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Correlation between effects of electric current and magnetic field on transport properties of electron-doped manganite $\text{La}_{0.7}\text{Ce}_{0.3}\text{MnO}_3$ thin films

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Abstract

We report the effect of electric current and magnetic field, separately and in conjugation, on the transport behaviour of patterned $\text{La}_{0.7}\text{Ce}_{0.3}\text{MnO}_3$ thin films. In the absence of a magnetic field, a significant reduction in peak resistance (R_p) was found with increasing bias current. This effect is also present when a magnetic field is applied, though the magnitude of the electroresistance ($\text{ER} = [R(I = 0.05 \mu\text{A}) - R(I = 50 \mu\text{A})]/R(I = 50 \mu\text{A})$) decreases. The metal-insulator transition temperature (T_p) increases both with increasing current and with magnetic field. We observe an interesting correlation between electric current and magnetic field: the magnetoresistance ($\text{MR} = [R_{H=0} - R_{H=1 \text{ T}}]/R_{H=0}$) decreases with increasing bias current, while ER decreases with increasing magnetic field. Both ER and MR show a maximum near T_p . The interesting correlation between these two effects suggests that both these effects arise from the same origin.

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

One of the major challenges in doped rare-earth manganites is to understand the large change in their electronic properties at high temperatures (100–300 K) that arise from relatively small perturbations. The most celebrated of these effects is the phenomenon of colossal magnetoresistance (CMR), where the resistivity of the materials such as $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ and $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ decreases by an order of magnitude close to the ferromagnetic transition temperature, under the application of a field of a few Tesla. These materials however have been shown to exhibit large response to other perturbations as well, such as chemical and hydrostatic

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pressure [1] and electric field [2, 3]. Recently it has been shown that the resistivity of these materials can also be tuned by passing an electric current [4–7]. This effect, commonly termed as electroresistance (ER), has mostly been studied in weakly hole-doped and charge-ordered manganites where large changes in resistance with transport current have been observed [8–11]. In optimally hole-doped manganites exhibiting a clear metal–insulator transition the ER effect at low currents has been observed to be weak [12].

Despite several studies on the ER effect in hole-doped manganite thin films and single crystals the origin of this effect has remained unclear. Simultaneous investigation of ER and MR effect in hole-doped manganites [8, 12] revealed striking similarities between the effect of magnetic field and transport current on the electrical resistance, suggesting a common origin for the ER and MR. However, studies on optimally hole-doped manganites also revealed that the metal–insulator transition temperature (T_p) shifts towards lower values with increasing transport current, which is in direct contrast with the increase in T_p under the application of magnetic field [9]. The weak ER effect in the optimally doped material however required these measurements to be carried out at large current densities ($\sim 10^4$ A cm $^{-2}$), where the effect of self-heating cannot be ruled out.

In a recent paper [13] we reported large ER effects in thin films of the electron-doped manganite La $_{0.7}$ Ce $_{0.3}$ MnO $_3$ (LCeMO), where manganese is in a mixture of Mn $^{2+}$ and Mn $^{3+}$ valence states [14]. The magnetic and magnetotransport properties of this compound are very similar to its hole-doped counterpart La $_{0.7}$ Ca $_{0.3}$ MnO $_3$ and are believed to arise from the double-exchange interaction between the Mn $^{2+}$ and Mn $^{3+}$ ions [14]. However, in contrast to hole-doped manganites the electron-doped manganite was found to be much more susceptible to small currents [13], allowing these measurements to be carried out at moderately low current densities. In addition to the strong similarity between the temperature dependences of MR and ER in LCeMO we observed that T_p increases with increasing current. In this paper we explore the coupling between the ER and MR effect by studying the ER under the application of magnetic field and the MR at different currents. We observe a strong coupling between the effect of magnetic field and transport current: ER decreases with applied magnetic field, while MR decreases with increasing measuring current, and both ER and MR are maximum near T_p . These results provide strong indication of a common origin based on the basic framework of double exchange for the MR and ER effects.

Thin films of LCeMO were synthesized on (001) LAO substrate using the pulsed laser deposition technique. The synthesis of films and structural details of these films have been reported elsewhere [15]. The XRD pattern revealed that the films are of single phase, and are oriented along the c -axis⁵. The thickness of the film was ~ 39 nm, measured by using a Dektak profilometer. In the present work the LCeMO films were patterned into a 200 μ m wide 10 mm long strip line using conventional photolithography in order to ensure homogenous current distribution in the film. Current density was changed from 0.641 to 641 A cm 2 . Four gold contacting pads were made on the stripline patterned films. Current and voltage leads were connected by using In–Ag (5% Ag in In) solder for good and stable ohmic contacts. The resistance was measured as a function of temperature for various currents and magnetic fields by using a conventional dc four probe method in a continuous flow helium cryostat. The magnetic field was applied parallel to current using a conventional electromagnet.

To get the broad picture of electroresistance and magnetoresistance we have first measured resistance at various currents in zero and 1 T magnetic field as a function of temperature (R – T characteristics). Measurement was done during the warming up process and the heating

⁵ We cannot however rule out the possibility of nanocrystalline CeO $_2$ precipitates reported in some studies from transmission electron microscope data; e.g. [16].

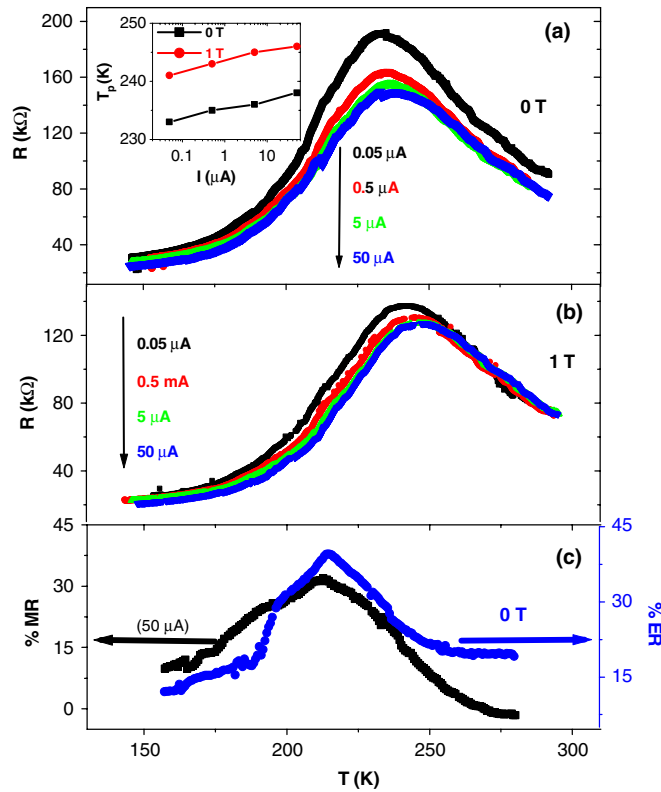


Figure 1. (a) Temperature dependence of resistance of the $\text{La}_{0.7}\text{Ce}_{0.3}\text{MnO}_3$ microbridge at various currents in zero field; (b) temperature dependence of resistance at various currents in magnetic field of 1 T; (c) magnetoresistance ($\text{MR} = [R_{H=0} - R_{H=1\text{T}}]/R_{H=0}$) (measured with transport current of $50\ \mu\text{A}$) and electroresistance ($\text{ER} = [R(I = 0.05\ \mu\text{A}) - R(I = 50\ \mu\text{A})]/R(I = 50\ \mu\text{A})$) (in zero field) as a function of temperature. The inset of (a) shows the variation of T_p with current with and without field.

rate was maintained at $3\ \text{K}\ \text{min}^{-1}$ during the measurements. The temperature variation of the resistance at various currents in zero magnetic field is shown in figure 1(a). We observe a significant reduction in peak resistance (R_p) as current increases from 0.05 to $0.5\ \mu\text{A}$ corresponding to current densities of 0.641 and $6.41\ \text{A}\ \text{cm}^{-2}$ respectively, as seen from figure 1(a). More interestingly, for the same range of current densities, T_p (inset of figure 1(a)) increases with increasing current density, consistent with our previous report [13]. This increase in T_p has not been observed in ER studies on optimally doped hole-doped manganites, where it has been reported to either remain constant or decrease due to Joule heating [9–11]. Figure 1(b) shows the same set of measurements in an applied field of 1 T. Though the trend remains the same, even a visual inspection shows that the relative decrease in R_p with increasing currents is much less in this case, showing the current is less effective in decreasing the resistance in the presence of a magnetic field. T_p continues to shift towards higher temperatures in the applied magnetic field also (inset in figure 1(a)). Figure 1(c) shows the temperature dependence of MR ($= [R_{H=0} - R_{H=1\text{T}}]/R_{H=0}$) and ER ($= [R(I = 0.05\ \mu\text{A}) - R(I = 50\ \mu\text{A})]/R(I = 50\ \mu\text{A})$) as determined from R - T characteristics. Both MR and ER are maximum at a temperature near to T_p .

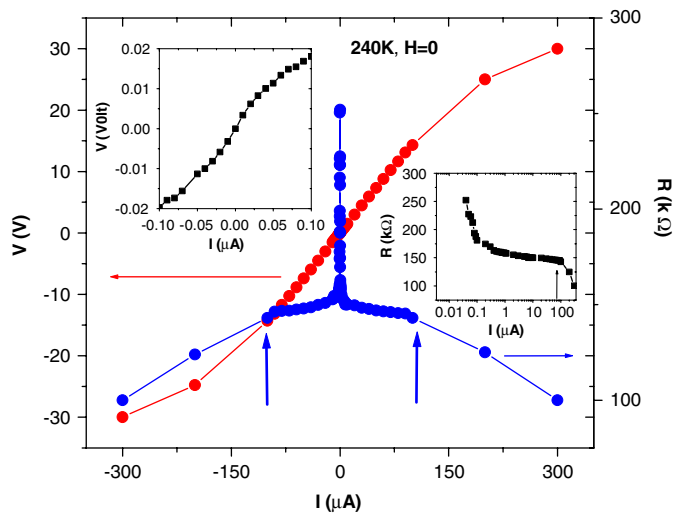


Figure 2. Variation of voltage and resistance with bias current at 240 K and in zero magnetic field. Vertical arrows indicate onset of Joule heating. The upper inset on the right-hand side shows nonlinearity in I - V curves for small voltages. The lower inset on the left-hand side shows the variation of resistance with current, where the current is plotted on a logarithmic scale.

Since in ER measurements the current is varied over several decades, it is of paramount importance to rule out the possibility of Joule heating due to the transport current in the range of current over which the measurements are carried out. In our measurements we rule out this possibility in two ways. First, we identified the onset of Joule heating as a 'knee' in the current versus voltage (I - V) characteristics at various temperatures (shown with an arrow in figure 2), above which the voltage saturates. The applied current was restricted to less than half of this 'knee' current for the actual measurements. The Joule heating was also confirmed by the thermometer placed very near to the sample from a rapid increase in the temperature for current values above this 'knee'. The onset current for Joule heating depends on the resistance of the sample and is lowest ($\sim 100 \mu\text{A}$) close to $T_p \sim 240 \text{ K}$. For example at 269 K the voltage saturates above $200 \mu\text{A}$, while at 295 K it saturates above $350 \mu\text{A}$. This shows that we can pump in nearly 4.5 times more power at 290 K ($\sim 9.2 \text{ mW}$) as compared to 240 K ($\sim 2.1 \text{ mW}$) without observing significant heating at 295 K. This is because in a continuous He flow cryostat the cooling power depends on the flow rate of l He. Typically, during measurement in the temperature range 210–300 K the flow rate of l He is kept constant and the temperature is controlled by heating against cooling with a heater fitted to the sample probe. Since the heat given by the heater is small at lower temperature a small additional heat causes the sample to heat up. At higher temperatures the temperature is stabilized by providing a much larger amount of heat with the sample probe heater and the same amount of heat generated by the sample has little effect. It requires much more heat to be dissipated by the sample to see any effect of heating. However, since the onset of Joule heating can be determined only within an accuracy of $\pm 10 \mu\text{A}$, we performed all our measurements up to $50 \mu\text{A}$, which is half the value of the onset current for Joule heating at T_p . The second proof that the effect observed in this experiment is not dominated by Joule heating comes from the variation of T_p with current. In the case of Joule heating, since the heating is local, the local temperature of the sample will rise faster than the sensor. In this case the sample temperature will be higher than the temperature recorded on the sensor and therefore T_p should show an apparent decrease with increasing current. The increase of T_p with current reinforces the conclusion that the current induced

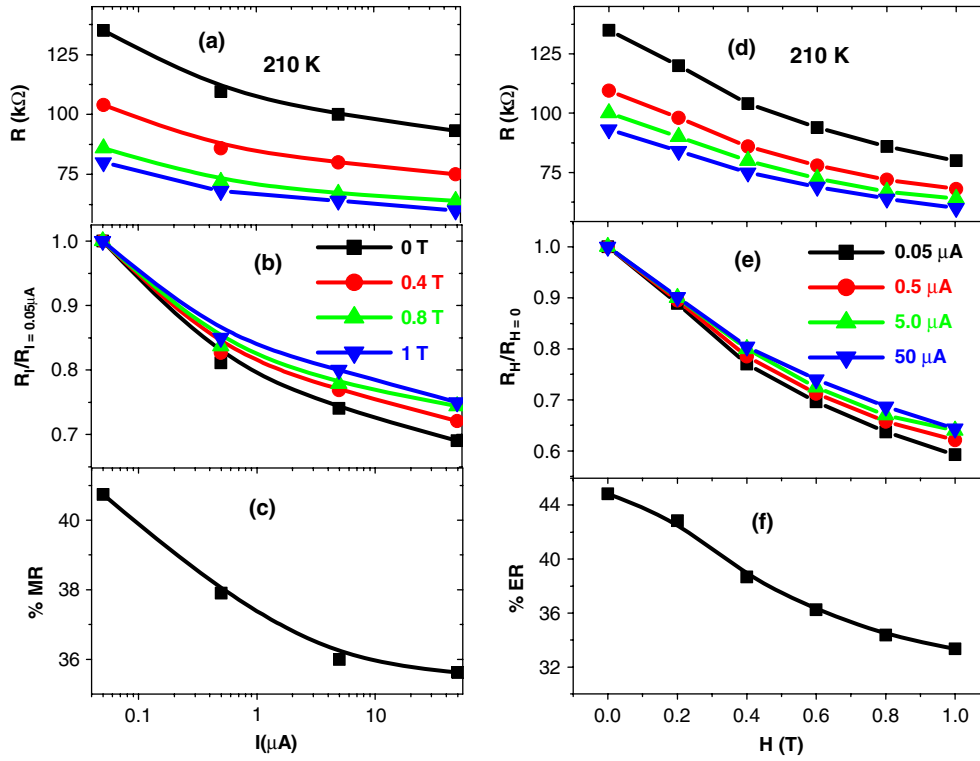


Figure 3. Variation of (a) resistance and (b) normalized resistance as a function of current at various values of magnetic field measured at 210 K. (c) Variations of magnetoresistance (defined as $MR = \frac{R_{H=0} - R_{H=1 T}}{R_{H=0}}$) with transport current. The current is plotted on the logarithmic scale. Variation of (d) resistance and (e) normalized resistance with magnetic field at different transport currents measured at 210 K. (f) Variation of electroresistance (defined as $ER = \frac{R(I=0.05 \mu A) - R(I=50 \mu A)}{R(I=50 \mu A)}$) with magnetic field.

effect is indeed an intrinsic one. The ‘intrinsic’ nonlinearity observed in I – V curves for small values of voltage is shown in the upper inset of figure 2. The nonlinearity and sensitivity of resistance for small currents is depicted in the lower inset of figure 2.

To investigate the coupling between the effect of current and magnetic field the resistance of the film was measured at different currents as a function of magnetic field at various temperatures. Magnetic field was varied between 0 and 1 T. Figure 3 shows the variation of resistance with electric current and magnetic field at 210 K. Figures 3(a) and (b) show the decrease in resistance (R) and normalized resistance ($R(I)/R_{I=0.05 \mu A}$) with applied current (I) at various magnetic fields. Figure 3(c) shows the MR ($= [R_{H=0} - R_{H=1 T}] / R_{H=0}$), as a function of increasing current. Figures 3(d) and (e) depict variation of resistance and normalized resistance ($R(H)/R_{H=0}$) with magnetic field (H) at various currents. Figure 3(f) shows the ER ($= R(I = 0.05 \mu A) - R(I = 50 \mu A) / R(I = 50 \mu A)$) as a function of magnetic field. Figures 3(c) and (f) together clearly demonstrate the central result of this paper: with increasing transport current the MR is suppressed, whereas with increasing magnetic field the ER is suppressed. This effect remains qualitatively the same above and below the metal–insulator transition temperature, as can be readily seen from figures 4(a) and (b). MR and ER values determined from R – T characteristics are consistent within 5% with those determined from I – V characteristics.

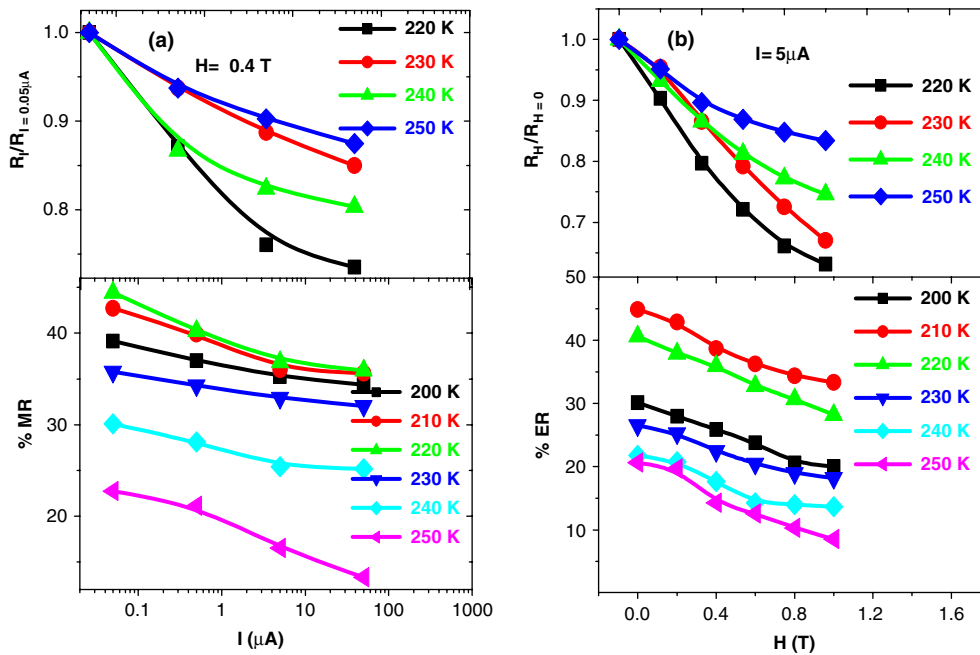


Figure 4. (a) Variation of normalized resistance and magnetoresistance at 0.4 T with current at various temperatures. Current is plotted on a logarithmic scale. (b) Variation of normalized resistance and electroresistance ($ER = \frac{R(I=0.05 \mu A) - R(I=50 \mu A)}{R(I=50 \mu A)}$) with magnetic field at various temperatures.

The observed similarity in the effects of transport current and magnetic field on the electrical resistance in the present work strongly suggests a similar origin for both the effects. A natural explanation of this effect would be from the basic mechanism of double exchange. The basic mechanism of Zener [17] double exchange follows from the fact that the spin of the electron is conserved while it hops from Mn^{2+} to a neighbouring Mn^{3+} via the intermediate oxygen. Thus if the spins of the two manganese ions are not parallel there is an energy loss of the order of the Hund's rule energy for the hopping process to happen. Thus the alignment of the manganese spins in a system where the manganese is in a mixture of two valence states, results in an increase in the hopping probability of an electron between adjacent Mn^{2+} and Mn^{3+} via the intermediate oxygen, thereby increasing the conductivity of the overall matrix. It follows from the principle of Zener double exchange that the converse should also be true: passing a current in such a system where the manganese spins are not all aligned would result in an increase in the potential energy associated with hopping of an electron from a Mn^{2+} to a neighbouring Mn^{3+} ion. The system would therefore rearrange itself to align the manganese ions, thereby increasing the overall conductivity of the matrix. This basic mechanism would explain most of our observation, namely

- (i) the peak in the ER at temperatures close to T_p where the spin disorder is largest;
- (ii) the increase in T_p with increasing current and
- (iii) the decrease in the ER under applied magnetic field where the spins are already partially aligned by the application of magnetic field.

It is however to be noted that the similarity between the effect of magnetic field and transport current should not be stretched too far: the first one is an equilibrium response of the system to

an applied magnetic field while the second one is a dynamic response to an applied transport current. The effect of magnetic field and transport current in our experiment is analogous to the 'spin valve' and the 'spin torque' effect in magnetic multilayers, where the magnetization reversal between two ferromagnetic electrodes can either be induced by magnetic field or electrical current respectively.

Though the basic mechanism in terms of Zener double exchange can explain our observations at a qualitative level, it cannot explain all the aspects observed in the data. In particular this simplistic picture would not explain the extreme sensitivity of the ER at very low currents and its moderate insensitivity at relatively higher currents ($>10 \mu\text{A}$). Even for measuring currents greater than $10 \mu\text{A}$ the MR at 1 T is significant, showing that a large fraction of the manganese spins are still not aligned. This model also does not explain why the ER tends to level off at about 18% at temperatures much higher than T_p . We would also like to note that this general mechanism of ER should be equally applicable to $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$, where we do not see the large ER at small current [13]. It is therefore likely that the magnitude of the ER effect crucially depends on the microscopic details of the system under investigation. This is hardly surprising given the fact that even the more well understood phenomenon of CMR and the metal-insulator transition cannot be explained based on double exchange alone. In hole-doped manganites there is strong experimental evidence that close to the Curie temperature the system gets spontaneously phase separated at submicron length scales with regions of high conductivity coexisting with regions of low conductivity [18–23]. This phase separation, which gives rise to a percolation behaviour in transport, is believed to play a central role in the origin of CMR. We could speculate that in our experiments, at microscopic length-scales the current is non-uniform, flowing through filamentary paths. In addition, such a filamentary conduction would produce intense magnetic field in its vicinity, which has to be further taken into account. In such a situation, the entire bulk of the sample would not respond uniformly to the applied transport current and the magnitude of ER would depend on the structure and length-scale of such phase separated domains. However, in order to extend our explanation beyond the simple minded picture we need to obtain microscopic details on the nature of current flow using local probes. We believe that our clear demonstration of the correlation between the ER and MR effects will motivate further studies to understand this effect at a more quantitative level.

In summary, we have demonstrated that electrical resistance of the electron-doped manganite $\text{La}_{0.7}\text{Ce}_{0.3}\text{MnO}_3$ can be influenced by both the magnetic field as well as the electric current and the two have similar kinds of effect on the transport properties. Furthermore, we have shown that the effects of electric current and magnetic field are coupled: application of a magnetic field causes a decrease in the electroresistance whereas increasing the transport current decreases the magnetoresistance. We believe that the clear elucidation of this coupling forms an important first step in the understanding of the ER effect at a more microscopic level.

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