Current localization and Joule self-heating effects in Cr doped Nd$_{0.5}$Ca$_{0.5}$MnO$_3$ manganites

A. S. Carneiro and R. F. Jardim

Instituto de Física, Universidade de São Paulo, CP 66318, 05315-970, São Paulo, SP, Brazil

F. C. Fonseca

Instituto de Pesquisas Energéticas e Nucleares, CP 11049, 05422-970, São Paulo, SP, Brazil

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The effects of dc excitation current on the current-voltage curves of polycrystalline samples of Nd$_{0.5}$Ca$_{0.5}$Mn$_{0.96}$Cr$_{0.04}$O$_3$ were investigated. The experimental results show that an abrupt jump in the voltage is concomitant with a huge increase in the temperature of the sample. A simple model and estimates for Joule self-heating effects support the experimental data. Moreover, the data strongly suggest that both the current localization in the metallic paths and local Joule self-heating effects are essential ingredients to understand the current-induced phase transition in phase-separated manganites.

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It has been reported that phase control in manganites can be achieved by both the appropriate chemical substitutions and by application of external stimuli such as pressure $P$, magnetic field $H$, high-power laser irradiation and electric fields $E$. However, while both $H$ and $P$ change the bulk properties of the colossal magnetoresistance (CMR) manganites, the influence of light, X-rays, and electric current $I$ may result in a rather localized phase transition. In particular, effects induced by applied $E$ have been studied in both the charge/orbital-ordered (CO-OO) and the ferromagnetic-metallic (FMM) states in single crystals and polycrystalline samples of CMR manganites. The proposed scenarios for the $I$ (or $E$)-induced phase transition usually consider mechanisms related to the interplay between charge and spin degrees of freedom. However, based on magneto-optical imaging of local magnetization and $I$ distribution, a different view has been recently proposed. It was suggested that application of high density $I$ reduces the metallic channels due to local Joule self-heating effects. Increasing $I$ changes the metallic paths into insulating regions, further localizing the inhomogeneous current flow, and propagating the Joule self-heating. This process results in a rapid collapse of the metallic channels, which is related to an abrupt jump in the $I$-$V$ curves at a well-defined $I$ threshold $I_{T}$.

Besides the common observation of nonlinearity in $I$-$V$ curves, this scenario is still not clear and other physical mechanisms have been proposed as the depinning of the OO states upon $E$ application, the change in the orientation of the OO in the insulating state upon application of large $E$ and an $E$-induced switching of the direction of order of the OO states of $e_g$ electrons. Results of $I$-$V$ curves under $H$ also indicated an increase of the magnetization at the same threshold $V$ where $I$ rises abruptly. It was argued that Joule heating effects were insignificant and the formation of a less-resistive mixed state, but with higher magnetization than the initial CO state, was suggested. More recently, the nonlinear conduction was described by considering a model for charge-density-wave (CDW) motion.

The nonlinearity of $I$-$V$ curves under $H$ is related to an abrupt jump in the $V$-$I$ curves at a well-defined $I$ threshold $I_{T}$. The inset displays the copper block sample holder experimental setup. Warming and cooling cycles in $V$-$I$ measurements ($I=0.05$ mA) are indicated by arrows.

We report the occurrence of abrupt jumps/drops on the $V$ across the phase-separated manganite...
Nd_{0.5}Ca_{0.5}MnO_{3}Cr_{0.04}O_{3} (Cr-NCMO). By using an appropriate experimental setup we have observed that the occurrence of nonlinearity in $I$-$V$ curves is a precursor of a drastic rise in the temperature of the sample $T_S$. A model to account for the increase in $T_S$ on increasing $I$ supports the experimental data. Moreover, based on the temperature dependence of the specific heat $C_p$, an estimate of the energy dissipation of the material further confirms the importance of Joule heating effects. The results allowed us to propose that Joule self-heating effects associated with localized conduction are essential ingredients to understand the abrupt $I$-induced phase transition in phase-separated manganites.

Polycrystalline samples of Cr-NCMO were prepared by solid state reaction. Four-wire dc $\rho(T)$ and $I$-$V$ measurements were performed between 30 and 300 K. Four gold contact pads were deposited on parallelepiped-shaped (typically of length $t=0.60$ cm, width $w=0.145$ cm, and thickness $h=0.075$ cm) samples to obtain excellent electric contacts by using Ag epoxy. In $I$-$V$ measurements, the current sweeps between -100 $\leq I \leq$ 100 mA were carried out in 0.5 mA steps. To avoid any memory effects, after each measurement the sample was heated to 300 K, cooled down to 30 K, and then heated to the desired measuring $T$. The experimental setup for the electrical measurements uses a cold finger connected to a closed-cycle helium refrigerator. The temperature of the system $T$ is monitored with a silicon diode, positioned near the sample, and mounted on the copper block. The temperature of the sample $T_S$ is measured by a Pt thermometer placed on the copper block. The sample stands over the Pt thermometer and is fixed to it by using a thin thermal conducting silicon grease layer. The inset in Fig. 1 displays a schematic draw of the sample holder setup. Magnetization $M(T)$ measurements were performed in a SQUID magnetometer under applied magnetic field of 1 mT in both FC and ZFC modes. $C_p$ measurements in Cr-NCMO crystals were performed in a Quantum Design PPMS apparatus.

The compound Nd_{0.5}Ca_{0.5}MnO_{3} has a CO antiferromagnetic-insulator (AFI) ground state below $T_{CO}$ $\sim$ 240 K where a partial OO and magnetic correlations of short range are observed. At low $T$, the OO increases and a long range CE-type AFI state is established below $T_N$ $\sim$ 170 K. Substitutions on the Mn site are an effective way to gradually modify the CO state. The material under investigation Nd_{0.5}Ca_{0.5}MnO_{3}Cr_{0.04}O_{3} is well studied and develops a FMM phase at low temperatures, as shown in Fig. 1. The transition to the CO regime is inferred from a subtle change of $d\rho/dT$ and a small cusp in the $M(T)$ data at $T_{CO}$ $\sim$ 240 K. With decreasing $T$, a MI transition, closely related to the FM transition, occurs at $T_{MI}$ $\sim$ 140 K when $I=0.05$ mA. The thermal irreversibility in both $M(T)$ and $\rho(T)$ data indicate a coexistence of CO/OO and FMM phases at low temperatures. Indeed, under Cr-doping and for $T < T_{MI}$, these compounds are comprised of a fine mixture of 20-30 nm domains of the FMM phase embedded in the CO/OO matrix.

Fig. 2 shows a typical $I$-$V$ curve ($T=50$ K) and the temperature rise of the sample $T_S$ as $I$ increases. The data reveal a clear correspondence of both $V$ and $T_S$ curves against $I$. At low $I$, the rising in $T_S$ is smooth and the $I$-$V$ data is here considered linear up to an $I$ value $I_L$ $\sim$ 14.5 mA. $I_L$ is defined as the current in which the temperature measured by the Pt-thermometer increases 1 K. As $I$ evolves, the $I$-$V$ data loose the linearity and $T_S$ rises progressively. Further rising in $I$ leads to a switch to a much higher $V/I$ value, as inferred from a steep increase in $V$ at $I_{Ti}$ $\sim$ 59 mA ($\sim$ 5.4 A/cm$^2$). This $I$-induced phase transition is accompanied by a remarkable increase in $T_S$ from $\sim$ 70 to $\sim$ 135 K in a narrow range of $I$ while $V$ displays a spike-like maximum. With further increasing in $I$, $V$ decreases monotonically, i.e., the $I$-$V$ curves exhibit a negative differential resistance $(dV/dI<0)$ up to $I=100$ mA. This feature is also reproduced in $T_S$ that rises continually, reaching $\sim$ 155 K for $I=100$ mA, while the temperature of the copper block varied $\sim$ 1.6 K.

The $I$-induced phase transition at $I_{Ti}$ is useful to separate two well defined regions in dynamical $I$-$V$ curves: below and above $I_{Ti}$, where the material can be considered as a metal or insulator, respectively. The decreasing branch of $I$-$V$ curves shows a pronounced irreversibility in $I$, preserving the more insulating state down to a lower value of the threshold current $I_{Td}$ ($T=50$ K) $\sim$ 40 mA. At this excitation current, $V$ drops rapidly, and the initial $I$-$V$ curve is recovered with further decreasing $I$. Again, $T_S$ follows essentially the same trend: it decreases from $T_S$ $\sim$ 150 K but it is kept above 100 K while the more insulating state is preserved. Accordingly, close to $I_{Td}$ ($T=50$ K) $\sim$ 40 mA, $T_S$ drops rapidly to $\sim$ 55 K and continuously decreases towards to the initial value ($50$ K).

Due to a thin conducting grease layer separating the
phase. The sweep to negative evidences the localized electric conduction in the FMM ing larger when the FMM phase is robust, and further phase competition in the phase-separated manganite, be-

From Fig.2, the spike-like maximum in $V$ at constant and rather low value of $I=0.05$ mA, must be also considered. For such low values of $I$ a homogeneous distribution of $I$ within the sample is expect, a feature hardly believed to occur at $I=60$ mA.

A set of $I$-$V$ curves taken at different $T$ is displayed in Fig.3. The most prominent feature here is the sharp jump in $V$ at $I\sim I_{T1}$, which decreases appreciably with increasing $T$ and is barely identified for $T>T_{MI}$ (inset of Fig.3). Increasing $T$ also results in a less pronounced ir-

From $I$-$V$ curves at different $T$, values of $I_L$ were computed and are shown in the inset of Fig.4. Such a dia-

gram is useful for separating regions where current localization takes place in these materials. The decrease of $I_L$ with increasing $T\leq T_{MI}$ reflects the reduction of the relative volume fraction (VF) of the FMM phase, which coexist with the CO/AFI\textsuperscript{19,20}. Such a reduction is consistent with a more localized $I$ distribution across the material and provides an explanation for the so-called $I$-induced change in $T_{MI}$\textsuperscript{19}. A considerable increase of $I_L$ for $T>T_{MI}$ suggests a much more homogenous distri-

We have carefully checked changes in the temperature of the copper block in all $I$-$V$ curves. The data show a maximum temperature rise of the block of $\sim1.9$ K at $T=30$ K, when the sample is subjected to $I=100$ mA and $T_S\sim165$ K. Thus, the temperature rise of the sample with respect to the copper block has been defined as $\Delta T = T_S-T$ and typical data of $\Delta T$ vs. $I$ are shown in Fig.4. At $T=30$ K, a huge rise of $\sim100$ K in $\Delta T$, at essentially the same $I=80$ mA where $V$ increases drastically, is observed. The average temperature reached by the sample $T_S$ just after the jump was measured to be $\sim155$ K, a value higher than $T_{MI}\sim140$ K. Such a rise in $T_S$ is certainly caused by Joule self-heating effects, a process that continues up to $I=100$ mA, where $T_S\sim165$ K. When the sample was subjected to a power dissipation of $\sim0.1$ W ($I=100$ mA), the highest value of $\Delta T\sim135$ K was observed for $T=30$ K. Again, a temperature gradient $T_G\sim15$ K, at $I=100$ mA, was estimated, further indicating a current localization, as already mentioned. Such a $T_G$ of the experimental
arrangement is more pronounced at high $I$ and $T \leq T_{MI}$, indicating that the local temperature of the sample is significantly altered by the current localization.

The behavior of $\Delta T$ also displays irreversibility in curves taken at low temperatures, a feature that vanishes for $T \geq T_{MI}$. Increasing $T$ results in smaller values of $\Delta T$, being $\Delta T \sim 2$ K for $I = 100$ mA at $T \sim T_{CO}$ (inset of Fig. 4). Therefore, the experimental data suggest that changes in $I$-V curves are close related to Joule self-heating effects, to the VF of the FMM phase, and to the localized distribution of $I$ in the metallic phase.

![FIG. 5: The $I$ vs. $\Delta T$ curve at 30 K (open squares) and the calculated Joule-self heating effect (solid circles and line). The inset displays the $C_p(T)$ data of a single crystal specimen.](image)

To support the experimental findings, the expected increase in $T_S$ due solely to Joule self-heating effects was calculated by considering the heat dissipation in the sample under applied $I$ and the heat conduction by the grease placed between the sample and the Pt thermometer. Since $T$ was found to be nearly constant during the experiment, the temperature rise of the sample $\Delta T(T)$ can be approximated by $P/\kappa_g (A/d)$, where $P$ is the power dissipated in the sample, and $\kappa_g$ is the thermal conductivity of the grease with area $A$ and thickness $d$. By using the experimental $R=V/I$ data and $P=I^2 \rho*(l/wt)$, $\Delta T(T, I) = P^2 \rho (T + \Delta T)/w t \kappa_g * (l/d, A)$, where $w$, $l$, and $t$ were already defined. Considering that $\kappa_g \sim 0.16$ Wm$^{-1}$K$^{-1}$ is nearly temperature independent in the $T$ range investigated (30-160 K), an example of the calculated $\Delta T(T=30$ K$)$ for a typical sample is shown in Fig. 5. The good agreement between the calculated and the measured $\Delta T(T)$ confirms that Joule self-heating provides an excellent description for the temperature rise of the sample. Small deviations in the high $I$ region are mainly related to: (i) the transition to the insulating regime where the VF of the FMM is low; (ii) changes in $T$ that are assumed to be constant; (iii) a thermal gradient within the sample; and (iv) a more homogeneous distribution of $I$ across the sample (see inset of Fig. 4). Furthermore, the $C_p(T)$ data of a Cr-NCMO single crystal specimen (inset Fig. 6) allowed us to estimate the energy required to heat the sample at a given temperature and to compare with the one due to Joule self-heating effects. By using the $C_p$ data, the energy required to heat a Cr-NCMO sample from 30 to 140 K was estimated to be $\approx 85$ mJ. Accordingly, the energy dissipated by $I_R(T=30$ K$)$ was found to be $\approx 80$ mJ, corresponding to a $T_S \sim 140$ K, in excellent agreement with the data shown in Fig. 6. This result supports the experimental data, and lends credence to the relationship between Joule self-heating effects and the temperature rise of the sample.

In summary, the combined experimental results indicate that the abrupt increase of $T_S$ can be doubtless ascribed to the heat generated by the $I$ flow through localized pathways in the FMM phase. Under relatively low $I$, the weaker metallic regions become insulating, increasing the current density in the remaining FMM paths. This is reflected in both the initial rise of $T_S$ and changes in the $I$-$V$ behavior. As $I$ evolves, the weaker metallic paths are progressively reduced and the Joule self-heating effect decreases, culminating in a rapid collapse of the FMM phase. This is mirrored in the observed jumps in $V$ and the dramatic rise of $T_S$. The Joule self-heating is strong enough to promote the sample to the insulating phase ($T_S > T_{MI}$), resulting in a less appreciable self-heating due to a much more homogeneous distribution of $I$.

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The irreversible behavior of both $V$ and $T_S$ vs. $I$ curves below $T_{MI}$ are further evidence of the phase-separation in this manganite. In this $T$ range both $I_{Ti}$ and $I_{Td}$, along with the difference $I_{Ti}-I_{Td}$, decrease with increasing $T$ and approach zero at $T \sim T_{MI}$.

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