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Electric current-induced first-order effects on the insulator–metal transition and the colossal electroresistance in rare-earth manganates

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Passing electric currents through a single crystal of $La_{0.77}Ca_{0.23}MnO_3$ causes a marked decrease in the temperature of the insulator–metal transition, T_{IM}, the T_{IM} decreasing with increasing current. The transition exhibits thermal hysteresis, the magnitude of which increases with an increase in applied current. Large negative electroresistance is observed close to room temperature while large positive electroresistance occurs at low temperatures. Similar results are observed with $La_{0.9}MnO_3$ as well. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1595137]

Rare-earth manganates of the formula $Ln_{1-x}A_xMnO_3$ (Ln = rare earth, A = alkaline earth) exhibit a variety of fascinating properties including colossal magnetoresistance (CMR), charge ordering, and electronic phase separation.[1–3] All of these aspects have been investigated extensively in the last few years. Subjecting the rare-earth manganates to external magnetic fields give rise to CMR in certain compositions, and melt the charge-ordered insulating state to the ferromagnetic metallic state in some others.[1,2] An aspect of particular interest to us is the effect of electric fields on the rare-earth manganates. It has been shown that passing moderate electric currents through thin films and single crystals of charge-ordered manganates changes the resistivity behavior, bringing about insulator–metal (IM) transitions.[4–8] Even relatively small currents are found to cause a small decrease in the resistivity in thin films of $La_{0.7}Sr_{0.3}MnO_3$.[9] Current-induced metastable resistive states occur in $La_{0.82}Ca_{0.18}MnO_3$ wherein the current-induced low resistive state exhibits memory effects.[10] We were interested to explore electric-field effects on rare-earth manganate compositions which exhibit CMR in order to find out whether we can observe colossal electroresistance and any other effects induced by electric fields in these manganates. For this purpose, we have studied current-induced effects on $La_{0.77}Ca_{0.23}MnO_3$ and $La_{0.9}MnO_3$ which exhibit IM transitions near the ferromagnetic Curie temperatures.

Single crystals of $La_{0.77}Ca_{0.23}MnO_3$ and $La_{0.9}MnO_3$ were grown by the floating-zone furnace fitted with two ellipsoid halogen lamps, having prepared the polycrystalline samples of the materials by the solid-state route. Monophasic polycrystalline samples were hydrostatically pressed and sintered at 1400 °C for 24 h to obtain feed and seed rods of dimensions 8 cm in length and 4 mm in diameter. The crystals thus obtained were cut and subjected to oxygen annealing for 48 h. Magnetization data were obtained with a vibrating sample magnetometer operating between 300–50 K. The $La_{0.77}Ca_{0.23}MnO_3$ and $La_{0.9}MnO_3$ showed ferromagnetic Curie temperatures, T_C of 230 and 259 K, respectively. Electrical resistivity measurements were carried out on crystals of 4–6 mm diameter and 1 mm thickness by the standard four-probe method.

In Fig. 1, we show the effect of varying electric currents on the IM transition in $La_{0.77}Ca_{0.23}MnO_3$, in the cooling and warming cycles. We readily see that the IM transition temperature, T_{IM}, which is close to the ferromagnetic T_C, decreases with the increase in current, although there is little change in the peak resistance. In Fig. 2, we have plotted the T_{IM} values in the cooling and warming cycles against the applied current. The T_{IM} value decreases markedly with increasing current, in both the cooling and warming cycles. While there is little hysteresis at a small value of the current (0.1 mA), the magnitude of hysteresis, ΔT, increases with increasing current linearly as shown in the inset of Fig. 2. These data demonstrate that the applied current increasingly induces first-order characteristics on the IM transition and that the observed effects are thermodynamic.

If we designate the IM transition temperature at the lowest applied current (0.1 mA) as T^o_{IM}, we observe a current-induced decrease in resistance at $T > T^o_{IM}$ and a marked increase in resistance at $T < T^o_{IM}$. We can define an electroresistive ratio (ER) as

$$\%\mathrm{ER} = \frac{R(I) - R(I\,\mathrm{min})}{R(I\,\mathrm{min})} \times 100,$$

where $R(I\,\mathrm{min})$ is the resistance at the lowest applied current at a given temperature and $R(I)$ is the resistance at a higher value of the current. In Fig. 3(a), we plot %ER for $T > T^o_{IM}$ for different values of the applied current. We see that the %ER reaches −40% around 240 K and remains around −30% even around room temperature. The positive ER in Fig. 3(b) for $T < T^o_{IM}$ reaches very high values in the 200–150 K temperature range. Such colossal electroresistance could find applications.

We have examined current-induced effects in self-doped $La_{0.9}MnO_3$ as well. In Fig. 4, we show the effect of applied current on the IM transition at different current values in the cooling cycle. While the observed effects are somewhat

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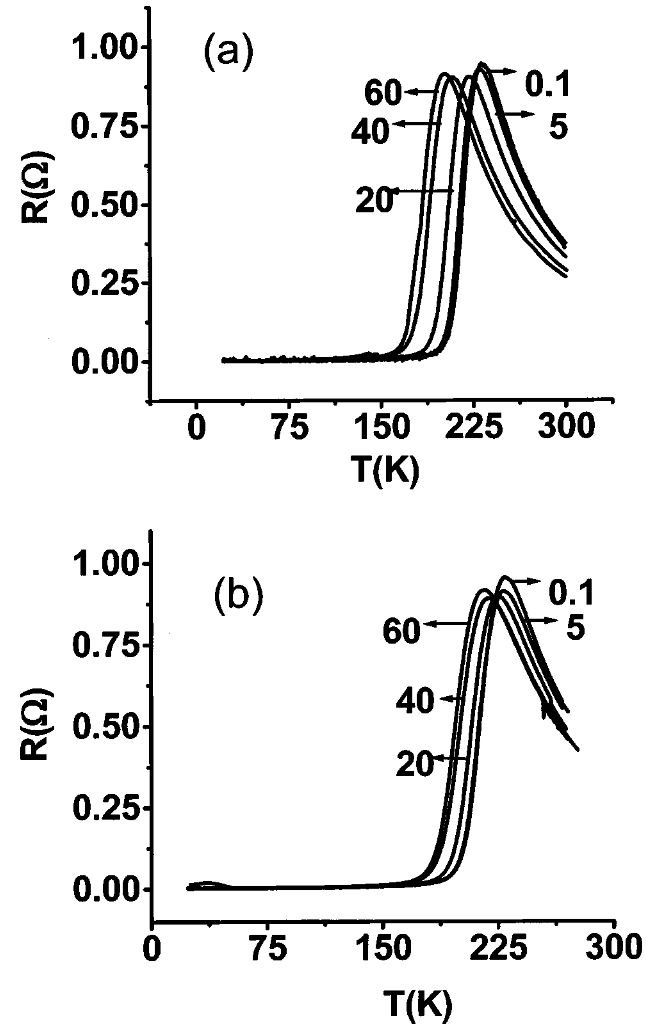

FIG. 1. Temperature variation of resistance of $La_{0.77}Ca_{0.23}MnO_3$ at different currents recorded in (a) the cooling cycle and (b) the warming cycle. Note the shift in the IM transition temperature with current. The numbers indicate the current values in mA.

smaller than in $La_{0.77}Ca_{0.23}MnO_3$, there is little doubt that T_{IM} decreases with increasing applied current. Here again, the thermal hysteresis increases with increasing current as can be seen from the inset in Fig. 4.

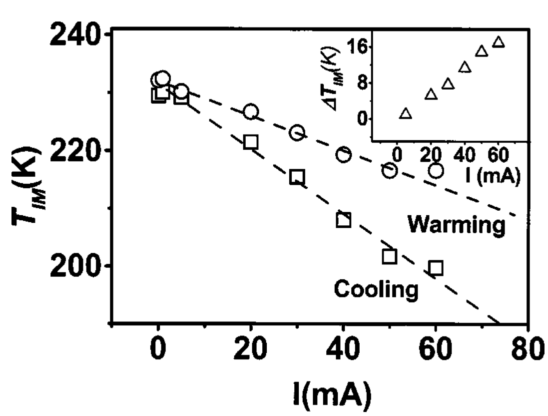

FIG. 2. Variation of T_{IM} of $La_{0.77}Ca_{0.23}MnO_3$ with the applied current. Open squares represent the data for cooling cycle and open circles represents the same in the warming cycle. Inset shows the difference in the T_{IM} in cooling and warming cycles.

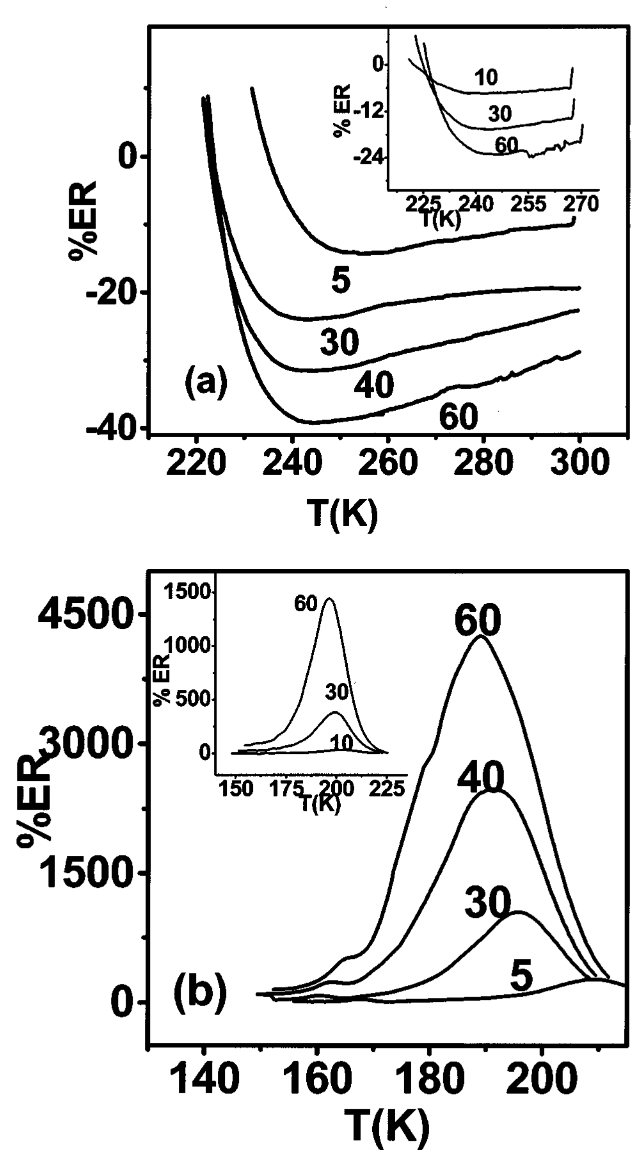

FIG. 3. Temperature variation of negative %ER of $La_{0.77}Ca_{0.23}MnO_3$ (a) in the cooling cycle and inset shows the data for warming cycle. (b) Temperature variation of %ER of $La_{0.77}Ca_{0.23}MnO_3$ in the cooling cycle and inset shows the data for warming cycle. The numbers indicate the value of current in mA.

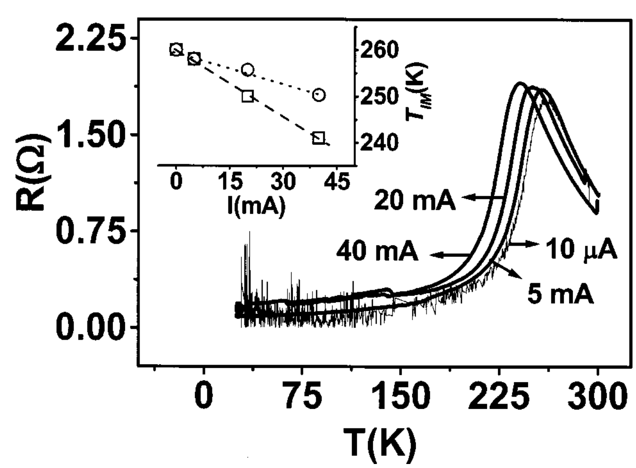

FIG. 4. Temperature variation of resistance of $La_{0.9}MnO_3$ at different current values, recorded in the cooling cycle. Inset shows the variation of T_{IM} in the cooling (squares) and warming (circles) with the current through the sample.

It must be noted that the effects discussed herein are not due to ohmic heating. Such heating cannot explain our observations and one would expect, if any, an opposite effect with a decrease in the peak resistance for high currents. The marked decrease in T_{IM} with applied current is understandable since the magnetic order and its fluctuations are coupled strongly to electron transport in the manganates. The coupling is strong around T_{IM} (T_C), manifesting as a resistance peak due to critical spin disorder scattering of the conduction electrons. In the presence of finite transport current under nonequilibrium conditions, one expects a softening of the spinwave excitation spectrum which would, in turn, enhance the spin fluctuations and lower the T_C. Such a charge-transport driven phase transition can indeed become first order.

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