Revealing polarons with high pressure on low electron-doped manganites

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Pressure sensitivity (up to 1 GPa) of the electrical resistivity and of the ac susceptibility was measured for low electron-doping levels of $Ca_{1-x}Y_xMnO_3$ (CYMO) and of $Ca_{1-x}Sm_xMnO_3$ (CSMO) ceramic samples ($0.05 \leqslant x \leqslant 0.15$). A very weak pressure dependence of the Curie temperature (T_c) was observed for both systems (~ 6 K GPa^{-1}), when compared to the hole-doped manganites of the same T_c (~ 20 K GPa^{-1}). Our results can be interpreted within a modified double exchange scenario, where pressure alters the reduction of the bandwidth produced by the electron-phonon interaction associated with small Fröhlich polarons in the weak to intermediate coupling regime.

I. INTRODUCTION

The study of high-pressure effects on the magnetic transitions and the electrical transport properties of manganite compounds can provide useful information about the relevant mechanism in magnetic ordering and its relation to transport properties. Indeed, this was the case for the pressure dependence of the Curie temperature $[T_c(P)]$, mostly studied, up to now, in the hole-doped part of the phase diagram of these compounds.

It was experimentally shown[1,2] that the pressure coeficient $[d \ln(T_c)/dP]$ as a function of T_c follows a sort of universal curve, independent of the mean A ionic radius R_A (1.124 Å $< R_A < 1.147$ Å) for the $A_{0.67}B_{0.33}MnO_3$ compounds ($A=Pr, Sm, Nd, Y, La; B=Ca, Sr$), or the doping level x ($0.2 \leqslant x \leqslant 0.4$) for $La_{1-x}Sr_xMnO_3$ and $Nd_{1-x}Sr_xMnO_3$.

It has also been shown[1] that a qualitative understanding of this curve can be obtained in terms of the double exchange (DE) model.[3,4] Within this model, pressure increases the transfer integral of the e_g electron, which is hopping from Mn^{3+} to Mn^{4+}. This results in a broadening of the bandwidth (W_0), which yields an increase of T_c. This can be understood by considering the pressure dependence of two geometric or steric factors, both of which control W_0: the Mn-O distance (d_{Mn-O}) and the bending angle (Θ) of the Mn-O-Mn bond. However, calculations based on a simple model related to pressure variations of the steric factors do not give a good quantitative agreement with the experimental data. A better quantitative prediction can be obtained by considering the polaronic modifications of W_0 due to the Jahn-Teller (JT) cooperative effect. The existence of these polaronic contributions was clearly established for some of the manganite compounds.[5–8] It has been shown that a positive contribution to the pressure dependence of T_c arises from the negative pressure derivative of the isotope exponent, α

$= 1/2 \gamma E_{JT}/h\omega$, where γ is a positive constant ($\leqslant 1$), E_{JT} is the JT energy, and ω is an appropriate optical frequency. Even though the experimental results could not be suitably fitted by this model, as for example, in the case of the studies of the oxygen-isotope effects under pressure in the $La_{0.65}Ca_{0.35}MnO_3$ samples,[9] the $d \ln T_c/dP$ value observed for the ^{18}O sample (23%) is higher than the one predicted by this model (6%). It was shown[10] that this controversy can be solved considering the polaron theory in the intermediate electron-phonon coupling region ($\lambda \sim 1$) and in the adiabatic approximation,[11] which also gives an adequate description of the electrical conductivity for temperatures above T_c.

The study of electron-doped manganites can shed light on the issue whether polarons are relevant or not to the $T_c(P)$ dependence in manganites. As the low e_g level occupancy of the Mn^{4+} ions no longer favors the JT distortion, JT polaronic effects are not expected to contribute to transport properties or to magnetic ordering in the same way they are expected to participate in the hole-doped part of the phase diagram. Then, a small $T_c(P)$ dependence for the electron-doped manganites would reveal the important role played by JT polarons in the hole-doped samples and may also indicate, on the other hand, an active role of other types of charge carriers, like lattice or magnetic polarons, associated with electron-doped manganites in other studies.[12,13]

For low doping levels ($0.05 \leqslant x \leqslant 0.15$), $Ca_{1-x}Y_xMnO_3$ (CYMO) and $Ca_{1-x}Sm_xMnO_3$ (CSMO) compounds are typical electron-doped manganites. For this doping regime a competition between antiferromagnetic (AF) superexchange and ferromagnetic (FM) DE interactions has been revealed by dc-magnetization and electrical transport measurements as a function of temperature and magnetic field for the CYMO system.[14–16] Neutron diffraction studies[17,18] for CSMO ($x=0.10$ and $x=0.15$) showed that, besides the FM phase, a Pnma structure associated with a G-type AF is also

present in the paramagnetic state. At 10 K, the sample with $x=0.10$ exhibits a G-type AF phase, but for $x=0.15$ the C-type magnetic order (characterized by a P $2_1/m$ space group) dominates, coexisting with smaller G AF clusters.

In this paper we study the high-pressure effects on the resistivity and ac susceptibility as a function of temperature of the low electron-doped CYMO and CSMO manganese perovskites. Although an expected weak pressure dependence of $T_c(P)$ was observed, a quantitative agreement could not be obtained by solely considering the pressure sensitivity of the steric factors within a DE scenario. Our results indicate that the characteristic hopping conduction and the value of the pressure coefficient d ln T_c/dP can be ascribed to the polaronic nature of carriers, which can be associated with small Fröhlich polarons in a weak to intermediate coupling regime.

II. EXPERIMENTAL

Single-phase and well-oxygenated CYMO ($x=0.06;0.07;0.08;0.10$) and CSMO ($x=0.10;0.15$) ceramic samples were prepared by solid-state reaction. These samples had been previously studied and the details of their synthesis and characterization have been published elsewhere.[14,15,17–23]

Resistivity $[\rho(T)]$ and ac susceptibility $[\chi_{ac}(T)]$ were measured as a function of temperature (4 K$\leq T\leq$ 300 K) applying high hydrostatic pressures up to 1 GPa. A self-clamping cell was used with a 50-50 mixture of kerosene and transformer oil as the pressure transmitting medium. $\rho(T)$ was measured following a standard four-terminal DC technique, while $\chi_{ac}(T)$ was evaluated using an applied ac magnetic field of 1 Oe at an excitation frequency of $\simeq$1 kHz.

III. RESULTS AND DISCUSSION

Resistivity as a function of temperature for different pressures is displayed in Fig. 1 for CYMO ($x=0.08;0.10$) and CSMO ($x=0.10$). A metallic conductivity at room temperature followed by a semiconductinglike behavior when decreasing temperature can be observed for both compounds. The low-temperature resistivity shows in all cases a divergence characteristic of an insulator behavior.

The FM transition temperature (T_c) can be associated with an inflexion or a small drop in the resistivity, depending on doping level, as well as with a peak in the ac susceptibility, as can be noticed in the inset of Fig. 1. The low temperature part of $\chi_{ac}(T)$, which can be associated with the behavior of a cluster glass,[20] will not be considered here, as we want to focus our study on the appearance of the FM ordering and its sensitivity to the applied pressure.

Pressure decreases resistivity and enhances the resistivity drop at T_c. T_c was determined using the peak of the logarithmic temperature derivative of $\rho(T)$ and $\chi_{ac}(T)$ curves, as illustrated in the inset of Fig. 2. The obtained pressure sensitivity of T_c, depicted in Figs. 2 and 3, depends on the technique used to characterize the sample. A nonmonotonic dependence is observed for the resistivity data: T_c remains nearly constant for low pressures and then softly increases for higher pressures with a slope $\sim$5–7 K GPa^{-1}. On the
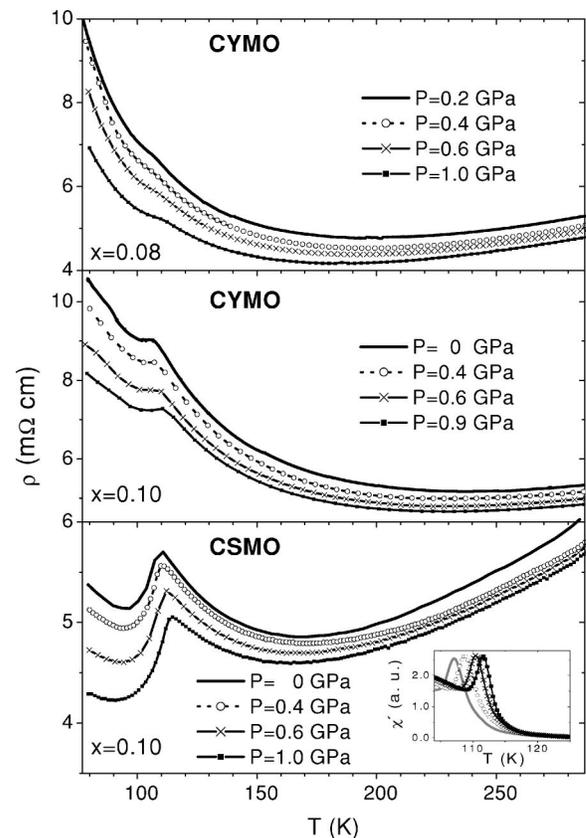
FIG. 1. Pressure dependence of the resistivity as a function of temperature of CYMO for $x=0.08;0.10$ and of CSMO for $x=0.10$. The inset shows the peak observed in the real part of the ac susceptibility $\chi(T)$ for the FM transition of this latter sample. Similar transport and magnetic results are obtained for $x=0.06$ (CYMO) and $x=0.15$ (CSMO), but are not shown here for clarity.

other hand, susceptibility data shows essentially a linear increase of T_c with pressure with a similar slope.

These differences can be associated with the fact that $\rho(T)$ is related not only to the magnetic ordering of the sample, but also to a percolation problem. The percolation scenario can be generated by the coexistence of different conducting
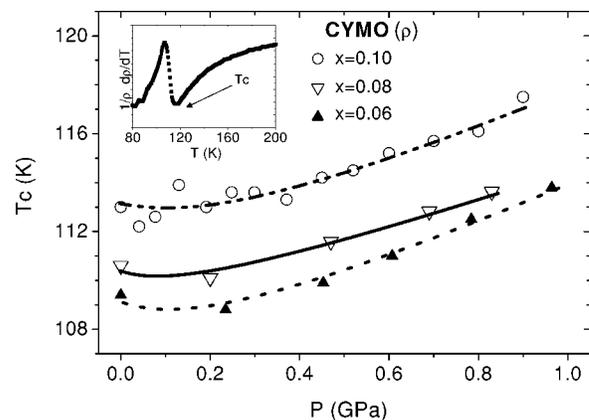
FIG. 2. Pressure dependence of the FM transition temperature T_c of CYMO. In the inset, the criteria to define T_c from the $\rho(T)$ measurements are presented.
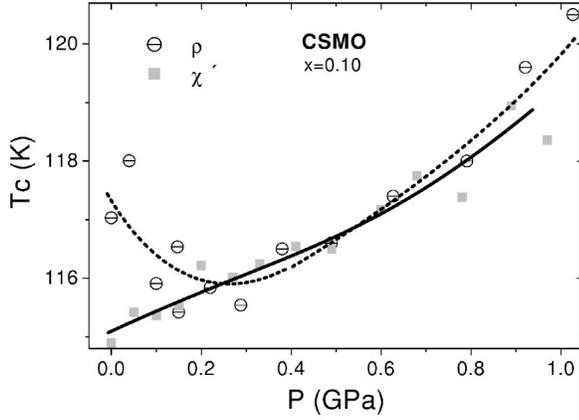
FIG. 3. Comparison of the pressure dependence of the FM transition temperature T_c of CSMO obtained using the temperature logarithmic derivative of $\rho(T)$ and $\chi(T)$.

phases and by their pressure-dependent relative distribution. This was already demonstrated by neutron diffraction studies of these samples, as the coexistence of FM clusters in an AFM matrix. Although the pressure dependence of T_c obtained from $\chi_{ac}(T)$ should then be more reliable, both methods produce similar results if only the high-pressure portion of the curves is analyzed. Considering this criterion, the pressure coefficient d ln T_c/dP can be determined from the $T_c(P)$ curves obtained using both techniques. As shown in Fig. 4, this coefficient is for all the samples (0.05 ± 0.01) GPa^{-1}, which is far from the usual values[2] (universal curve) obtained for hole-doped manganites in this T_c range (~ 0.20 GPa^{-1}).

In a DE scenario the FM transition temperature (T_c) is proportional to the bare bandwidth (W_0), which depends on the steric parameters d_{Mn-O} and $\Theta(P)$.[24] It can be shown that the pressure coefficient can then be estimated as:[1]

$$\frac{d \ln T_c}{dP} = - 3.5\kappa(d_{Mn-O}) - 2\tan(\Theta)\Theta\kappa(\Theta), \qquad (1)$$

where $\kappa(d_{Mn-O})$ and $\kappa(\Theta)$ are the bond length and bond angle compressibilities, respectively. Using Eq. (1) and the
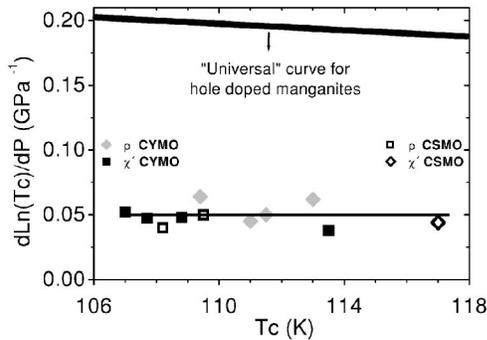
FIG. 4. Dependence of the pressure coefficient d ln T_c/dP as a function of T_c for the CYMO and CSMO electron-doped manganites [determined from $\rho(T)$ or $\chi(T)$ measurements]. The universal curve is a guide to represent data of hole-doped manganites as described in the text.

values already measured for other manganese perovskites,[25] we can evaluate a contribution, at the most, of 0.01 GPa^{-1} to the pressure coefficient produced by the variation of these steric factors. This small value, compared with our experimental results in Fig. 4, reveals the existence of other contributions, which may be based on the polaronic nature of electrical carriers. Though JT collective polarons are not expected in these samples, local lattice distortions or magnetic interactions may play an important role, which may be noticed in the transport properties.

In this sense, the temperature dependence of the resistivity of these samples seems to be a characteristic behavior of the electron-doped manganites, as previously reported in the literature.[14,15,20,26] The observed dependencies (see Fig. 1) can be associated with small polaron hopping at intermediate temperatures, while the room-temperature regime can be assigned to thermally dissociated polaronic states, which yield a metalliclike conductivity, as was recently shown by S. Fratini and S. Ciuchi.[27] A scenario where the room-temperature metallic conduction would be associated with the electronic conduction of a metal, followed by a localization regime at low temperatures, can be ruled out considering the previous thermopower measurements performed on the CSMO samples.[20] Indeed, these measurements showed a linear dependence of the form A+BT (where A and B are negative constants) and values near 60 μV/K ($x=0.10$) at room temperature, which are too large for a metal and reveal the existence of a hopping mechanism for the conduction process,[28] particularly associated with small polarons.[29]

Therefore, we analyze our data considering that the electrical transport at intermediate temperatures is effectively related to the hopping of polarons. As it was shown for the hole-doped manganites,[10] an adequate description should be performed in the adiabatic limit, which must be maintained for the electron-doped samples, as similar characteristic times are expected to be involved. In this case, the resistivity can be expressed as

$$\rho(T) = \rho_0\exp\left(\frac{E_a}{k_BT}\right) \qquad (2)$$

with an activation energy E_a that can be approximated by

$$E_a = \frac{1}{2}E_p - J \simeq \frac{1}{2}E_p - \frac{W_0}{2z}, \qquad (3)$$

where E_p is the binding energy of the polaron; J, the electron transfer integral between nearest neighbor sites; W_0, the bare bandwidth; z, the lattice coordination number; and k_B, the Boltzmann constant.

In the case that carriers could be associated with a polaronic nature, the bare bandwidth W_0 should be narrowed as a consequence of the electron-phonon interaction, producing an effectively smaller W. W and W_0 can be related by the following expression:

$$W = W_0 F^\lambda(E_p,\lambda,\omega_0), \qquad (4)$$

where E_p is the polaron binding energy, $\lambda=E_p/(W_0/2)$ the dimensionless electron-phonon coupling constant and ω_0 a characteristic optical phonon frequency. The F^λ are different
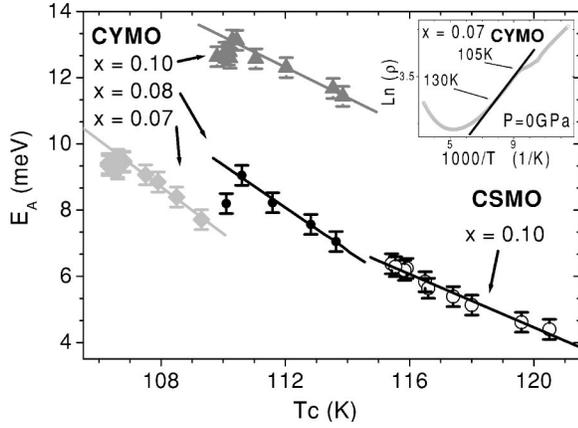
FIG. 5. Activation energy E_a as a function of T_c for CYMO and CSMO with a different doping level. The inset shows an example of the $\ln(\rho)$ versus $1/T$ curves from which the E_a values were extracted.

functions, defined according to the value of λ, which determines the coupling regime.[11,30] For example, in the strong coupling regime $(\lambda \gg 1)$, $F^\lambda \simeq \exp(-E_p/\hbar\omega_0)$, but this function is not valid anymore for the intermediate coupling regime as pointed out by Alexandrov and Mott.[30] From Eqs. (3) and (4) we obtain that

$$E_a = \frac{1}{2}E_p - \frac{CT_c}{F^\lambda}. \tag{5}$$

Following the description of Lorenz *et al.*,[10] we have plotted in Fig. 5 the activation energy E_a as a function of T_c obtained for different pressures for the CYMO and CSMO with different doping levels. Except for the lowest T_c values, where, as already mentioned, their determination is influenced by the percolation problem, an almost linear dependence is obtained. This indicates that E_p and F^λ are essentially constants within our experimental pressure range, independently of the pressure variation of all relevant parameters.

A linear fit of the curves of Fig. 5 yields an estimation of $E_p \simeq 120$–180 meV. The increasing value with increasing doping may point to a tendency to localization, produced by the size mismatch of replacing Ca with small ions like Y or Sm. In this case, considering that $W_0 \simeq 1$ eV, the coupling constant can be estimated as $\lambda \simeq 0.3$, which determines a weak to intermediate coupling regime. This value is over 4 times smaller than the one typically obtained for the hole-doped manganites.[31]

Based on the fact that the electron-doped manganites are very poor conductors, particularly for the low doping range here analyzed, we can assume the framework introduced for doped manganites and other oxides by Alexandrov and Bratkovsky[32] and Alexandrov and Kornilovitch,[33] where small or lattice polarons with a long-range Fröhlich interaction are the quasiparticles involved in the electrical transport properties of these materials. Thus, for these small Fröhlich polarons we can consider the Fröhlich electron-phonon coupling constant

$$\alpha = \frac{e^2}{K_p}\left(\frac{m}{2\hbar^3\omega_0}\right)^{1/2}\left(\frac{1}{4\pi\epsilon_0}\right), \tag{6}$$

where $K_p^{-1} = \epsilon_\infty^{-1} + \epsilon_s^{-1}$ is the effective dielectric constant and ϵ_s and ϵ_∞ are the static and high-frequency dielectric constants, respectively, e is the electron charge, m the electron band mass, and ϵ_0 the vacuum permittivity.

Following Feynman's approach for the weak-to-intermediate coupling regime,[34,35] we can express E_p and the polaronic effective mass, m^* as a function of α:

$$E_p = (\alpha + 0.0123\alpha^2)\hbar\omega_0, \tag{7}$$

$$\frac{m^*}{m} = 1 + \frac{\alpha}{6} + 0.025\alpha^2. \tag{8}$$

Considering a typical phonon frequency (ω_0) of the order of 400–500 cm^{-1}, estimated from Raman spectroscopy studies of CaMnO$_3$ (Ref. 36), and taking a mean value of $E_p = 0.15$ eV from our data, we obtain from Eqs. (6)–(8) that $K_p = 11 \pm 1$, $\alpha = 1.5 \pm 0.1$, and $m^*/m \simeq 1.3$, respectively. The last two values are in accordance with the assumption of being in the intermediate coupling regime[35] $(1 \leqslant \alpha \leqslant 6; m^*/m < 3)$. Also, assuming that the electron band mass is similar to the electron mass $(m \sim m_e)$, the polaron localization radius $R_p = (\hbar/m\omega_0)^{1/2} \leqslant 8$ Å ~ 2 d$_{Mn-O-Mn}$ determines a polaron size in the limit of being considered small. The radius of the lattice distortion produced by the long-range Fröhlich interaction can be estimated[28] as $R_d = 5\hbar K_p/(me^2) \sim 30$ Å. These values are in good accordance with the assumption of an electrical transport mediated by small Fröhlich polarons, i.e., a small size for the electron localization and a large size for the lattice distortion. To our knowledge, there is, at this time, no experimental determination of ϵ_∞ for any electron-doped manganite, though a very high low-frequency dielectric constant ϵ up to 10^8 was measured at room temperature in Ca$_{1-x}$La$_x$MnO$_3$ $(x \leqslant 0.1)$ (Ref. 37). As $K_p \sim 3$ was estimated for LaMnO$_3$ (Ref. 32), a higher value for the electron-doped CaMnO$_3$ can be expected, as a consequence derived from Eq. (6) and its lower coupling constant, when compared with the former manganite.

At this point, we can try to make a rough approximation to determine what the contribution is to the pressure coefficient produced by the dependence of the bandwidth on the mass enhancement, related to the polaronic nature of carriers. To do so, we can consider the approximation of slow polarons $(k \ll q_p$, where k and q_p are the electron and phonon momenta, respectively)[30] where the bandwidth W is essentially modified as

$$W = \frac{W_0}{m^*/m}. \tag{9}$$

Using Eqs. (8) and (9), the pressure coefficient can then be expressed as

$$\frac{d \ln Tc}{dP} \simeq \frac{d \ln W_0}{dP} - \frac{m}{m^*}\frac{d(m^*/m)}{d\alpha}\frac{d\alpha}{d\omega}\frac{d\omega}{dP}. \tag{10}$$

From Eqs. (6) and (7) and assuming that $d\omega/dP = \delta\omega_0$ (with $\delta \sim 0.01$ GPa^{-1}) from Ref. 38, we can evaluate the sec-

ond term of Eq. (10), which gives a positive contribution of ~ 0.02 GPa^{-1}, which is in good agreement with the results presented in Fig. 4. Therefore, these results reveal the necessity of considering an additional contribution to the pressure coefficient besides the one that arises out of the steric factors. The additional contribution can be consistently related to the pressure sensitivity of intermediate-coupling Fröhlich polaronic effects on the bandwidth.

IV. CONCLUSIONS

We have studied the pressure sensitivity of the resistivity and of the FM ordering of electron-doped manganese perovskites (CYMO and CSMO) in the low doping regime. The obtained values of d ln T_c/dP are considerably smaller than the values measured for hole-doped manganites in the same T_c range. Within a DE scenario these values cannot be explained as a consequence of the pressure variation of steric factors. A better quantitative agreement can be obtained if a scenario of small intermediate-coupled Fröhlich polarons is considered.

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