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EVIDENCE FOR A UNIVERSAL RELATIONSHIP BETWEEN MAGNETIZATION AND CHANGES IN THE LOCAL STRUCTURE

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X-ray Absorption Fine Structure (XAFS) measurements of the colossal magnetoresistance (CMR) sample $La_{0.79}Ca_{0.21}MnO_3$ at high fields indicate a decrease in the width parameter of the pair distribution function, σ, as the applied magnetic field is increased for T near T_c. The change in σ^2 from the disordered polaron state varies approximately exponentially with magnetization irrespective of whether the sample magnetization was achieved through a change in temperature or the application of an external magnetic field. This suggests a more universal relationship between local structure and the sample magnetization than was previously indicated.

Keywords: XAFS; $La_{0.79}Ca_{0.21}MnO_3$; CMR; polaron distortion.

1. Introduction

In previous studies, we have shown that as the temperature (T) is lowered below T_c, σ^2 decreases rapidly for CMR samples. This is attributed to a decrease in the amount of polaron-induced disorder. Others have also observed this in many XAFS and neutron pair distribution function analysis (NPDF) studies.[1–7] In addition, the change in σ^2 ($\Delta\sigma^2$) below T_c depends exponentially on the magnetization, M.[1,2] However, each point in plots of $\Delta\sigma^2$ versus M is at a different temperature.

In this study, we show that σ^2 also decreases as the applied B-field is increased at constant temperature. XAFS results indicate that $\Delta\sigma^2$ remains an exponential function of magnetization regardless of whether the sample magnetization was achieved through lowering the temperature or by applying a field.

2. Experimental Details

Mn K-edge data were collected on $La_{0.79}Ca_{0.21}MnO_3$ as a function of temperature and magnetic field at the Stanford Synchrotron Radiation Laboratory (SSRL) using beamline 7-2. The data were reduced using standard procedures and Fourier transformed to r-space. The r-space Mn–O peak was then fit using the RSFIT program, using standards calculated from FEFF6.[8] The number of neighbors was constrained to 6, and only the r-space peak position and the width parameter of the pair distribution function, σ, were allowed to vary.

3. Results

Our preliminary results show that there is a large temperature dependent change in the broadening parameter of the pair distribution function, σ, when polarons form near and above T_c (about 190 K for this sample) [see Fig. 1(a)]. At low temperatures the sample is ordered and σ^2 is small while at high temperatures there is a large amount of polaron-induced disorder. Furthermore, there is also a small field dependent change in σ^2. Near T_c, σ^2 decreases as the applied field is increased indicating that the application of a magnetic field removes polaron

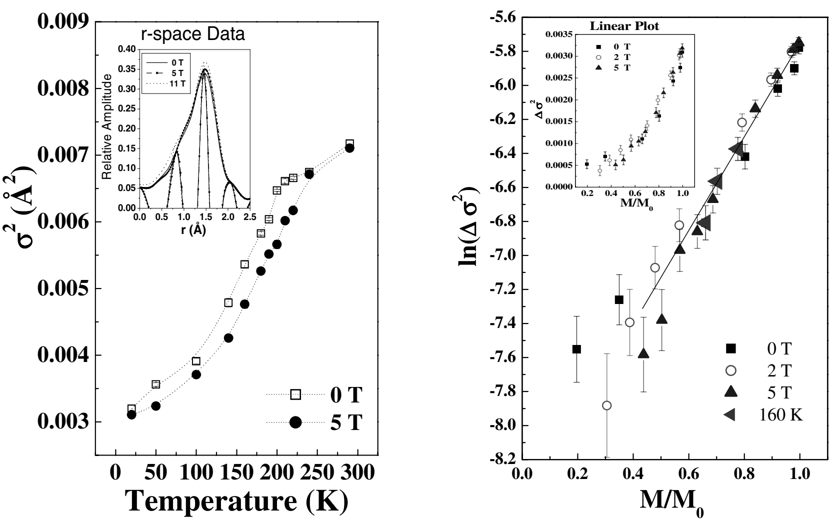

Fig. 1. (a) σ^2 versus T for the Mn-O peak for the 21% Ca sample. The dotted lines are guides to the eye. The insert shows the corresponding changes in the Mn–O peak at T_c (190 K) — the amplitude increases (σ^2 decreases) with increasing field. (b) $\ln(\Delta\sigma^2)$ versus relative magnetization for various fields. $\Delta\sigma^2$ is the decrease in σ^2 as T is lowered below T_c that is attributed to the loss of polaronic distortion. The insert shows a linear plot of $\Delta\sigma^2$ versus M/M_0.

disorder from the sample [see Fig. 1(a)]. The change in σ^2 is defined as

$$\Delta\sigma^2 = \sigma^2_{\rm T} + \sigma^2_{\rm FP} + \sigma^2_{\rm static} - \sigma^2_{\rm Mn-O}\,, \quad (1)$$

where $\sigma^2_{\rm T}$ is the thermal contribution calculated from[9] $CaMnO_3$ and $\sigma^2_{\rm Mn-O}$ is the data plotted in Fig. 1(a). The difference between $\sigma^2_{\rm T} + \sigma^2_{\rm static}$ and $\sigma^2_{\rm Mn-O}$ at high temperatures is called the full polaronic distortion, $\sigma^2_{\rm FP}$ in Eq. (1) above.[1,2] $\sigma^2_{\rm static}$ is the excess (above $\sigma^2_{\rm T}$) contribution at low temperatures.

In Fig. 1(b), $\ln(\Delta\sigma^2)$ vs M/M_0 is plotted for several fields (M_0 is the saturation magnetization at low temperature). There is a linear relationship between $\ln(\Delta\sigma^2)$ and M/M_0 for a relative magnetization above 0.5. However, each of these points is at a different temperature. For comparison, three points at the same temperature (160 K) are also shown to lie along the same line [see Fig. 1(b)]. Thus, we have extended our previous results[1,2] to show that the relationship between σ^2 and magnetization, given by $\ln(\Delta\sigma^2) = A\ (M/M_0) + B$, where A and B are constants, is more general. The relatively slow change in $\Delta\sigma^2$ at low M suggests that the low-distortion sites become magnetized first, possibly in pairs — an undistorted "Mn^{+4}" site and a distorted "Mn^{+3}" site. Further analysis needs to be done to investigate the nature of this relationship.

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