing and breaking of the last percolating junction in the constriction show up as macroscopic and often reproducible conductivity switching.[7,8] It should be cautioned, however, that the fabrication process itself could introduce inhomogeneity in the sample. Furthermore, the notion of electric field as a driving force is not obvious at all in these examples considering the relatively large power consumption necessary for the transition (for example, approximately 20 mW for the case of thin film in Ref. 9). The sample heating has been demonstrated as a cause for the switching behavior in certain cases.[10]

In this paper, we present another route to realize the metallic phase, the current-stabilized metallic state. In contrast to the mechanisms described above, the metallic phase is dynamically maintained by the forced charge motion. Since the additional interaction between neighboring Mn sites mediated by the externally injected current favors uniform charge distribution and ferromagnetism through the double-exchange interaction,[11] it is not surprising that the balance between ferromagnetic metal (FM-M) and charge- and orbital-ordered insulators (COO-Is) tips toward the metal side by the electric current. By choosing a proper film sample and hysteretic conditions, we show that the response of one mesoscopic path to the varying current can be studied. A clear percolative behavior is demonstrated by monitoring the average electronic state (the IR transmission) and the electrical conductivity simultaneously.

II. EXPERIMENT

The sample is a thin film of $Pr_{1-x}(Ca_{1-y}Sr_y)_xMnO_3$ ($x=0.45$, $y=0.25$) grown on a perovskite $(LaAlO_3)_{0.3}(SrAl_{0.5}Ta_{0.5}O_3)_{0.7}$ (011) substrate. The growth condition and the characterization have been described previously.[12] It shows a bicritical behavior with a paramagnetic insulator at higher temperatures, and COO-I and FM-M are competing at lower temperatures.[13] Near the bicritical point, CMR and photoinduced insulator-to-metal transition have been observed.[4,13] A reference film with the composition of $y=0.4$, which is far from the critical point, was fabricated in a similar fashion.

The IR transmission spectra (T_{IR}) were taken using a monochromatized IR lamp light and an InSb detector. The transmittance of the film was calculated using a bare substrate as a reference. The reflectance correction turned out to be small and ignored.

III. RESULTS AND DISCUSSION

Figure 1(a) shows (T_{IR}) spectra at 90 K. The sample was initially cooled to 90 K in zero magnetic field. The spectrum is typical of an insulator with a charge transfer gap ($\sim$0.25 eV). The application of a magnetic field of 5 T made the sample metallic, signified by much reduced transmission toward the low photon energy. The response is in good accord with that found in the resistivity in Fig. 1(b), in which the temperature dependence of the resistivity under various magnetic fields is depicted. Compared with the CMR effect found in a $y=0.4$ film [Fig. 1(c)], in which the COO-I phase is absent[14] and the transition into the metallic phase is less complicated, one may safely say that the field-induced metallization is saturated and completed at 5 T for a $y=0.25$ sample. Therefore, we take the resistivity of $\sim$mΩ cm and $T_{IR}\sim 0.4$ at 0.5 eV as our reference values for the fully metallized film. It is clear, by noting T_{IR} in Fig. 2, that the sample is only partially metallic in zero magnetic field even

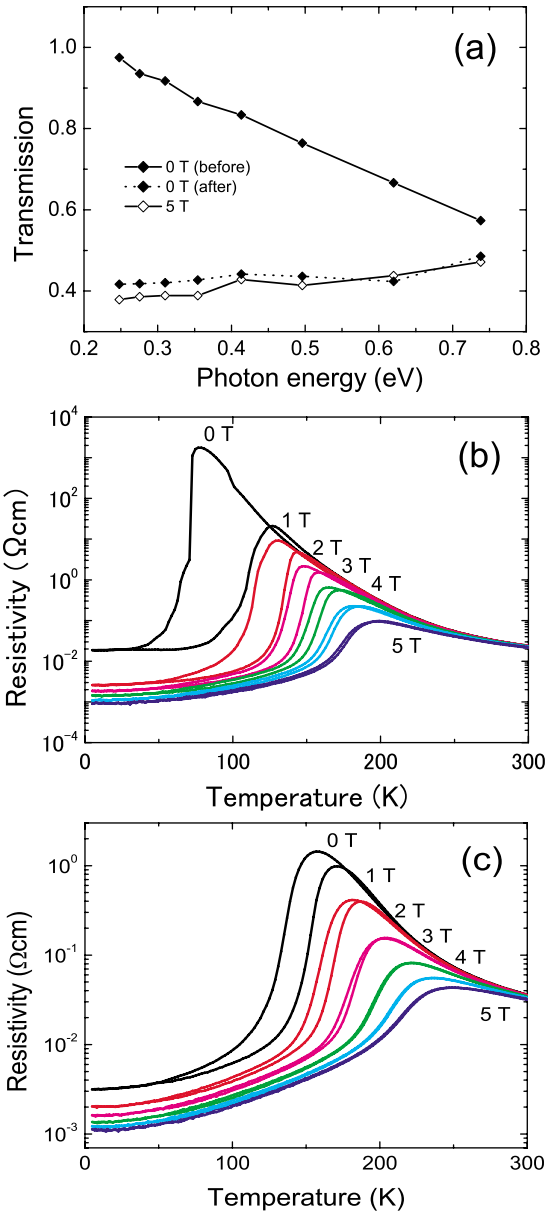

FIG. 1. (Color online) (a) IR transmission spectra of $y=0.25$ sample after cooled to 90 K in zero magnetic field and after subsequent application and removal of a magnetic field of 5 T. (b) The CMR effect in $y=0.25$ and (c) $y=0.4$ films.

at the lowest temperature. Note also that at 90 K, the metallic phase is more stable and the film stays metallic after the removal of the magnetic field. We found a similar persistent effect in the current-induced metallic phase as described below.

In the following, we use the resistance (ρ) and T_{IR} at 0.5 eV to characterize the sample; the former probes the most conducting path and the latter is a signature of the average electronic state. The sample area between the electrodes was illuminated uniformly for the transmission measurements, and both T_{IR} and ρ were monitored simultaneously.

In Fig. 2, typical temperature dependence of ρ and T_{IR} is presented. The resistance was measured under a constant current of 1 μA. They both show insulator-to-metal transition with a large hysteresis between the cooling and warming processes. Although their detailed temperature dependence differs from run to run, there are qualitatively reproducible aspects as follows:

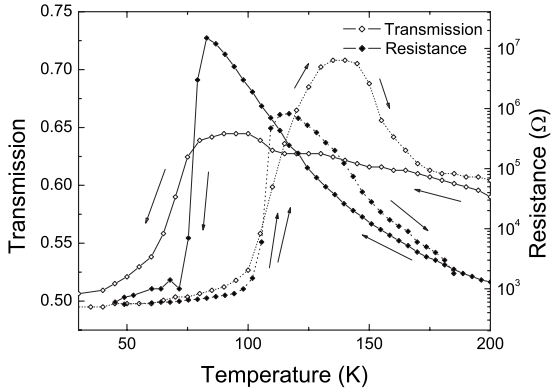

FIG. 2. Temperature dependence of resistance and transmission at 0.5 eV. Resistance and transmission were measured at the same time.

(1) T_{IR} peaks at higher temperatures than ρ in both the cooling and warming runs,

(2) ρ changes much more precipitously than T_{IR} in both cooling and warming runs,

(3) both ρ and T_{IR} overshoot the corresponding cooling curves in the warming run, and

(4) the cooling and warming curves merge above the charge-orbital-ordering temperature.

The aspects (1) and (2) clearly indicate that the electrical conduction is dominated by the filamentary metallic path rather than the average electronic state. A dramatic demonstration of this is shown in Fig. 3, in which ρ and T_{IR} were monitored as a function of the applied current at 70 K. As will be discussed in more detail shortly, the metal phase is induced by the electric current, a mere 10 nA in this case. At

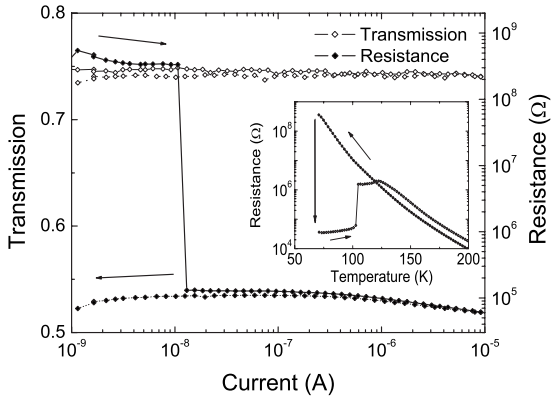

FIG. 3. Current dependence of resistance and transmission at 0.5 eV and 70 K. Resistance and transmission were measured simultaneously. The inset shows temperature dependence of the resistance at a constant current of 1 μA. Right after the current-induced insulator-to metal-transition in the cooling run, the temperature sweep was reversed.

this temperature, the metallic state is more stable. Therefore, the metallic path formed by the current stays metallic even when the current is reduced just as the case for the magnetic-field-induced metallic state as noted above. On the other hand, there is no sign of the corresponding transformation in T_{IR}. Obviously, the fractional increase of the metal phase (the additional conducting path) is too small to be detected in T_{IR}.

The resistance overshoot in the warming process [aspect (3)] has been noticed before. A qualitative explanation has been put forward,[15] in which the overshoot in ρ is due to the stronger segregation of metallic and insulating phases in the warming run, while the metallic fraction is the same as in the cooling process at the same temperature. This scenario is not consistent with our observation that the average electronic state is much more insulating (higher T_{IR}) as well. Together with aspect (4), it seems that there is a mechanism that locks the insulating part into a more stable form at low temperature, resulting in the large hysteresis. One candidate is a long range lattice deformation.[16,17] However, a more elaborate structural study is clearly in need, and we will leave this for a future study.[14]

The effect of the electric current stabilizing the metallic phase is revealed both in cooling and warming runs. In the cooling process, as was noted above, ρ starts to drop *after* the average metallic fraction starts to grow: a clear signature of the percolative conduction. Here, the current flows in an inhomogeneous landscape seeking the low resistance terrain. As the high current density flowing in a restricted geometry in the percolating texture favors the conversion to the metallic phase, the resistance of the region with the higher current density diminishes further. Thus, created lower resistance attracts more current. The positive feedback process, then, ends up with a precipitous resistance drop. Since the metallic phase is more stable at this temperature, the metallic conduction path formed is stable after the removal of the current (Fig. 3) a persistent current-induced insulator-to-metal transition. If the temperature is cycled right after a one sharp drop, the resistance increases sharply (inset of Fig. 3), suggesting that only one filamentary path is involved in this case. The temperature dependence of the path confirms that it is metallic.

The efficiency of the current to induce the metallic state depends on the relative stability of the two phases and hence a function of the temperature. This is illustrated in Fig. 4. Although the magnitude of the drop varies, due probably to the eventual width of the stable metallic path, the current at which the first resistance drop occurs shows systematic behavior, as illustrated in the inset of Fig. 4. Although this does not imply any critical behavior because the current is not a thermodynamic variable, it suggests that the end point is the absolute stability limit in a first-order phase transition separating COO-I and FM-M.

The metal-phase-stabilizing effect of the current is seen more clearly in the warming process starting from the metallic phase. As the sample temperature rises, metallic paths are broken while the insulating phase grows. Since the resulting increase in resistance is inversely proportional to the number of paths, it is hardly noticeable at first. However, at the last stage in which only one path is left, ρ directly reflects the microscopic dynamics. Figure 5(a) shows *I-V* curves in the

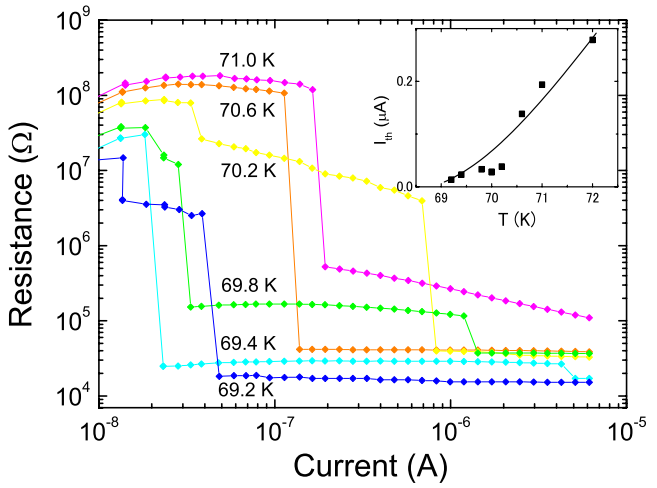

FIG. 4. (Color online) Current dependence of resistance at various temperatures. The inset shows temperature dependence of the critical current I_{th}, which is defined as the current at which the first resistance drop in the main panel occurs. The line is a guide for the eye.

warming process. At 120 K, far above the transition temperature, *I-V* curve shows Ohmic behavior. However, below but near the transition temperature, *I-V* curve is not linear. This is because, at this temperature, the conducting path is much restricted and the current density is thus high enough so that the path widens in response to the increase in the

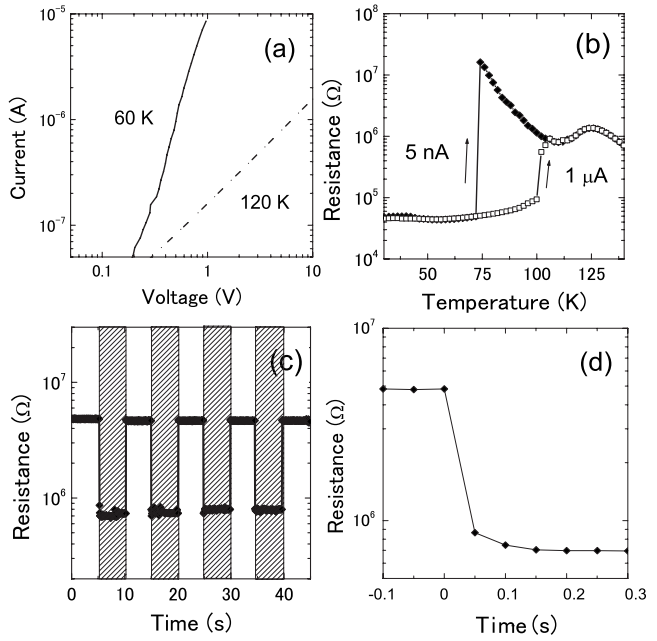

FIG. 5. (a) *I-V* relationship at 60 and 120 K in the warming process. (b) Temperature dependence of resistance in the warming process under constant current of 5 nA and 1 μA. (c) Change of resistance by switching the applied current at 90 K in the warming process. Applied current was switched between 5 nA and 1 μA (hatched) every 6 s. (d) Transient resistance drop in going from 5 nA to 1 μA.

current. By the same mechanism, if a constant current is maintained in the resistance measurement on warming, the temperature of the resistance jump depends on the current, as is illustrated in Fig. 5(b). There is a large difference in the transition temperature for currents of 1 μA and 5 nA. Note that the shift of the temperature is *opposite* to what is expected from the heating effect of the current. In our sample, the higher the temperature is, the less favorable the metallic phase becomes. Therefore, one would expect that the larger current induces the transition at lower temperature if the Joule heating was the cause of the shift.

Because the last metallic path is dynamically maintained by the current in the more stable insulating background, one can manipulate the resistance by switching the current. An example is shown in Fig. 5(c). While the sample was kept at 90 K, the current was switched between 5 nA and 1 μA. The resistance switches between two well defined values in response to the current. The larger current clearly widens the path by converting the surrounding insulator to metal. The dynamics of the effect is the most interesting, since it may reveal the mechanism of the metal-phase-inducing effect of the current. Unfortunately, the high resistance of the sample does not allow us a faster measurement. Figure 5(d) shows the transient resistance change in switching from 5 nA to 1 μA. This is the fastest speed that we can digitize in the high resistance measurement mode of our setup. The detailed dynamics is unclear at the moment.

It would be interesting to comment on the size of the filament. Since we do not know the real shape of the conduction path, we cannot estimate the size in a quantitative manner. Assuming that the film is uniform across its thickness ($\sim$60 nm) and assuming that the narrowest constriction in the conducting path is rectangular in shape in the film plane, all we can determine from the macroscopic measurements is the aspect ratio of the rectangle. The widest estimate is arrived if we assume that a single uniform and straight filament connects the two electrodes. Using the resistivity of the pure metallic state of mΩ cm, as was determined above, and the electrode separation of 2 mm, the width of the filament would be 500 nm for 1 μA and 70 nm for 5 nA. The current densities would be 3 kA/cm^2 and 100 A/cm^2, respectively. This should be the uppermost estimate in the width, and thus, the conduction path is indeed mesoscopic in size.

One may question if the inhomogeneity is intrinsic or extrinsic. Structural defects and nonuniform chemical composition, for example, can act as a source from which a metallic path grows. Although we do not have a definitive answer to this question, we argue that the inhomogeneity is intrinsic based on the following observation. (1) Each line in Fig. 4 was taken after annealing the sample above the COO-I transition temperature. The scatter in data suggests that each cooling run is statistically independent. (2) In a macroscopic but space-resolved resistance measurement, the inhomogeneity pattern varies after heating above the room temperature.[18] (3) The space-resolved photoinduced insulator-metal transition experiment indicates that the most sensitive spot varies from run to run.[19]

There have been many reports on the effect of electric current inducing metallic state.[20] However, no conduction measurement has been so far done at so low current level as done here, which helps us to study the behavior of a single mesoscopic path.

We are aware of one theoretical study on the effect of the forced charge motion on the correlated electronic state,[21] but it does not deal with the insulator or metal stability described here, and the subject is largely untouched. Experimentally, the heating side effect is the annoying problem in many cases. However, in the example shown here, the power dissipation is many orders of magnitude lower than most of the previous transport measurements and, moreover, the temperature rise would impede the current-induced effect since the high temperature phase is an insulator. We expect that the present study stimulates the study of the correlated electron systems away from the thermal equilibrium.

IV. CONCLUSION

We have shown clear evidence for percolating conduction in a manganite thin film by monitoring the IR transmission and conductivity simultaneously. The effect of electric current to stabilizing the metallic state, free of the doubt of the Joule heating, has been presented. The response of a single mesoscopic conducting path has been studied by employing very much reduced current.

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