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# UNIVERSAL RELATIONSHIP BETWEEN MAGNETIZATION AND CHANGES IN THE LOCAL STRUCTURE IN QUASICUBIC AND BILAYER MANGANITES

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We extend our previous EXAFS investigations on the relationship between magnetization and local distortions to the bilayer materials $La_{2-2x}Sr_{1+2x}Mn_2O_7$ and to higher magnetic fields, for quasicubic $La_{0.7}Ca_{0.3}MnO_3$. For each, there is a significant change in the broadening parameter $\sigma$ of the Mn-O pair distribution function (PDF) associated with polaron formation as T is increased through $T_c$. Applying a magnetic field reduces the local distortion of the Ca sample for temperatures near $T_c$. For samples with sharp transitions, there is still a significant change of $\sigma^2$ with T below $T_c$ when the sample is fully magnetized, i.e. distortions are still present and continuing to be removed as T is lowered well below $T_c$. A simple model is presented for the magnetization process.

*Keywords*: EXAFS; manganites; CMR.

## 1. Introduction

For the quasicubic manganites we have shown some limited evidence for a *universal* relationship between the decrease in local distortions and the magnetization M as T is decreased below $T_c$[1,2]. We show here that that behavior extends to other samples and at high magnetic fields up to $\sim$ 10T. Of particular interest is that for samples with a very sharp magnetic transition, the sample is still clearly distorted just below $T_c$ when M has reached the low-T saturation level $M_0$. This means that the ferromagnetic coupling (double exchange or other models) is operative even when significant distortions of the Mn-O bonds are present. These distortions decreases as T is decreased to 10K. A similar result is found for the bilayer material - the transition is sharp and some distortion is still present when M $\sim$ $M_0$.

## 2. EXAFS Results

Transmission EXAFS data were collected as a function of T and B-field for the quasicubic materials using powdered samples, while polarized fluorescence EXAFS data (ab plane and c-axis) were collected as a function of T for the bilayer material using oriented single crystal samples. In Fig. 1 we plot the r-space data (Mn-O peak) for the 30% Ca quasi-cubic sample (function of B) and the ab-plane data for the 32 and 40% Sr bilayer samples (function of T); note that when the peak amplitude increases the disorder decreases. For each material an EXAFS analysis was made using FEFF standard functions[3]. One broadening parameter $\sigma$ for the Mn-O PDF was used in order to have a *single* structural parameter for comparison with M.

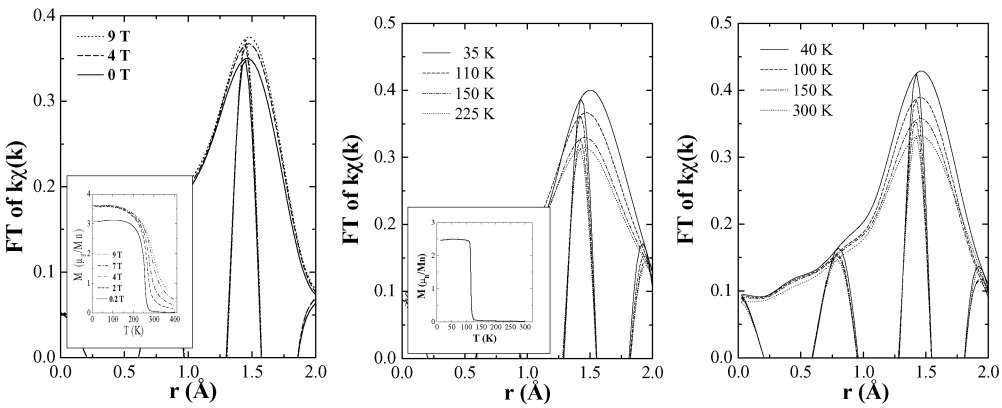


Fig. 1. r-space plots. left: 30% Ca sample at T = $T_c$ = 260K for various B-fields. mid.: 40% Sr ($T_c$ = 120K) and right: 32% Sr ($T_c$ = 115K), B = 0, at various temperatures for the polarization in the ab-plane. Insets shown M vs T (several B-fields for 30% Ca sample)

$\sigma^2$ vs T plots for the Ca-doped samples are shown in the insets (Fig. 2); the B-field induced reduction of the distortions for 30% Ca are mainly above $T_c$ in contrast to the earlier data on the 21% sample. Magnetostriction results on another CMR system, $Sm_{1-x}Sr_xMnO_3$, show similar behavior[4]. In addition, the 30% sample becomes nearly fully magnetized for T $\leq$ 150K, yet there is still a significant distortion present. It might be a small percentage of very highly distorted sites (a few sites (<3%) might not be magnetized) but then the distortion/site would have to be *larger* than in $LaMnO_3$; instead we propose there is still a small average distortion of all the sites. Clearly the coupling mechanism (e.g. double exchange) is operative when a distortion of the Mn-O bonds is still present. For the 32 and 40% Sr bilyaer samples, $\sigma^2$ again drops very rapidly with T below $T_c$ (not shown) but still has a significant value at T$\sim$100K when the sample becomes fully magnetized. Thus in the bilayer samples, distortions are also still present when the sample is in the metallic, ferromagnetic state.

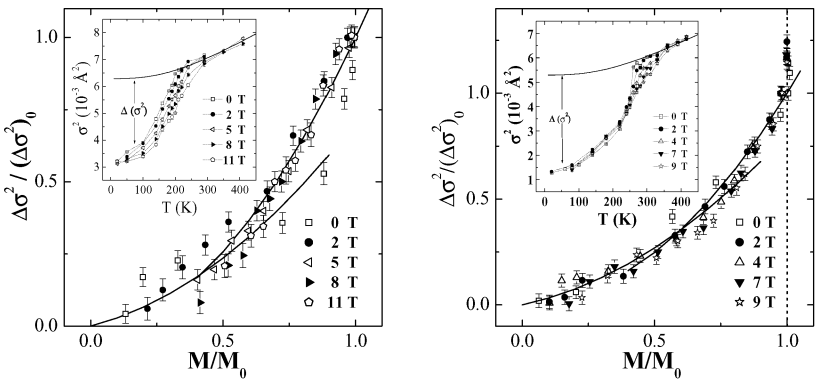


Fig. 2. $\Delta\sigma^2$ vs relative magnetization for the 21% (left) and 30% Ca samples (right) for various magnetic fields. $\Delta\sigma^2$ and $M$ have been normalized to their respective maximum (low-T) values at each magnetic field. Inset shows $\sigma^2$ vs T for several magnetic fields, and defines $\Delta\sigma^2$ as the decrease in $\sigma^2$, attributed to the loss of polaronic distortions, as T is lowered below $T_c$. The dotted lines are guides to the eye. The solid lines in the main figure represent a model whereby the sample becomes magnetized in pairs (hole and electron sites), but with a range of distortion for the electron sites; the least distorted electron sites become magnetized first.

Plots of $\Delta\sigma^2$ vs relative magnetization (Fig 2) show that the data fall roughly onto two lines with a break-point for $M/M_0$ just above 2 times the Ca concentration ( $M/M_0 \sim .45$-$.5$ for 21% and $\sim .65$-$.7$ for 30% Ca). Preliminary results for a 45% sample have the break near $M/M_0 = 0.9$. Conductivity plots vs $M/M_0$ (linear scale) show a similar break point at $M/M_0 = 0.65$-$0.7$ for a 30% sample. This suggests that initially, the magnetization develops through pairs - an undistorted hole site and a distorted electron site – this requires less energy to undistort the electron site per magnetized Mn. The distortions removed remain small (20-30%) until all the hole sites are used up - then the sample is at least 50% magnetized.

## 3. Conclusions

EXAFS measurements as a function of B and T indicate that $\Delta\sigma^2$ is a universal function of M. Very little local distortion is removed below $T_c$ till the sample is nearly 50% magnetized, suggesting a mechanism in which pairs of Mn atoms (hole and distorted site) become magnetized first. Distortions exist even when samples are fully magnetized. Similar results are found in the initial results for bilayer samples.

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