

Pressure Sensitivity of the Ferromagnetic Transition in Perovskite Manganites Revisited

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Abstract. It was already shown that a qualitative understanding of the pressure effects on the ferromagnetic transition temperature (T_c) of the perovskite manganites ($AMnO_3$) results from the variation of the geometric factors that control the bandwidth, the Mn-O-Mn bond angle and the Mn-O bond length and also from the reduction of the effective mass related to the polaronic nature of the carriers. But, up to now, a quantitative agreement was not really obtained, particularly for the hole-doped manganites. Here, we present results on the pressure effects in the T_c of $La_{0.8}(Ca,Sr)_{0.2}MnO_3$ ceramic samples. Our results are described within a phenomenological approach based on the well known dependence of T_c on the mean A ionic radius, that gives a good quantitative agreement of the pressure sensitivity of T_c for several hole-doped manganites.

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In the last years many efforts have been devoted to determine the relevant electronic and structural parameters that determine the Curie temperature T_c of manganites. Historically, the Double Exchange model (DE) was successfully applied in order to correlate electrical transport properties and magnetic ordering in these compounds [1]. But, from the early experiments in the middle of the 90's, the necessity of including polaronic contributions was clear, in order to account for their experimental evidences [2]. Particularly, although a good qualitative understanding for the pressure sensitivity of T_c was achieved, a quantitative agreement was not obtained [3]. Here, we present a phenomenological model based on the A mean ionic radius dependence of T_c for the large bandwidth $AMnO_3$ hole-doped perovskites (where A is a trivalent rare-earth ion that can be partially replaced by a divalent dopant), which usefully describes the quantitative dependence of T_c with pressure (P).

In order to modify the A mean ionic radius ($\langle r_A \rangle$) and to analyze the $T_c(P)$ dependence for these samples at a fixed nominal doping, we have prepared by liquid mix good quality ceramic samples of the hole-doped large bandwidth $La_{0.8}Ca_{0.2-y}Sr_yMnO_3$ compound ($0 \leq y \leq 0.2$). Details of its synthesis will be published elsewhere [4]. We have performed 4 terminal DC

resistivity measurements as a function of temperature (T) and pressure (P) in the $4\text{ K} < T < 300\text{ K}$ and $P < 1.2\text{ GPa}$ range. Pressure was applied hydrostatically using a CuBe piston-cylinder cell with a mixture of kerosene-oil as the pressure transmitting medium. Pressure was measured at room temperature by using a calibrated InSb sensor and remains constant (within a 1% of variation) over all the temperature range, in spite of thermal contractions. Temperature was measured using a well calibrated carbon-glass thermometer.

The $\langle r_A \rangle$ dependence of T_c for these samples is shown in Fig. 1. The ambient pressure $\langle r_A \rangle$ values were calculated using the reported ionic radii in Ref. [5]. We have also included in this curve all the $T_c(\langle r_A(P) \rangle)$ points obtained from the pressure experiment by assuming that $\langle r_A \rangle$ varies linearly with pressure. This assumption was previously verified [6] from experimental results ($d(\langle r_A(P) \rangle)/dP = \gamma$; $\gamma = 0.000375\text{ \AA kbar}^{-1}$). We have also included points from other $D_xT_{1-x}MnO_3$ compounds, where D and T are divalent or trivalent rare-earth atoms, respectively.

A general parabolic behavior can be observed in Fig. 1 for the $T_c(x, \langle r_A \rangle)$ curves, which can be expressed as follows:

$$T_c(x, \langle r_A \rangle) = T_c^M(x) \left\{ 1 - \beta(x) \left(\langle r_A^M \rangle - \langle r_A \rangle \right)^2 \right\}, \quad (1)$$

where x is the nominal doping, $\langle r_A^M \rangle$ ($\sim 1.255 \text{ \AA}$) the A ionic mean radius where T_c reaches its maximum $T_c^M(x)$, and β a parameter which depends on x .

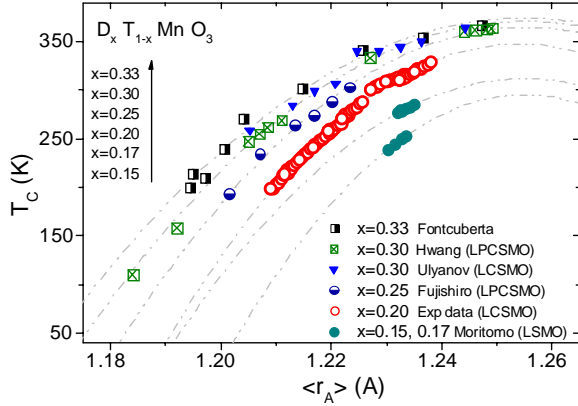


FIGURE 1. $T_c(\langle r_A \rangle)$ dependence for several $D_x T_{1-x} \text{MnO}_3$ compounds. Data from references [6]-[10] are also shown.

Although this $T_c(\langle r_A \rangle)$ dependence was already known, its application to the understanding of the pressure sensitivity of T_c was not taken into account. Instead, most of the efforts were oriented to explain the pressure dependence of T_c considering the compressibility of the steric factors that govern the bandwidth (the Mn-O-Mn bond angle and the Mn-O bond length) within a Double exchange (DE) scenario. But a poor quantitative agreement was obtained, even considering polaronic effects. If we define the pressure coefficient as $d \ln T_c / dP$, then, from Eq. (1) we obtain the following expression:

$$\frac{d \ln [T_c(x, \langle r_A \rangle)]}{dP} = \frac{d \ln [T_c^M(x)]}{dP} + 2\beta(x) \left[\frac{\langle r_A^M \rangle - \langle r_A \rangle}{1 - \beta(x) (\langle r_A^M \rangle - \langle r_A \rangle)^2} \right] \quad (2)$$

where we have also assumed that $\langle r_A^M \rangle$ and $\beta(x)$ are pressure independent by noting, from data of Fig. 1, that pressure essentially modifies $\langle r_A \rangle$, without a noticeable change of the other parameters that define the parabolic dependence of $T_c(\langle r_A \rangle)$. It is also clear that the logarithmic derivative of $T_c^M(x)$ should be small when compared to the second term of the right side of Eq. (2). This term can be calculated by obtaining the $\langle r_A^M \rangle$ and $\beta(x)$ parameters from the curves of Fig. 1. The measured pressure coefficient as a function of T_c can be observed in Fig. 2, where we have also plotted the calculated term as solid lines, added to the estimated steric contributions that control

T_c within the DE model [3]. The obtained quantitative agreement is quite remarkable.

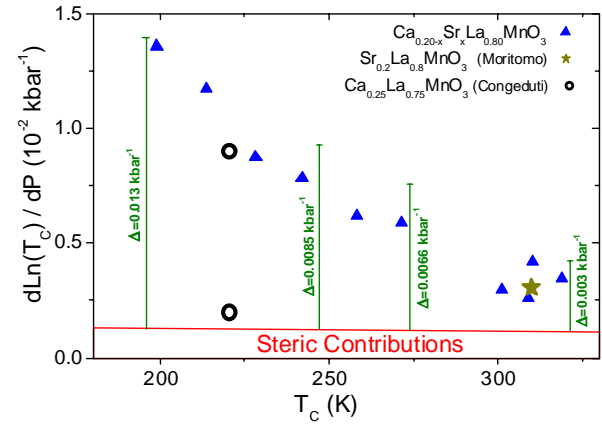


FIGURE 2. Pressure coefficient ($P < 10 \text{ kbar}$) as a function of T_c for our samples. Data from references [10]-[11] are also shown. The solid vertical lines were calculated from Eq. (2).

This result naturally associates the DE steric contributions with the logarithmic derivative of $T_c^M(x)$, while we may relate the term between brackets in Eq. (1) to the effective valence of the Mn, controlled by the value of $\langle r_A \rangle$, but calculations and more experimental evidences are needed to confirm this assumption.

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