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Electroresistance and field effects in epitaxial thin films of Pr$_{0.7}$Sr$_{0.3}$MnO$_3$

J. Gao · H.Y. Liu

Abstract Highly epitaxial thin films of Pr$_{0.7}$Sr$_{0.3}$MnO$_3$ were grown on (100) SrTiO$_3$ single crystal substrates by laser ablation. Similar to other manganite compounds, these Pr$_{0.7}$Sr$_{0.3}$MnO$_3$ films exhibited remarkable magnetoresistance. Application of electric currents could induce a remarkable reduction in resistivity, demonstrating a strong electroresistance effect. The ratio of the resistance variation, \( ER = \frac{R(0) - R(I)}{R(I)} \), is about 33% at metal-insulator transition temperature. Using a planar field effect configuration, significant field modulation of the metal-insulator transition was achieved. The observed field effects were discussed based on the strong interactions between carrier spins and localized spins in Mn ions, as well as the percolative mechanism of phase separation.

1 Introduction

Perovskite manganites R$_{1-x}$A$_x$MnO$_3$ (where R and A are rare- and alkaline-earth ions, respectively) are known as a typical system with a strong electron correlation. In such compounds, spin, charge, lattice, and orbital are strongly coupled, resulting in various ground states, such as the ferromagnetic metallic state and charge/orbital ordering state, etc. These ground states have comparable energies so that an external disturbance can easily drive the system from one state to another state, leading to a dynamic coexistence of phases with different magnetic and electronic properties. Much research effort has been devoted to understand the underlying physics and related phenomena like colossal magnetoresistance (CMR) and electroresistance (ER) effects [1–3]. It is generally agreed that the double-exchange interaction via spin-polarized conduction electrons is the main cause. External magnetic fields can align the magnetic moments of Mn ions, thereby enhancing the double-exchange transfer integral and decreasing the resistivity. The strong competition among the coexisted multiphase might also be responsible for their extremely rich phase diagram and may even lie at the core of the CMR and ER effects for manganites with different doping concentrations. Recent investigations proved that the equilibrium of multiphase coexistence can be influenced not only by a magnetic field but also by an electric field or current bias [6].

Since the magnetic field required for a significant MR is very high, and usually exceeds several Tesla, the prospect for a direct application of MR effect is not clear. In contrast, the practical feasibility to use electric fields/currents to modulate the insulator–metal (I–M) phase transition and transport properties of manganites is of great merit [4–7]. The compound Pr$_{1-x}$Sr$_x$MnO$_3$ is known being very sensitive to external disturbances, especially in the vicinity of percolation threshold [8–10]. It mimics the behavior of the La$_{1-x}$Ca$_x$MnO$_3$ system. In this paper, we report a study on the field/current effects in Pr$_{0.7}$Sr$_{0.3}$MnO$_3$ (PSMO) thin films grown by laser ablation. From the phase diagram, the selected doping concentration \( x = 0.3 \) makes the PSMO behaving as a ferromagnetic metal.

2 Experimental

The studied PSMO films were grown on (100) SrTiO$_3$ single crystal substrates by laser ablation. The target was made by standard solid state reaction from metallic oxides and
carbonates. The nominated composition of the target was $\text{Pr}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$. The deposition took place in oxygen of 0.5 mbar. The beam energy was $\sim 380$ mJ with the wavelength of 248 nm and pulse frequency of 2 Hz. The beam was focused by a lens, which produced an energy density $\sim 5$ J/cm$^2$ at the target. The distance between substrate and target was 6 cm. The substrate temperature was $75^\circ\text{C}$ measured by a $k$-type thermocouple inserted into the heater block. The film thickness was between 150 to 300 nm, controlled by the deposition time. The electric measurements were done by using the standard four-probe technique in a closed cycle cryostat. In order to apply a current with high density in the electric measurements, the films were patterned into a microbridge with the width of 50 μm and length of 200 μm using lithography technique. Field effect was studied using a planar field effect configuration (FEC), which is formed on a monolayer of PSMO [11].

### 3 Results and discussions

The grown thin films of PSMO are of highly epitaxy as characterized by X-ray diffraction and rocking curve measurements. The spectra of X-ray diffraction always reveal the sharp peaks of the formed $\text{ABO}_3$ phase with the c-axis perpendicular to the substrate surface (Fig. 1). Besides the reflections from substrate and the (00l) peaks of the PSMO, no peaks of secondary phases are visible. The value of full width at half maximum for rocking curve of the PSMO (002) peak is less than 0.2 degree, implying very good epitaxy and crystallinity formed in our films.

As a typical feature of doped manganites, the grown films show a clear phase transition from an insulator to a metal near the Curie temperature. Such a phase transition can be affected by oxygen deficiency. For the as-grown film, which is suspected being oxygen deficient, the transition peak is found $T_p \sim 210$ K. After annealed in 1 bar oxygen at $700^\circ\text{C}$ for 1 hour, the resistance of the film is reduced remarkably and the I–M transition temperature increase to over 250 K (Fig. 2). The resistance of the sample also decreased from $\sim 60$ kΩ to $\sim 7$ kΩ. The differences between as-grown and annealed films are attributed to the influence of oxygen defects on the phase separation. It is thought that the equilibrium between metallic phase and insulating phase in as-grown films is much unstable due to the oxygen deficiency. In addition, the resistance of as-grown films is one order larger than that of the annealed films. An external disturbance to as-grown films could impact the equilibrium and may result in a more significant reduction of the resistance. On the other hand, the transition temperature of the post-annealed PSMO films is slightly higher in comparing with that of its bulk ($\sim 245$ K). This is probably due to the strain introduced by the lattice mismatching between SrTiO$_3$ substrate and grown PSMO layer [9].
When a magnetic field is applied, these PSMO films showed a very strong magnetoresistance. As can be seen in Fig. 2, a magnetic field of 1.0 Tesla can induce a remarkable MR effect. The ratio of the resistance variation \( \{MR = (R(0) - R(H))/R(0)\} \) is found \( \approx 57\% \), which is more significant in comparing with some other CMR compounds \([5, 6, 8]\). The ER effect induced by electric field/current is known as a common feature for thin films of doped manganites \([6, 7, 11]\). The influence of the electric field/current on the transport properties of our PSMO films has also been explored. Figure 3 shows the temperature dependences of resistance for a 300 nm film of PSMO with different currents applied. It is found that the transition temperature peak, \( T_p \), remains nearly unchanged, whereas the resistance, especially the peak resistance, is modulated by the applied currents. When the current increases from 1 \( \mu \)A to 50 \( \mu \)A (the current density \( \sim 3.4 \times 10^5 \) A/cm\(^2\)), the peak resistance drops remarkably from \( \sim 105 \) k\( \Omega \) to 70 k\( \Omega \). The changing ratio of the peak resistance (defined as \( ER = (R(0) - R(I))/R(0) \)) reached 33\% under a current of 50 \( \mu \)A, which is also much pronounced than what we observed on \( La_{0.8}Ca_{0.2}MnO_3 \) and \( La_{0.9}Ba_{0.1}MnO_3 \) thin films \([6]\). To ensure that the obtained results are reliable, similar measurements were performed on a number of samples and all the results were consistent.

Such an ER effect is fully reversible as the transition curve could return to the initial state if the applied current is removed. Obviously, the observed ER effect cannot be caused by the loss of oxygen. On the other hand, the self magnetic field generated by the applied current also cannot be the main cause for the observed ER effect. Supposing the film is an infinitely large plane carrying a uniform current with a surface current density \( \sim 1 \times 10^3 \) A/cm. The magnetic field produced by the applied current is estimated \( \sim 10^{-2} \) T. Such a value of the self-field is far below the magnetic field required to cause a MR equivalent to 33\%.

The influence of a static electric field on the I–M transition has been studied as well. In our work, a very simple planar FEC formed on a monolayer film is employed (see the inset of Fig. 4). As such a FEC is formed in a monolayer, a transverse electric field is applied to the microbridge of PSMO. Significant modulation of the I–M transition was achieved. The modulation of the resistance for the PSMO channel upon a bias of +350 V (\( +1.75 \times 10^5 \) V/cm) reaches 44\%, whereas the modulation caused by a negative bias of \(-200 \) V (\( -1.0 \times 10^5 \) V/cm) is \(-18\% \). The results of field effect for PSMO channel are similar to that reported by Wu et al. and Gao et al. \([12, 13]\). The gate voltage applied to our planar FEC is remarkably larger in comparing with the results obtained on multilayer FEC. Actually we find that the field gradient, rather than the absolute field value, plays a more important role for the field effect. In our experiments, the bias applied to the sample is a transverse field with a spacing \( \sim 20 \) \( \mu \)m. The field gradient is therefore much smaller. Moreover, it is material dependent, e.g., the needed gate voltage is \(-80 \) V for a \( La_{0.7}Ca_{0.3}MnO_3 \) channel \([11]\).

The sign of the field modulation is found depending on the bias direction. Similar to what reported on multilayer FEC \([12]\), the resistance enhances when a positive gate voltage is applied and decreases with a negative gate voltage. Also, the magnitude of modulation is asymmetric with the bias polarity. When the gate potential sweeps between positive and negative directions, nonlinear dependence appears and no saturation trend is revealed. All measured films could return to their pristine state completely if the bias was removed.

At this moment, the underlying physics behind the observed field effects has not been well understood yet. In our opinion, such effects induced by the applied dc-current/field should be related to the percolative mechanism of phase separation \([11]\). A dynamic coexistence of phases with different magnetic and electronic properties is known as one of the most characteristic features of doped manganites. These coexisted phases have comparable energies so that external disturbances can easily drive the system transiting from one state to another. Application of an electric field/current in such an inhomogeneous system may change, by accumulation of charge, the relative volume fractions of the metallic and insulating phases. The boundary between metallic and insulating phases can be therefore moved. The direction of such interface shift is controlled by the polarity of the applied field. Application of a negative field shifts the I–M phase boundary to the insulating phase. It increases the fraction of metallic phase, hence reduces the resistance. Whereas by moving the I–M interface to the metallic site, a positive field may enhance the resistance. More clear understanding of the observed effects needs more detailed studies.
4 Conclusion

The effects of electric fields/currents on the I–M phase transition and transport properties of Pr$_{0.7}$Sr$_{0.3}$MnO$_3$ epitaxial thin films have been investigated. These films are epitaxially grown with c-axis perpendicular to the film surface and show a good crystallinity, as revealed by X-ray diffraction and rocking curve measurements. Application of electric currents to these PSMO films causes remarkable changes in their resistance, demonstrating a strong ER effect. The observed ER ratio of the peak resistance reached 33% with a current of 50 $\mu$A, which is much pronounced than what we observed on La$_{0.8}$Ca$_{0.2}$MnO$_3$ and La$_{0.9}$Ba$_{0.1}$MnO$_3$ thin films. Significant modulation on the I–M transition peak by a static electric field was obtained using a simple planar FEC. The sign of the field modulation changes with the polarity of gate voltage, which is similar to what reported on multilayer FEC. Such ER and field effects might be related to the percolative mechanism of phase separation.

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