Resistance Steps and Asymmetric Conduction Induced by Currents in $La_{0.8}Ca_{0.2}MnO_3$ Films

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A constant voltage mode of a power supply was adopted to study the current-induced effects in $La_{0.8}Ca_{0.2}MnO_3$ epitaxial thin films. After processed by a current of the density of $\sim 3 \times 10^5$ A/cm^2 for 1 minute, they exhibited anisotropic conduction. Like the behavior of p-n junctions, the current starts to increase dramatically in the forward direction at a threshold voltage (opening voltage V_{OP}), e.g., ~2.3 V at 30 K. For a processing with extending duration, the resistance while processing dropped from 10 kΩ to 2 kΩ. Current voltage curves measured following such a step showed more stable asymmetric characteristics and substantial reduction of V_{OP}, which is ~0.8 V at 290 K and ~1.6 V at 50 K. Our results are useful for revealing the origin of asymmetric conduction induced by current and for potential application.

***Index Terms*—Asymmetric conduction, resistance steps, transport.**

I. INTRODUCTION

HOLE doped manganites with a general formula of $R_{1-x}A_xMnO_3$ (R = rare earth ions, A = alkaline ions) have been extensively studied since the discovery of colossal magnetoresistance (CMR) [1], [2]. In such a compound, cross couplings often occur between different degrees of freedom [3], [4], such as charge, orbital, spin, and lattice. It results in a variety of electronic phases, which are energetically close to each other. As a consequence, these materials are sensitive to various external stimuli, including magnetic field, pressure, X-ray radiation and electric field/current [5], [6]. The responses of manganites to such perturbations provide an effective way to reveal the underlying physics and are promising for practical applications. Asamitsu *et al.* [6] reported the early study of resistance change of manganite ($Pr_{0.7}Ca_{0.3}MnO_3$) upon an applied electric field/current. Because the resistance drop upon the electric field/current was as large as several orders, it was named as "colossal electroresistance" (CER). Subsequently, many investigations on current induced effects appeared. The studies at the beginning were limited on materials with the charge ordered state. Later, the investigations were extended to materials with other ground states. Gao *et al.* [7] reported that the electric current can suppress the peak resistance of $La_{1-x}Ca_xMnO_3$ (x = 0.2, 0.3). It was also found that a metastable state, which is sensitive to weak currents, can be induced when the applied current is larger than a critical value [8]. More intriguingly, distinct asymmetric conduction was induced from an initially uniform system by a large current [9]–[12]. On the other hand, there have been more and more interests in pulsed voltage and current induced resistance switching [13], [14]. In this study, we report the co-occurrence of asymmetric conduction and abrupt resistance steps induced by currents in $La_{0.8}Ca_{0.2}MnO_3$ films.

II. EXPERIMENTAL

Thin films of $La_{0.8}Ca_{0.2}MnO_3$ were fabricated by pulsed laser deposition technique using KrF exicmer laser with the center wavelength of 248 nm and the repetition rate of 5 Hz. The films were grown in an oxygen atmosphere of 70 Pa on $SrTiO_3$ (001) substrates kept at 750 °C. To avoid severe oxygen vacancy, the as grown films were post annealed in air at 850 °C for 2 h. The thickness of the films is about 160 nm as controlled by deposition time. Standard $\theta - 2\theta$ X-ray diffraction data show that the films are highly epitaxial and are of a single phase. These samples were patterned into microbridges by conventional photolithography technique. The dimension of the microbridges is $\sim 10\ \mu m \times 100\ \mu m$. Four silver pads were evaporated on the film thermally as electrodes. Standard four probe method was used to measure temperature dependence of resistance and current voltage characteristics. As shown in the inset of Fig. 1, a constant voltage mode of a power supply (Sorensen DCS 300 V–3.5 A) was used to study the current processing. Here, the load resistance of $\sim 2000\ \Omega$ was used to protect the film against a burst of current.

III. RESULTS AND DISCUSSION

The temperature dependence of resistance shows the typical metal-insulator transition (MIT) expected in CMR materials, as shown in Fig. 1. This transition occurs at $T_P \sim 263$ K, much higher than bulk material. Such a phenomenon is consistent with previous reports [8], [9]. It was found that due to strain the unit cell volume is smaller than that of bulk samples, which would enhance the electron hopping integral. No obvious change was found on the temperature dependence of resistivity when the film was patterned to a wire with a width of 10 μm. It was reported that distinct changes, such as the appearance of a second robust metal insulator transition [15] and giant ultra-sharp steps [16], were found in La-Pr-Ca-Mn-O films when the width was reduced to a similar scale. These reemergent phenomena were due to the large scale phase separation in La-Pr-Ca-Mn-O. Therefore, it is safe to say that the length scale of phase separation in the present LCMO film should be much smaller.

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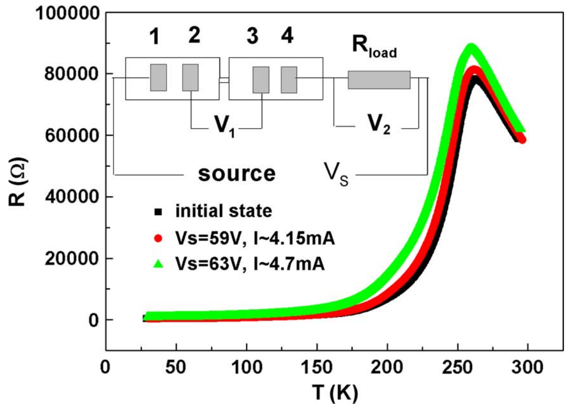

Fig. 1. R-T curves measured with a current of 10 μA before and after current processing with parameters $V_S = 59\ V, I \sim 4.15\ mA, duration = 30\ min$ and $V_S = 63\ V, I \sim 4.7\ mA, duration = 30\ min$. The circuit used for current processing and schematic geometry of the microbridge are in the inset.

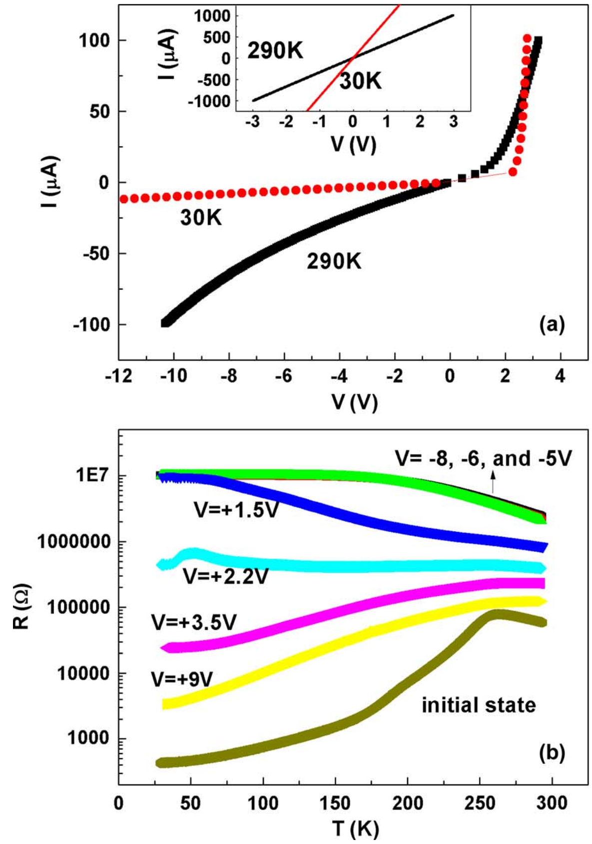

Fig. 2. (a) Asymmetric I-V curves and (b) R-T curves at selected bias after current processing with parameters $V_S = 64\ V, I \sim 4.8\ mA, duration = 1\ minute$. The positive direction is that of the processing current. The inset of (a) shows the I-V curves measured between electrode 1 and 2 after current processing.

To study the current-induced effects, currents larger than 1 mA were applied to the microbridge at room temperature in air using a constant voltage mode. R-T curves and I-V curves were measured subsequently using a four probe method to check the changes. With the processing parameters $V_S = 59\ V$, $I \sim 4.15\ mA$, and $duration = 30\ min$, only slight increase of resistivity was found [Fig. 1]. When the processing current reached ~4.7 mA ($V_S = 63\ V, duration = 30\ min$), the increase of resistivity became substantial and the decrease of T_P is as large as ~4 K, but the I-V curves are still linear. Previous studies [9]–[12] found that asymmetric I-V curves appeared only when the processing current exceeded certain critical value. Here, the substantial changes of transport properties imply that 4.7 mA is close to the threshold.

Fig. 2 shows the anistropic conduction after the film was processed by a current of ~4.8 mA $(V_S = 64\ V)$ for 1 min. The resistance is much smaller when the current is applied in the same direction as the processing one, which is defined as the forward direction. The asymmetric feature exists in a wide temperature range, and is especially strong at low temperature. From Fig. 2(a), we can see that the current starts to increase abruptly in the forward direction at a threshold voltage (opening voltage $V_{OP} \sim 2.3\ V$) at 30 K. In the backward direction, the current increases slowly with increasing bias and remains small at $V = -12\ V$.

To further investigate this asymmetric conduction, the R-T curves recorded at selected voltage bias in both directions were measured [Fig. 2(b)]. The R-T curve of the initial state is also presented for comparison. Under the reverse bias, the sample is completely insulating and the bias voltage has little influence on the resistance. In contrast, increasing bias voltage in the forward direction depresses the resistance significantly. The temperature coefficient of resistance is negative under low voltage bias and positive under high voltage bias. For $V \sim 2.2\ V$, the resistance is almost temperature independent. The resistance in either direction is larger than that of initial state, and no anomaly was found at T_P.

Interestingly, extending the processing duration led to steep resistance decrease. Since a constant V_S mode was used, the current and voltage across the microbridge also changed while the resistance dropped. Fig. 3 shows the time dependence of voltage, current and resistance. The resistance while processing first changed from $\sim\ 10000\ \Omega$ to $\sim\ 2000\ \Omega$. The I-V curves measured following this step are shown in Fig. 4. Compared with that in Fig. 2(a), these I-V curves are more asymmetric and less temperature dependent. Another salient change is the reduction of V_{OP}, which is ~0.8 V at 290 K and ~1.6 V at 50 K.

It is important to clarify whether these are dominated by the bulk or the interface between the electrode and the sample. The asymmetric conduction was first speculated as an interfacial effect [10]. However, careful rearrangements of electrodes configuration and redeposition of electrodes revealed that the changes took place within the film rather than at the electode-manganite interfaces [9], [11], [12]. In this study, due to the microbridge configuration, the current density near the electrodes is two orders smaller than that in the microbridge. The two lead I-V curves measured between electrode 1 and 2 [or between electrode 3 and 4, see the inset of Fig. 2(a)] after current processing are linear in the whole temperature range. Further, no change was found after redepositing the electrodes. As a matter of fact, silver was known to be a nonreactive metal when contacting with manganites [17]. Therefore, it is unlikely these effects are

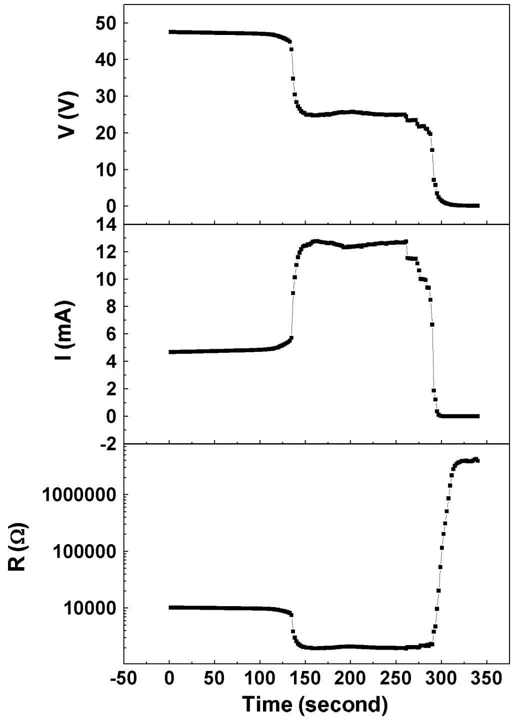

Fig. 3. Time dependence of voltage, current, and resistance while processing.

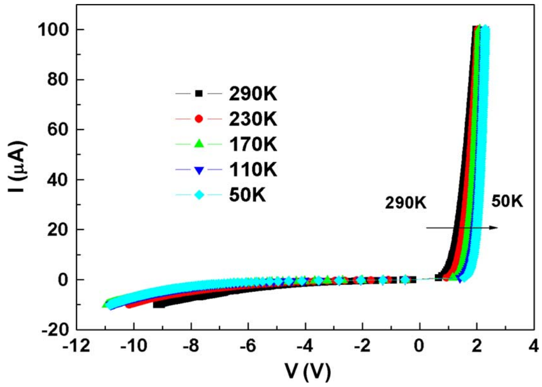

Fig. 4. I-V curves measured after the first resistance step.

mainly from the interfacial layer although changes induced by current processing may present in and near the interfaces.

The co-occurrence of abrupt resistance changes and asymmetric conduction implies the close relation of their underlying mechanisms. To understand these phenomena, it was noted that these steep resistance steps share some similarities with resistance switching studied in metal-oxide-metal structures [13], [14]. Currently, the exact origins of asymmetric conduction and resistance switching remain elusive. Proposed mechanisms for asymmetric conduction include double Schottky barriers at electrode-manganite interfaces [10], electric field/current affected charge orbital states [9] and oxygen electromigration at grain boundaries [11], [12]. To explain the resistance switching effects, various models, including carrier trapping/detrapping [18], defect creation/annihilation [19] and oxygen electromigration [20], have been suggested. The models proposed for both phenomena can be roughly categorized into two groups. One is based on pure electronic processes, and the other is based on processes involving lattice. Since both phenomena are rather universal, they are probably not dominated by a pure electronic process that depending on one particular detailed electronic structure. In present study, the appearance of asymmetric transport in the whole temperature range, as well as the weak temperature dependence of I-V curves, favor a model involving lattice rather than that of a pure electronic process. Although the detailed process is very complicated, it seems that imperfections caused by high electric field/currents, e.g., lattice distortion or bond breaking, take effect. The formation of percolation paths between such lattice sites is a possible origin for the first switching. In this framework, the asymmetric conduction could be understood as a consequence of space charge regions developed around these sites. If the accumulated imperfections reach a critical value, a hard breakdown may occur. That could be the reason that about two minutes after first step both voltage and current decrease to near zero while the resistance has a tremendous increase.

IV. SUMMARY

In summary, we studied current induced effects in epitaxial thin films of $\mathrm{La_{0.8}Ca_{0.2}MnO_3}$ fabricated by pulsed laser deposition. Asymmetric conduction was found when the processing current was larger than a certain value. Properly extending the processing duration led to abrupt resistance change, after which the I-V curves were more asymmetric and less temperature dependent. The co-occurrence of both phenomena implies the close relation of their underlying mechanisms.

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