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Electroresistive effects in electron doped manganite $La_{0.7}Ce_{0.3}MnO_3$ thin films

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Abstract

The influence of electric current on the transport behaviour of $La_{0.7}Ce_{0.3}MnO_3$ thin epitaxial films on (001) $LaAlO_3$ substrate is studied. Measurements were carried out in the regime of low current densities (0.0325–32.5 A cm^{-2}), for dc current in the absence of magnetic field. Significant reduction of the peak resistance (R_p) with monotonic increase in the metal–insulator transition temperature (T_p) was found with increasing bias current. The voltage current characteristics at various temperatures above and below T_p show nonlinearity for small currents. The behaviours of the resistance with the current are similar at various temperatures: an initial decrease with increasing current and then a levelling off. The magnetoresistance (MR) measurement under a field of 1 T revealed that it is maximum at the temperature near T_p at which the electroresistance (ER) is also maximum. The variation of resistance with electric field and magnetic field is non-hysteretic. We explain the origin of the observed ER and MR in our samples on the basis of double-exchange interaction and the phase separation phenomenon.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The perovskite material $R_{1-x}A_xMnO_3$ (R being trivalent rare earth ions) can be either hole doped or electron doped depending on whether A is a divalent alkaline earth ion such as Ca, Ba, Sr, Pb etc or tetravalent such as Ce, Sn etc. Both classes of this material exhibit a rich phase diagram as a function of temperature, magnetic field and doping, and show interesting phenomenon like colossal magnetoresistance (CMR) and metal–insulator transition [1, 2].

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Double-exchange interaction via spin polarized conduction electrons is believed to be the main cause of CMR. In hole doped manganites there is strong experimental evidence that there exist ferromagnetic clusters with size ranging from a few Å units to a fraction of a micron, above and below the Curie temperature [3–8]. This phase separation which gives rise to a percolation behaviour in transport is an intrinsic feature of hole doped manganites and is even believed to play a central role in the origin of CMR. Because of the coexistence of different phases in equilibrium, the physical properties of hole doped materials can be tuned by external agencies such as magnetic field, pressure, light illumination and electric current [9–18].

Recently the focus has been on the influence of electric field and electric current on hole doped CMR materials for their technological applications [19–24]. In hole doped and charge ordered manganites, it has been observed that the increasing electric current forces the resistivity to drop sharply, leading to colossal electroresistance (CER) [15, 16, 25]. Application of electric current has been reported to lead to a transition from the electrically insulating charge ordered (CO) state to the ferromagnetic metallic state for $Y_{0.5}Ca_{0.5}MnO_3$ in which the CO state is very robust, unaffected by a magnetic field up to 40 T [15]. The current bias can significantly influence the balance of multiphase coexistence and a current higher than a critical value may induce a new equilibrium state of coexistence [25]. Correlation between CER and CMR has been established in $La_{0.82}Ca_{0.18}MnO_3$ single crystal having heterogeneous structure [16]. Phase separation is believed to be the main cause of both CER and CMR. In the majority of the studies reported on hole doped manganites the phenomenon of CER is attributed to the phase separation [17, 18, 25].

In this paper we report on the effect of electric current on the transport properties of electron doped manganite $La_{0.7}Ce_{0.3}MnO_3$ (LCeMO). This material is known to exist in the single phase only in the form of thin epitaxial films prepared by the pulsed laser deposition method [2]. The x-ray absorption spectroscopy studies performed on $La_{0.7}Ce_{0.3}MnO_3$ thin films confirmed that cerium exists in a tetravalent state and the manganese is in a mixture of Mn^{2+} and Mn^{3+} valence states in this material [26]. Electron doped manganites have not been as intensively studied as compared to their hole doped counterparts. There are so far no reports on the current effects in these materials. In this study we observed an interesting feature that the metal–insulator transition temperature increases monotonically with increasing current, which has not been observed in numerous studies carried out so far on hole doped manganites. The peak resistance is found to be sensitive to small currents. We were able to identify the threshold current at which the Joule heating dominates at various temperatures and measurements of ER were performed at currents lower than this threshold. To verify the reliability of the measurement, the measurements were carried out on several samples and the results obtained were consistent. We also performed the measurements on hole doped $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) thin films having same thickness as the LCeMO films, for comparison.

2. Experimental details

The $La_{0.7}Ce_{0.3}MnO_3$ films were grown on (001) $LaAlO_3$ (LAO) substrate by the pulsed laser deposition technique. The sample preparation and characterization are described elsewhere [27]. The resistance was determined as a function of temperature for various dc currents in the absence of magnetic field by using a conventional four-probe method in a closed cycle cryostat. A Keithley 220 current source and a Keithley 196 digital multimeter were used for current and voltage measurements. Four gold contacting pads were deposited on the film. Current and voltage leads were connected to gold pads by using In–Ag (5% Ag in In) solder for good and stable ohmic contacts. Magnetoresistance was recorded at a constant current of 1 μA under a magnetic field of 1 T and field parallel to current. The LCMO films were grown

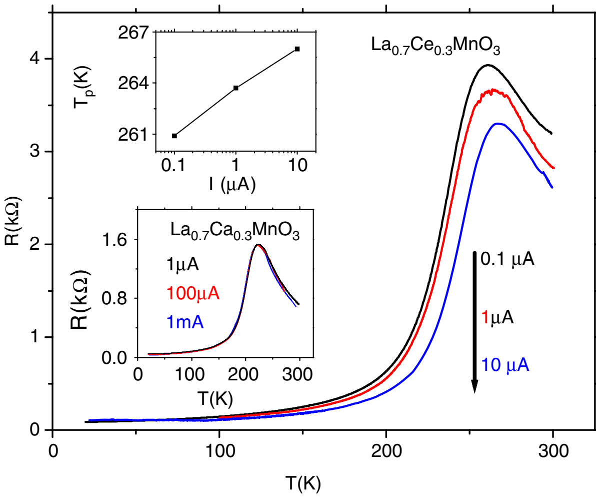

Figure 1. Temperature dependence of the resistance of $La_{0.7}Ce_{0.3}MnO_3$ thin films for different values of the bias current. Upper inset: variation of the metal–insulator transition temperature with the bias current. Lower inset: temperature dependence of the resistance of $La_{0.7}Ca_{0.3}MnO_3$ thin films for different values of the bias current.

on (001) LAO substrate by pulsed laser deposition. The energy density for LCMO films was 2.5 J cm^{-2}, at the oxygen pressure of 400 mTorr during the growth.

3. Results and discussion

The LCeMO films were characterized by energy dispersive spectroscopy and x-ray diffraction analysis, described elsewhere [27]. The thickness of the films is 120 nm. In figure 1 temperature variation of the resistance (R–T) is shown for different bias currents, under zero field. As can be seen, the resistance is very sensitive to small currents with peak resistance decreasing as the current increases from 0.1 to 10 μA corresponding to current densities of 0.0325 to 3.25 A cm^{-2} respectively. The striking feature is an increase in T_p from 260 to 266 K with increase in the bias current (upper inset in figure 1). This feature has not been observed in earlier studies on hole doped LCMO and LBMO films [18, 25] and their heterostructures [28, 29]. The electrical resistance saturates for current >10 μA. There is no significant change in the resistance with current at temperatures below 150 K. Further, there is no thermal hysteresis in heating and cooling curves. The lower inset in figure 1 shows the temperature dependence of the resistance for LCMO films. There is a small drop in the peak resistance with T_p shifting towards lower temperature as current is increased from 1 μA to 1 mA corresponding to current densities of 0.307 to 307 A cm^{-2} respectively.

Figure 2 shows the voltage versus current (V–I) characteristics of LCeMO films at various temperatures. The V–I curves are nonlinear over a current range from 0.1 μA to between 6 and 8 μA (inset in figure 2). Another nonlinearity is observed at high currents (indicated by arrows), which is due to Joule heating. We were able to identify the onset of Joule heating as a knee in the V–I characteristics at various temperatures. Above this knee current, the voltage saturates and the applied current was restricted to being below this threshold value for measurement of the ER. The Joule heating was also confirmed by a thermometer placed very near to the sample, from the rapid increase in the temperature. The onset current for Joule heating depends on the

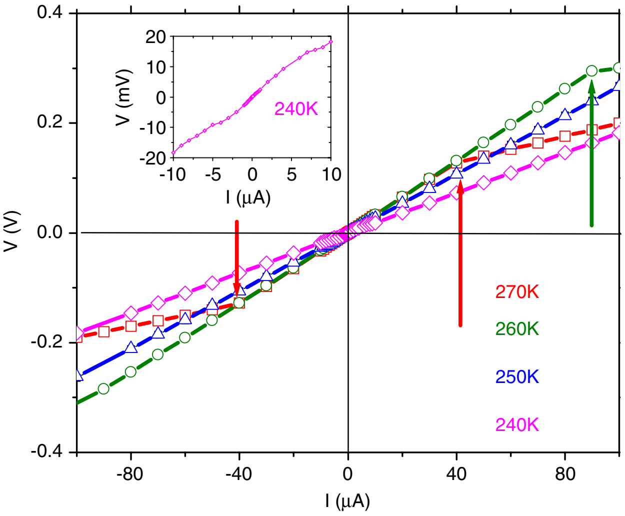

Figure 2. Voltage–current characteristics of $La_{0.7}Ca_{0.3}MnO_3$ thin films for different temperatures. The arrows in the figure indicate the onset of Joule heating. Inset: nonlinear behaviour of the voltage for small currents at 240 K.

resistance of the sample and is lowest (~50 μA) close to T_p. For example at 270 K the voltage saturates above 50 μA, while at 260 K it saturates above 100 μA. The variation of the resistance with the bias current at different temperatures as determined from V–I characteristics is shown in figure 3(a). The resistance decreases initially as the current increases and then levels off at currents above 10 μA (figure 3(b)). Like in the V–I curves, Joule heating can be seen quite distinctly at much higher currents in the range of 50–100 μA (indicated by arrows). The power dissipated at 270 K at the onset of the Joule heating is about 20 μW.

The ER behaviour of LCeMO is in contrast with that for LCMO where a small drop in the peak resistance with increasing current is observed but T_p shifts towards low temperatures, consistent with the previous reports [18, 25]. Earlier ER studies on hole doped manganites like LCMO and LBMO are reported at very high current densities [18, 25, 30]. The Joule heating is a serious issue at such high currents. We varied the current density in the case of LCMO from 0.307 to 307 A cm^{-2}. Joule heating is seen in V–I characteristics of LCMO films at applied currents greater than 1 mA. The maximum ER at temperature 215 K near to T_p is 6.6% when the current density is changing from 0.307 to 307 A cm^{-2} (by a factor of 1000, the same as for $La_{0.7}Ce_{0.3}MnO_3$ films). The values of ER calculated from R–T curves and V–I curves are consistent within 10%.

To compare the ER behaviour with MR observed in these samples, in figure 4(a) we have shown the resistance versus temperature for LCeMO measured under zero field and a field of 1 T at a constant current of 1 μA. There is a decrease in the peak resistance associated with an increase in T_p from 263 to 272 K. %MR and %ER calculated as $\%MR = 100 \times [R_{(H=0)} - R_{(H=1\,T)}]/R_{(H=0)}$ and $\%ER = 100 \times [R_{I_1} - R_{I_2}]/R_{I_2}$ where $I_1 = 0.01\ \mu A$ and $I_2 = 10\ \mu A$ are both maximum at 240 K (figure 4(b)). The variation of the resistance either with the bias current or the magnetic field is non-hysteretic.

The striking similarity between the MR and ER behaviours suggests a similar origin for these two phenomena for LCeMO films. Though the physics behind CER and CMR effects is far from being completely understood, double-exchange interaction is commonly adopted as the main cause. In the completely ferromagnetic phase, e_g (conduction) electrons of Mn^{3+} can

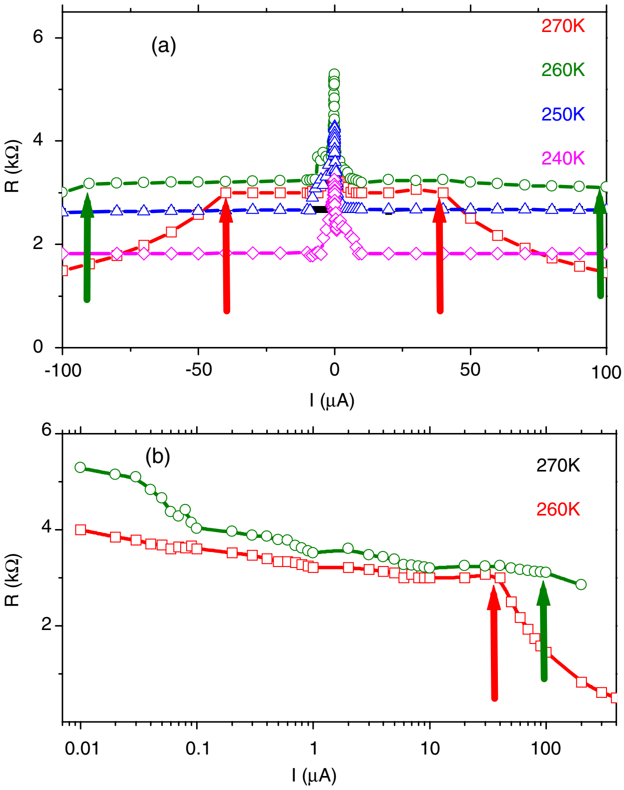

Figure 3. (a) Resistance as a function of bias current for $La_{0.7}Ce_{0.3}MnO_3$ thin films for different temperatures. The arrows in the figure indicate the onset of Joule heating; (b) expanded view of (a) for two values of the temperature. The current is plotted on a logarithmic scale. The arrows in the figure indicate the onset of Joule heating.

hop coherently without magnetic scattering by t_{2g} (localized) spins, while they become strongly incoherent if the t_{2g} spins are disordered. The reduction in peak resistance and increase in T_p in the present work could possibly arise due to strong interaction between carrier spins and localized spins in Mn ions caused by the field. The carrier spins are forced to become parallel to localized spins on the same site due to a strong Hund's rule coupling. The hopping rate of carriers gets enhanced if the localized spins on the nearest neighbours are parallel. Under the applied electric current the conduction electrons are forced to move, irrespective of their spins being parallel or not to the localized spins. However the spin exchange coupling will force the localized spins to be parallel. Alignment of localized spins would reduce the spin scattering of conduction electrons, thus increasing the hopping rate of the conduction electrons and magnetic coupling among Mn ions and as a consequence reducing R_p and shifting T_p towards higher temperature. However, we would like to point out that while the double-exchange mechanism outlined above is likely to be the root cause of our observed CER, this mechanism alone cannot explain the extreme sensitivity of the ER effect to very small current and its relative insensitivity to moderately high currents. Also this general mechanism of ER should be equally applicable to $La_{0.7}Ca_{0.3}MnO_3$ where we do not see the large ER at small current. This is hardly surprising given that even the better understood phenomenon of colossal magnetoresistance could not be explained on the basis of double exchange alone. Since the number of electrons hopping

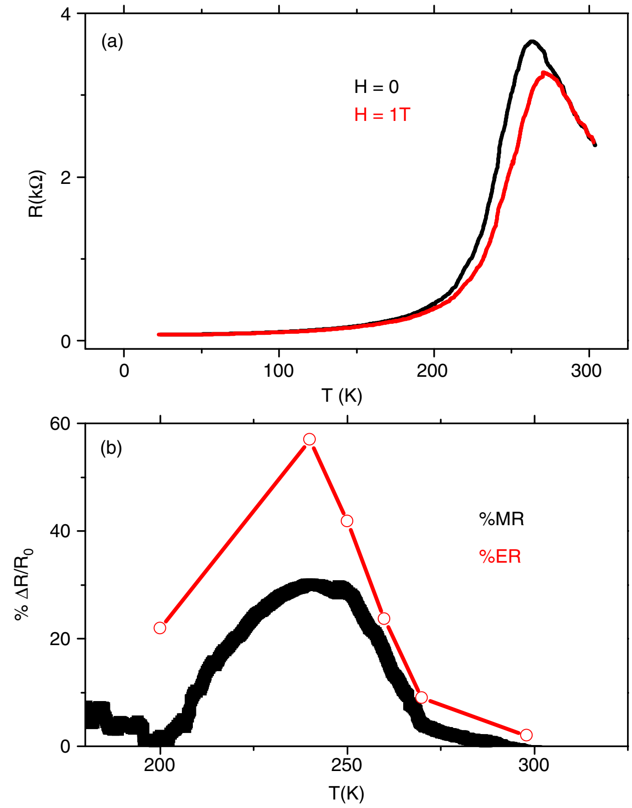

Figure 4. (a) Temperature dependence of the resistance of $La_{0.7}Ce_{0.3}MnO_3$ thin films for different values of the magnetic field. (b) Comparison of the temperature dependences of the electroresistance and magnetoresistance of $La_{0.7}Ce_{0.3}MnO_3$ thin films.

from Mn^{2+} to Mn^{3+} at a current density of 3.25 A cm^{-2} is a tiny fraction of the number of Mn^{2+}–Mn^{3+} adjacent sites present in this system, the ER is expected to continue to much higher currents if the current distribution is uniform at nanometre length scales. However, this high sensitivity could be explained if the sample is phase separated with conduction channels with higher conductivity where the current density is much larger than the average current density in the sample. When the response in these channels saturates, the conductivity of the sample will show no further enhancement. A confirmation of this conjecture will require further studies such as the measurement of the magnetic response in the presence of current as well as microscopic evidence of phase separation in this system. In addition, the role of orbital interactions and the polaron effect on the electroresistive properties also needs to be explored for this material [31, 32].

4. Conclusion

In summary we have investigated the temperature behaviour of the resistance for different applied dc currents of $La_{0.7}Ce_{0.3}MnO_3$ thin films. We observed that the ER is maximum (56%) at 240 K, which is near to the metal–insulator transition temperature, and a shift of T_p towards the higher temperature side. We also observed similar behaviour of the resistance under a magnetic field at constant applied bias current. The striking similarity of the temperature dependences of ER and MR suggests that the two effects arise from similar origins.

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