Electron spin resonance study of Nd 0.7 Sr 0.3 Mn O 3 single crystals

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Electron spin resonance study of $Nd_{0.7}Sr_{0.3}MnO_3$ single crystals

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Electron spin resonance (ESR) measurements in $Nd_{0.7}Sr_{0.3}MnO_3$ single crystals provide evidence for the existence of spin and lattice correlated clusters above and below T_C=205 K. The ESR linewidth deviates from the quasilinear "universal" temperature dependence of the linewidth observed in several manganites. A linear temperature dependence of the linewidth is attributed to the spin-phonon interaction. Thus the linewidth of the paramagnetic resonance line indicates the presence of a strong spin-phonon interaction rather than the spin-spin interaction seen in other manganites. The gradual increase observed in the g value above T_C is attributed to the presence of orbital correlations. The split in the ESR spectra only below 180 K (T_C=205 K) and more noise in the spectra between 150 and 180 K are ascribed to the strong competition between localization due to lattice distortions and delocalization of charge carriers. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1846613]

I. INTRODUCTION

Perovskite manganites, $R_{1-x}A_xMnO_3$ (where R is the rare-earth ion, such as La, Nd, Pr, etc. and A is the divalent alkaline earth ion, such as Ca, Sr, Ba, Pb, etc.), have been the subject of intense research from fundamental as well as application view points.[1] It has been shown that other interactions, such as electron-phonon interaction, intersite exchange interaction between the e_g orbitals [orbital ordering (OO) tendency], intrasite and intersite Coulomb repulsion interactions among the e_g electrons, antiferromagnetic (AFM) superexchange interaction between the t_{2g} local spins etc., compete with the ferromagnetic (FM) double exchange interaction in manganites and result in electronic phase separation.[2] Various experimental and theoretical results suggest the coexistence of FM and AFM and/or charge ordered phases in the ground state of manganites either in macroscopic form or, more often, through the presence of nanoclusters of one phase embedded into another.[3] The X-band electron spin resonance (ESR) measurements on $Nd_{0.7}Sr_{0.3}MnO_3$ single crystal reported here suggest the importance of the presence of such nanoclusters in the paramagnetic as well as ferromagnetic states.

II. EXPERIMENT

The NSMO (0.3) single crystal used in this study was grown by the floating-zone method using a double ellipsoidal infrared image furnace.[4] The crystal was characterized by x-ray diffraction, dc electrical resistivity, and magnetization measurements. The resistivity shows a peak at 214 K (T_P) whereas magnetization shows a sharp paramagnetic-to-ferromagnetic transition at 205 K (T_C). The ESR measurements were performed in the temperature range of 380–100 K. The c axis of the crystal was kept parallel to the magnetic field.

III. RESULTS AND DISCUSSION

Figure 1 shows the representative ESR spectra in the paramagnetic and ferromagnetic states. A single asymmetric line was observed in the paramagnetic state. There was no change in line shape at T_C (205 K). The spectrum splits into prominent low-field and high-field ferromagnetic resonance (FMR) lines only below 180 K. The lines are well separated. A shoulder develops on the low-field side of the low-field line, grows in intensity with decreasing temperature, and merges with the prominent low-field line below 140 K. The spectra become less noisy below 130 K compared to the spectra between 140 and 180 K. The asymmetric ESR spec-

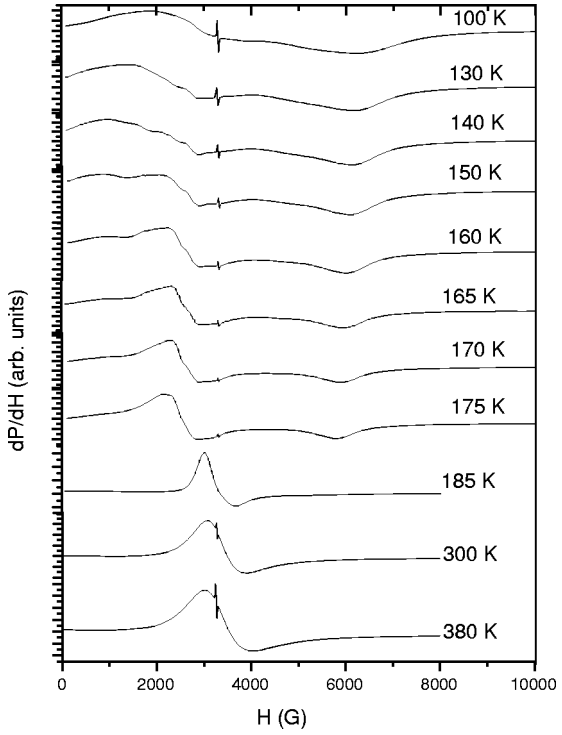

FIG. 1. ESR spectra of NSMO (0.3) crystal in the paramagnetic state and ferromagnetic state.

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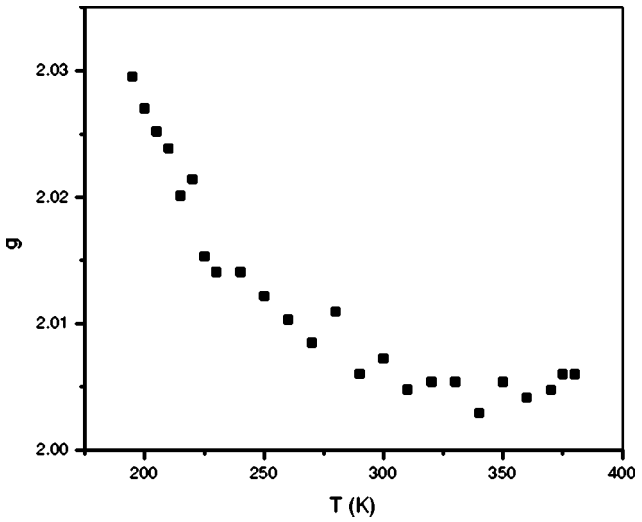

FIG. 2. Temperature dependence of g for NSMO (0.3) crystal.

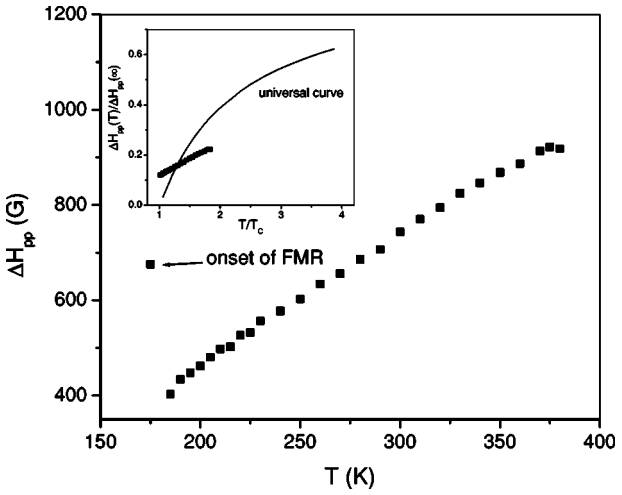

FIG. 3. ΔH_{pp} vs temperature for NSMO (0.3) single crystal. Inset shows $\Delta H_{pp}(T)/\Delta H_{pp}(\infty)$ vs temperature for NSMO (0.3) and universal dependence of $\Delta H_{pp}(T)/\Delta H_{pp}(\infty)$ for manganites.

tra could be fitted to asymmetric Lorentzian line shapes which include both absorption and dispersion.[5] The paramagnetic spectra of the NSMO (0.3) crystal could be fitted to a single asymmetric Lorentzian line while four asymmetric Lorentzians were needed to fit to the ferromagnetic spectra below 180 K. The FMR spectra are discussed elsewhere. The temperature evolution of the spectra above and near 180 K is discussed here.

The skin depth is responsible for the observed asymmetric line shapes in the paramagnetic state of NSMO (0.3) crystals. The g value, which is calculated with respect to the 2,2–diphenyl(picrylhydrazyl) (DPPH) line, increases from 2.00 to 2.03 as the temperature decreases from 380 to 195 K (Fig. 2). Such a behavior is unusual in the paramagnetic state and it may arise from additional spin correlations. The g-value of $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal also showed an increase with decreasing temperature in the paramagnetic state.[5] A prominent increase in the g value of $Nd_{0.5}Ca_{0.5}MnO_3$ is observed in its charge/orbital ordered state ($T_{CO}=240$ K) well above the Neel temperature ($T_N=140$ K).[6] Orbital degrees of freedom in $LiCuVO_4$ manifest as an increase in g value in the paramagnetic state.[7] Considering these results, we suggest that orbital degrees of freedom influence the spin correlations in the paramagnetic state of NSMO (0.3).

The linewidth of the ESR line in the paramagnetic state (Fig. 3) is found to be linear. The inset of Fig. 3 shows that the linewidth deviates from the quasilinear "universal" temperature dependence of the linewidth observed in several manganites.[8] The quasilinear linewidth is dominated by the variations of $\chi(T)$,

$$\Delta H_{pp}(T) = \Delta H_{pp}(\infty)\left(\frac{C}{T\chi}\right), \tag{1}$$

where C/T is the single-ion (Curie) susceptibility, χ is the measured paramagnetic susceptibility of a magnetically coupled clusters, and $\Delta H_{pp}(\infty)$ is the linewidth expected at temperatures high enough for the dc susceptibility to follow a Curie–Weiss law. This behavior indicates that the linewidth does not reflect a coupling to the lattice vibrations but rather is caused by a spin-only mechanism. However, the deviation in the linewidth of NSMO (0.3) crystal clearly indicates that the spin-relaxation mechanism in NSMO (0.3) is different from that observed in other ferromagnetic metallic manganites.

Seehra *et al.*[9] explained linear dependence of linewidth in manganites using a mechanism including critical and noncritical contributions. The noncritical contribution arises from the spin-spin interactions for $T \gg T_C$ whereas the critical contribution is due to the spin-phonon interaction. The resulting expression has the form

$$\Delta H_{pp}(T) = \frac{[c + f(\varepsilon)]}{T\chi}, \tag{2}$$

where $f(\varepsilon)$ is the critical contribution to ΔH which is significant only for $\varepsilon=(T-T_C)/T_C \geq 0.1$ If there is no contribution to the linewidth from sources other than spin-spin interactions, c would approach a temperature-independent value for $T \gg T_C$. In this case, the product $\Delta HT\chi$ would approach a temperature-independent constant value for $T \gg T_C$. To investigate this, a plot of the product of $\Delta HT\chi$ vs T is shown in Fig. 4 and it is found to increase with an increase of temperature. Then, following the discussion of Seehra *et al.*, it is inferred that the observed temperature dependence of linewidth is due to the spin-phonon coupling in the paramagnetic state of the NSMO (0.3) crystal.

The double integrated intensity of the ESR spectra was calculated using the formula

$$I \propto \Delta H_{pp}^2 Y', \tag{3}$$

where $2Y'$ is the peak-to-peak derivative amplitude. The intensity is found to decrease exponentially as the temperature increases. This is characteristic of the presence of spin clusters and the clusters dissociate upon increasing temperature. The ESR intensity is multiplied by a constant, proportional to the quality factor and dimension of the cavity, to determine ESR susceptibility (χ_{esr}).[5] χ_{esr} follows the dc susceptibility χ_{dc} in the manganites studied.[5,8] χ_{esr} of NSMO (0.3) crystal deviates from χ_{dc} at temperatures below 1.5 T_C, as

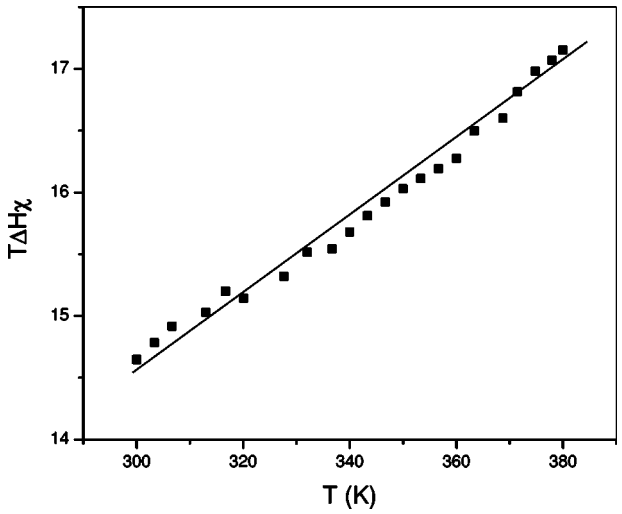

FIG. 4. Temperature dependence of the product $T\Delta H\chi$ for NSMO (0.3) crystal. Solid line is a guide to the eye.

shown in Fig. 5. This suggests that there is a marked deviation in the spin correlations in NSMO (0.3) near T_C compared to the other manganites, which points to the presence of a competing interaction which competes with the spin-spin interaction and grows in strength when the crystal is cooled.

The split in the ESR spectrum only below 180 K and the increased noise in the spectra between 180 and 140 K show

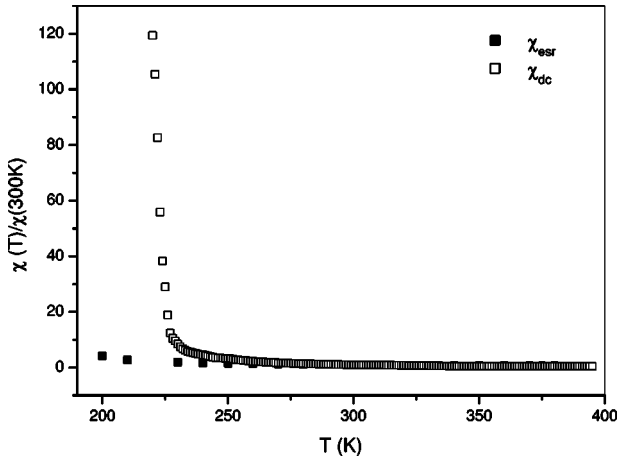

FIG. 5. Temperature dependence of χ_{esr} and χ_{dc} of NSMO (0.3) crystal normalized to χ(300 K).

the correlation of the lattice degrees of freedom on the onset of ferromagnetic spin order. Raman and ^{55}Mn NMR measurements on NSMO (0.3) crystals also show an abrupt increase in spin order below 140 K (Refs. 10 and 11) with prominent lattice correlations below T_C. This striking correspondence of the temperature evolution of spin dynamics in NSMO (0.3) as observed from different microscopic probes combined with the observation of spin clusters that are strongly coupled to lattice degrees of freedom possibly with orbital correlations in the paramagnetic state suggests that the spin clusters are strongly correlated with each other and significantly influence the spin dynamics in NSMO (0.3).

IV. CONCLUSIONS

The ESR study on the $Nd_{0.7}Sr_{0.3}MnO_3$ single crystals reveals the presence of spin-lattice correlated clusters above and below T_C. The linewidth, the g value of the paramagnetic spectrum, and the difference between χ_{ESR} and χ_{dc} close to T_C are indicative of the presence of strong electron-phonon interaction rather than the spin-only interaction seen in other manganites. The split in the ESR spectra only below 180 K (T_C=205 K) and more noise in the spectra between 150 and 180 K are attributed to lattice correlations.

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