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Current-induced effects in $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ ($y = 0.35$) single crystals

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Abstract

DC electrical current-dependent resistance and pulsed current–voltage characteristics as a function of temperature of mixed valent Mn oxide-based $La_{5/8-y}Pr_yCa_{0.375}MnO_3$ ($y = 0.35$) single crystals are reported. We find that the low-temperature regime of this material is strongly current dependent. For small current densities ($\sim 10\,\mu A/cm^2$), the metal–insulator transition related to the low-temperature enlargement of the ferromagnetic fraction is not observed down to 10 K. Higher current densities causes a large decrease of the resistance, which is temperature dependent and exhibits memory effects. Our results are interpreted within a scenario of strong competition between charge and ferromagnetic ordering.

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Keywords: Non-linear effects; Manganites; Percolation; Phase separation

1. Introduction

The magnetic phase separation scenario that presents some manganites can be very useful to produce samples with a controlled mixture of charge-delocalized ferromagnetic (CD-F) and charge-ordered, antiferromagnetic (CO-AF) phases. It will be technically interesting to externally control the proportions of these highly conducting and insulating mixtures as a change in the resistivity of several orders of magnitude can be obtained as a consequence of the percolating nature of the problem [1]. This kind of framework was already observed for hole-doped manganites

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[2–4], particularly for the $(La, Pr, Ca)MnO_3$ compound [5,6], where the conducting phase can be tuned by temperature, magnetic or electric field, time or grain size. Indeed, it was previously shown that this prototypical manganite is magnetically phase separated into the mixture already mentioned, as a consequence of the intricate interplay between metallic F and the AF interactions [1]. In this paper we present measurements of the electrical field dependence (DC and pulsed) of the resistance as a function of temperature in $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ ($y = 0.35$) single crystals. Our results can be interpreted within a percolation scenario where the proportion of the CD-F phase over the CO-AF is favored by the application of an electric field.

2. Experimental

Single crystals of nominal composition $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ (LPCMO, $y = 0.35$) were synthesized and characterized as was described previously [7]. The resistivity was measured as a function of temperature ($4\,K \leqslant T \leqslant 300\,K$) and electric field, following different configurations: a four terminal (4 W) standard configuration for constant current measurements and a two wire (2 W) configuration for high resistivities (up to 100 GΩ—using a Keithley 2400 SourceMeter) or for a constant voltage measurement. Pulsed measurements were performed, depending on the magnitude of their period, by generating a single square pulse of increasing voltages (up to 10 V) for 20 ms to 2 s (Agilent 33250A 80 MHz Function/Arbitrary Waveform Generator) and determining the current by measuring the voltage in a calibrated resistance using an oscilloscope or directly with the SourceMeter for longer pulses. Temperature was measured by a small diode thermometer thermally anchored directly to the sample.

3. Results and discussion

The resistivity of a LPCMO ($y = 0.35$) single crystal as a function of temperature for different constant voltages is displayed in Fig. 1. For small voltages ($V \leqslant 10\,V$), the sample remains insulating down to low temperatures. When higher voltages are applied ($10\,V \leqslant V \leqslant 100\,V$), a metal–insulator-like transition can be observed, reaching more than four orders of magnitude of resistivity drop for the highest voltages. Characteristic temperature hysteresis of this system can also be observed. The voltage dependence of the resistivity ($\rho(V)$), at a fixed temperature for a sample cooled in zero applied voltage, is shown in Fig. 2. When voltage is increased, we measured a very high resistance (HR) that remains nearly constant until a temperature-dependent critical voltage (V_c) is reached, where a drop of more than four orders of magnitude is observed. In these conditions, a time evolution of the resistivity can also be observed, a reduction of the resistivity can be noticed even for a constant applied voltage. When the voltage is decreased, the sample shows memory effects, remaining in a different and more conducting low resistance—regime (LR). If the voltage is increased again, a nonlinear dependence is observed, even for voltages lower than V_c.

Considering that the decrease of the resistance produces an increase of the Joule dissipation in the sample, the highest voltage parts of these $\rho(V)$ curves could be modified by overheating. To rule out this possibility, we performed pulsed $R(V)$

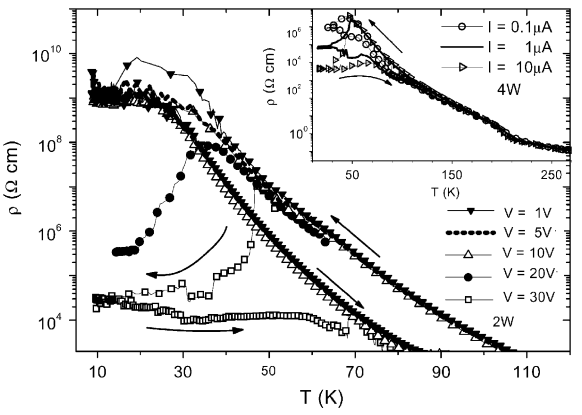

Fig. 1. Resistivity (2 W, voltage controlled) of LPCMO as a function of temperature for various applied voltages. The temperature evolution is indicated by arrows. The inset shows the 4 W, current controlled resistivity.

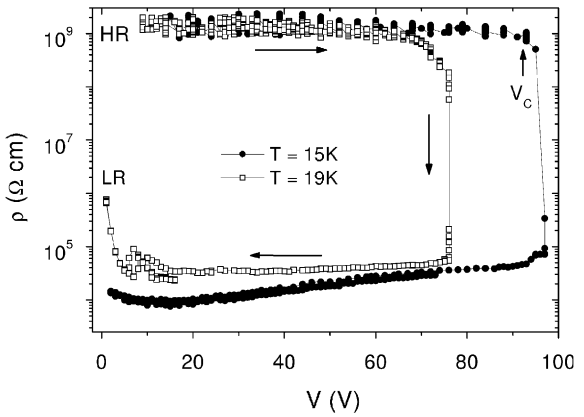

Fig. 2. Resistivity of LPCMO as a function of the applied voltage at $T = 15\,\mathrm{K}$ and $T = 19\,\mathrm{K}$. The system evolves from a HR to a LR regime, after applying a voltage $V \geqslant V_c$.

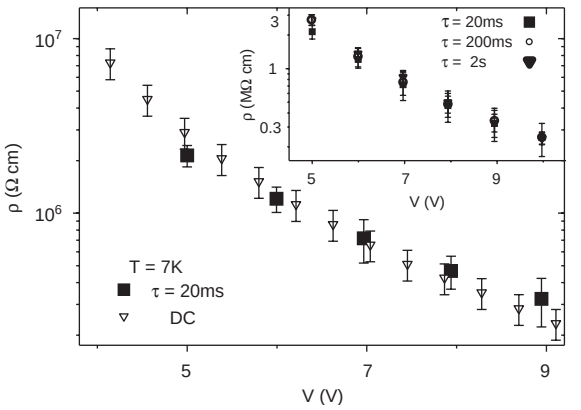

Fig. 3. Resistivity of LPCMO as a function of voltage measured applying single square pulses of $\tau = 20\,\mathrm{ms}$ compared to DC measurements. The inset shows the comparison of the $R(V)$ curves obtained with $\tau = 20\,\mathrm{ms}$, 200 ms, and 2 s.

measurements in the LR regime. As can be observed in Fig. 3, no time dependence can be observed for pulsed and DC voltages $V < 10\,\mathrm{V}$.

These results can be analyzed considering the general effective medium (GEM) equations developed by Mclachlan [8] to describe the electric or thermal conductivity of a binary mixture of conducting and insulator materials

$$f\frac{(\sigma_M^{1/t}-\sigma_E^{1/t})}{(\sigma_M^{1/t}+A\sigma_E^{1/t})}+(1-f)\frac{(\sigma_I^{1/t}-\sigma_E^{1/t})}{(\sigma_I^{1/t}+A\sigma_E^{1/t})}=0, \qquad (1)$$

where f is the volume fraction of the CDF domains, σ_M and σ_I the conductivities of the metallic and insulating phases, respectively. σ_E is the effective conductivity that we measure, t a critical exponent, and $A = (1 - f_c/f_c)$, where f_c is the percolation threshold.

Assuming a 3D percolation scenario ($t = 2, f_c = 0.17$) and that $\sigma_M(T) = \sigma(T)$ of $x = 0$ and $\sigma_I(T) = \sigma(T)$ of $x = 0.625$, the $f(V)$ and $f(t)$ values can be obtained, fitting our data with the GEM equation. The obtained $f(t)$ and $f(V)$ dependencies are shown in Figs. 4 and 5, respectively. In some cases, not shown here, we obtained that $f(t)$, $f(V) > 0.17$, with a corresponding metallic-like temperature-dependent resistivity for temperatures in the $10\,\mathrm{K} < T < 20\,\mathrm{K}$ range.

The nonlinearities observed in the LR regime ($I \sim aV + bV^{7/3} + cV^{7/2}$, as shown in Fig. 6) indicate the presence of a different conducting mechanism, which can be associated with tunneling processes [9] described, particularly, by the Glazman–Matveev (GM) theory [10]. Thus, these results point out a rich scenario where the voltage dependence of the resistivity of LPCMO single crystals can be related to different processes: a classical percolating framework in a conducting–insulating phase mixture and, when the conditions are favorable, tunneling processes between conducting paths separated by small insulating barriers.

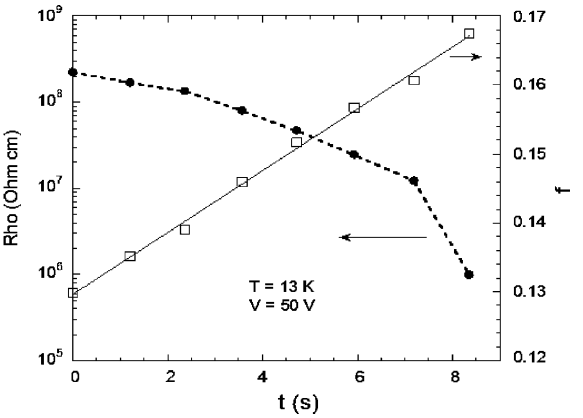

Fig. 4. Time evolution of the resistivity and of the calculated CD-F volume fraction $f(t)$ (Eq. (1)) after applying a voltage $V \geqslant V_c$ at $T = 13\,\mathrm{K}$.

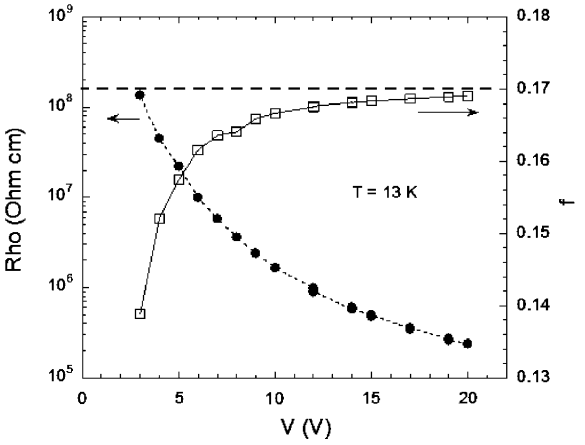

Fig. 5. Voltage sensitivity of the resistivity and of the CD-F volume fraction ($f(V)$) in the LR regime at $T = 13$ K. The dashed line indicates the critical value f_c.

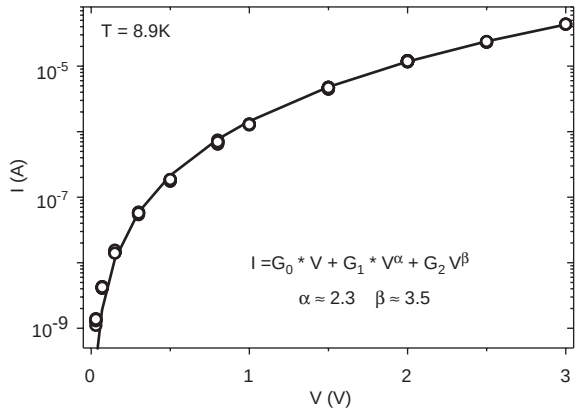

Fig. 6. Nonlinearities measured at $T \simeq 8.9$ K in the LR regime, which can be fitted (solid line) by the multi-step tunneling GM theory.

4. Conclusions

We have shown colossal electro-resistance effects on LPCMO single crystals. Thus, we have shown that the conductivity of the samples can be varied with several orders of magnitude by sweeping an external voltage over a temperature-dependent critical value. Memory effects are then observed as a zero voltage low-resistivity metastable state is obtained. Our results can be analyzed within the GEM formalism, indicating that the electronic properties of the LPCMO samples are dominated by a percolative 3D conduction through the CD-F phase embedded in a CO-AF matrix, while tunneling processes also seem to be present in the LR regime.

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