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*Title:* The Local Joah-Teller Effect in Lanthanum Strontium (n+1 Manganese(n)Oxide (3n+1)

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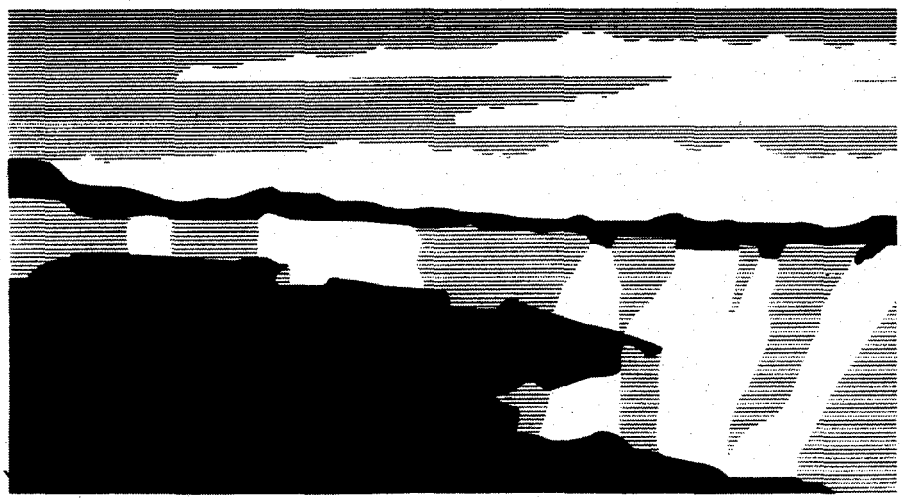

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# The Local Jahn-Teller Effect in $(La/Sr)_{n+1}Mn_nO_{3n+1}$

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## Abstract

Lattice polarons form an important constituent in the mechanism of magnetism and transport of the $(La/Sr)_{n+1}Mn_nO_{3n+1}$ (for n = 2 and ∞) colossal magnetoresistance manganites. The coupling of the polaronic distortions to the local Jahn-Teller (JT) effect has been characterized by the pulsed neutron pair density function (PDF) analysis. Distortions of equal magnitude to the ones found in perovskites arising from a JT effect are found in the 2-dimensional layer crystals. The coupling of the distortions to the lattice is similar in both systems suggesting that lattice polarons are formed independently to the crystal dimensionality.

Key words: JT distortions, lattice polarons, dimensionality

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## 1. Introduction

The phenomenon of colossal magnetoresistance (CMR) occurs in manganese oxides, $(La/A)_{n+1}Mn_nO_{3n+1}$, having the infinite perovskite and layered crystal structures [1]. The combined interaction of the spin, charge and lattice results in the unusually large drop of the resistivity at the Curie transition temperature, $T_C$. In the Zener picture of the interaction mechanism, the effective charge, introduced by the substitution of La, coupled to the local spins through the double exchange (DE) [2]. The lattice component has only been recently suggested [3] as a result of the surmounting evidence pointing towards it. Both structural and transport studies provided evidence for the lattice contribution in the form of polarons [4,5].

Although the layered and perovskite manganites undergo similar magnetic (different $T_C$'s however) and electronic conduction transitions, their crystal structures are quite different. In the perovskites, coulombic interactions induce a strong coupling of the electronic structure to the lattice producing the Jahn-Teller (JT) effect. The subsequent orbital ordering gives rise to the cooperative JT phenomena which induce the observed symmetry distortions from an ideal cubic environment. On the other hand, there has been no clear evidence of a JT effect in the 2 dimensional crystals. In spite of their tetragonal symmetry, the octahedral units in the layers are almost symmetric [6] and crystallographically, the structure does not show a distortion of the magnitude of the JT observed in the perovskites. However, the similarity of their properties at the macroscopic scale is in contradiction to the different crystal structures. In particular, it is questionable whether lattice polarons are present in the layer system as in the perovskite.

In the Mn system of the $3d^4$ configuration, the JT feature is represented as a distortion of the $MnO_6$ octahedron. While the presence of the JT distortions is undisputed in the case of $LaMnO_3$ as it can clearly be charac-

terized in the long range structure, it becomes less defined as the structure changes to rhombohedral as is the case with Sr doping. It is even less defined in the layered manganites as no direct evidence for a JT distortion is found on average. The relation of the JT distortions to the crystal dimensionality is addressed by this pulsed neutron powder diffraction study using the pair density function (PDF) to determine the local atomic structure.

## 2. The Jahn-Teller distortion in $La_{1-x}Sr_xMnO_3$

Pure $LaMnO_3$ is distorted from cubic symmetry lifting the degeneracy of the $e_g$ level. The resulting JT distortion is evidenced as a split in the manganese to oxygen bond distances giving rise to an elongated octahedron. With doping, the $e_g$ level is no longer degenerate indicating that the JT distortion is disappearing. This has been suggested to occur at the insulator-metal (IM) transition which is at $x \sim .15$ in the case of Sr. This coincides with the structural transition to rhombohedral symmetry where all Mn-O bonds are of the same length [7]. Thus, the apparent lack of long range ordering of the JT is consistent with the uniform structure of the metallic state.

However, *locally*, the structure in the rhombohedral, metallic state is quite different. Not only is the JT distortion present at the IM transition concentration, it persists beyond that point, to $x \approx .35$ [5]. The PDF of the local structure as a function of concentration shows evidence for the long Mn-O bonds. The decrease in the density of the JT octahedra with charge doping is shown in Fig. 1. Starting from $LaMnO_3$ with the baseline at 4, the number of short bonds in the octahedron, this number increases as the JT distortion is eliminated per site. The deviation of each concentration point of the 10 K data from 6, which denotes a uniform lattice, is an indication that structural uniformity is achieved at higher concentrations than expected. Thus both

distorted and undistorted regions in the lattice are most likely present [8] consistent with a two fluid model [9].

## 3. The Jahn-Teller distortion in $La_{2-2x}Sr_{1+2x}Mn_2O_7$

The PDF of the local atomic structure of the $La_{1.4}Sr_{1.6}Mn_2O_7$ shown in Fig. 2 is compared to the local structure of $LaMnO_3$ at 300 K. The PDF's are normalized to the respective average scattering lengths, $<b>^2$, to recover the true intensity. Although the two crystals have very different symmetries and it is easily seen in the local structure, it is interesting to note that the local octahedral environment is surprisingly the same. This is represented by the two negative peaks at ~ 1.9 and 2.15 Å (the normalization with the negative Mn b results in the negative peak). In particular, one can notice that the Mn-O bondlengths are split in the 2 layer manganite with a magnitude that is about the same as the split observed in the perovskite [10]. Such a split can only be driven by an effect similar to the JT in perovskites [10], providing the first clear evidence for the JT distortion in the 2 dimensional crystals.

A similar temperature dependence of the local lattice distortions is observed in the 2 layer crystal suggesting that lattice polarons play an important role in this system as well as in the perovskites. The anisotropic nature of the structure introduces an extra variable but the effects remain essentially the same. Thus the similarity in the properties of the two crystal systems is not surprising.

## 4. Implications to properties

The JT in the CMR oxides is strongly coupled to the properties of this system. It is consistent with the presence of two types of electronic states in the perovskites [8, 9] as well as antiferromagnetic domains in the 2 layer

structure [11]. The layer and perovskite manganites have many similarities in common. Namely, the nature of the distortions is the same in both systems. This suggests the following:

a) the JT distortion is equally present in both systems suggesting a critical role of the lattice in the magnetic and transport properties of these materials;

b) and lattice polarons are formed *independently of the dimensionality* of the crystal structure.

In conclusion, while the dimensionality is quite important to the magnetic and transport properties, the local microscopic properties are only affected by the electronic state.

## Acknowledgments

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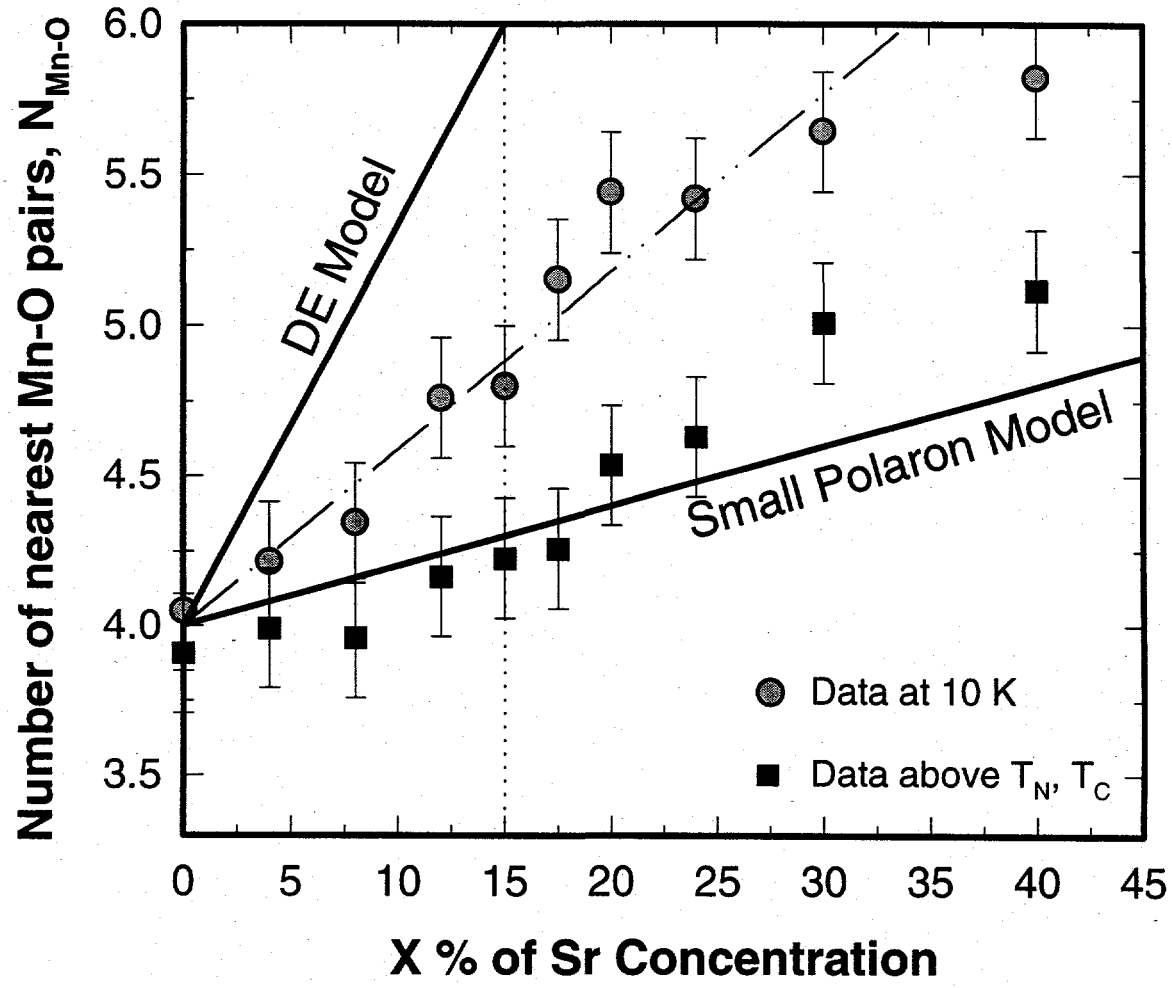
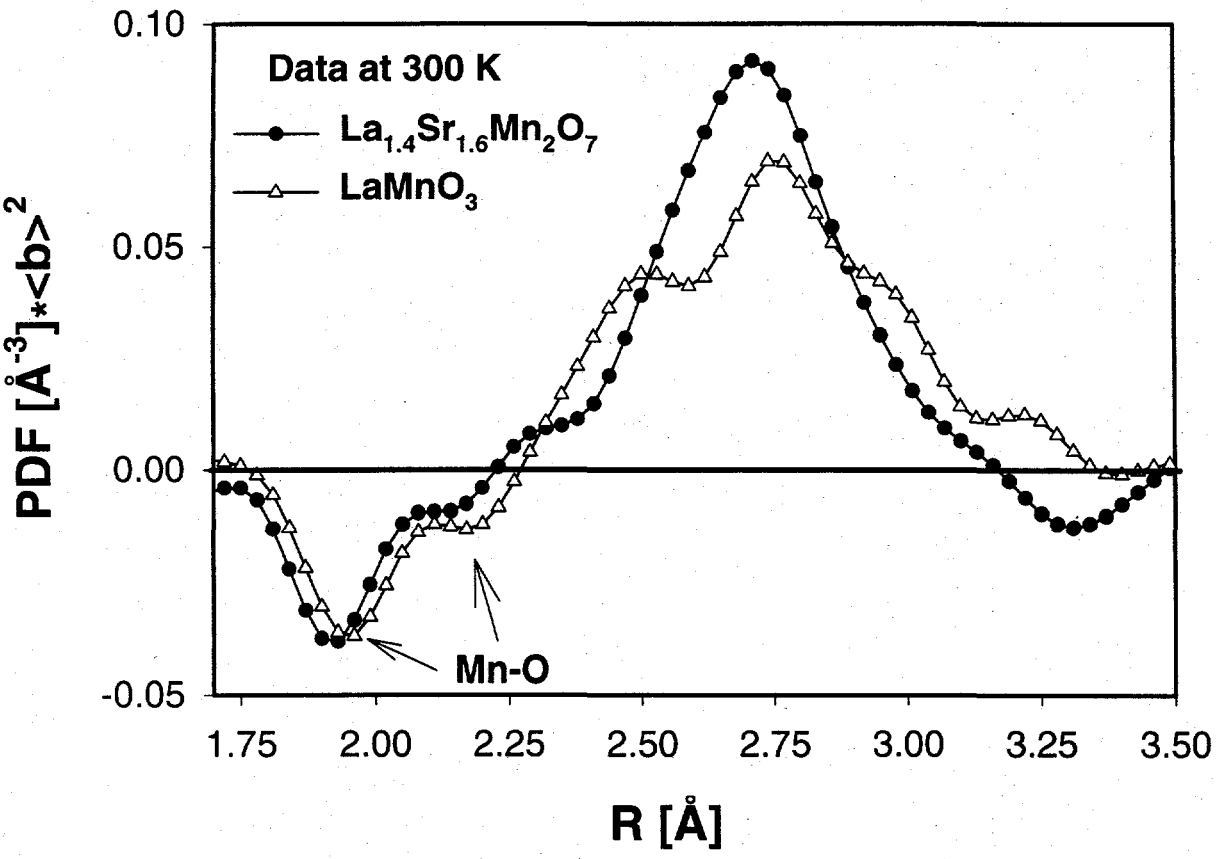
Figure captions:

Figure 1: The number of nearest O neighbors of Mn as a function of Sr doping at T = 300 K (320 K for x = 0.2, 350 K for x = 0.24 and 0.3, in diamonds), and 10 K (circles). In the small polaron model the charges are localized at $Mn^{4+}$ sites. At T ≈ RT, the data points are close to the "small polaron model" line. The solid line labeled "DE model" represents the predictions by the Zener-de Gennes model, while the chained line is a linear fit to the 10 K data. This result suggests that at room temperature a hole forms a single-site small polaron ($Mn^{4+}$), but at T = 10 K it is shared by about three octahedral sites forming a three-site polaron.

Figure 2: The local structure of the layered compound is compared to the one obtained from the pure $LaMnO_3$ at RT. Note that a split in the Mn-O peak is seen, at 1.92 and at 2.12 Å corresponding to short and long bonds, respectively, is as pronounced as the one observed in the perovskite and serves as a strong indication that the Jahn-Teller distortion is present in this system as in the perovskites.




D. LOUCA et al., Fig 1




D. LOUCA et al., Fig. 2