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ESR study of $Nd_{0.5}Sr_{0.5}MnO_3$ single crystal

S. Angappane[a,*], M. Pattabiraman[a], G. Rangarajan[a], K. Sethupathi[a], G. Balakrishnan[b], D.McK. Paul[b], M.R. Lees[b], V.S. Sastry[c]

[a] *Low Temperature Laboratory, Department of Physics, Indian Institute of Technology, Chennai 600036, India*
[b] *Department of Physics, University of Warwick, Coventry CV4 7AL, UK*
[c] *Materials Science Division, Indira Gandhi Centre for Atomic Research, Kalpakkam 603 302, India*

Abstract

The paramagnetic g-value and linewidth of $Nd_{0.5}Sr_{0.5}MnO_5$ single crystal show the presence of spin clusters in the paramagnetic state. The anisotropy field splits the spectra below 250 K (T_C). An A-type antiferromagnetic phase ($T_N \sim 225$ K) is found to coexist with the ferromagnetic phase down to the charge ordering temperature of 150 K, at which a transition takes place also to a CE-type antiferromagnetic state.

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Keywords: Rare earth-transitionmetal compounds; Electron-spin resonance; Magnetic ordering; Charge ordering

The rare-earth manganese perovskites, ($RMnO_3$; R = La, Nd, Pr, etc.), when doped with alkaline earths, ($R_{1-x}A_xMnO_3$; A = Ca, Sr, Ba, etc.), exhibit colossal magnetoresistance (CMR) and an insulator-to-metal transition concomitant with a paramagnetic to ferromagnetic transition [1]. When the doping concentration is increased to $x = 0.5$, the ferromagnetic metallic phase, upon cooling, undergoes another transition at lower temperatures to a CE-type antiferromagnetic insulating phase. Many experimental studies [2] suggest that the CE state is inhomogeneous.

We report here an ESR study in single crystals of $Nd_{0.5}Sr_{0.5}MnO_3$ (NSMO 0.5) aimed at understanding the temperature evolution of the magnetically ordered phases. ESR measurements were performed using a X-band Varian E-112 continuous wave spectrometer from 425 to 100 K. A spherical crystal of diameter ~1 mm, whose axes were determined using a Laue camera in back-scattering geometry, was used for the measurement. The c-axis of the crystal was aligned parallel to the static magnetic field. The angular dependence was measured by rotating the sample with respect to an axis perpendicular to the magnetic field.

A single asymmetric line was observed in the paramagnetic state. The asymmetric line shape, which is observed up to 425 K, is due to the effect of the skin depth. The skin depth of our sample is calculated to be 0.03 mm, which is much less than the sample size of 1 mm. When the temperature is reduced below 250 K ($= T_C$), the spectrum splits, and the lines broaden. Two or three asymmetric Lorentzians [3] were used to fit the experimental data at various temperature ranges.

The g-value, which is calculated using DPPH as g marker, is found to increase with decreasing temperature in the paramagnetic state from 1.80 at 425 K to 1.96 at 250 K (Fig. 1a). This increase is attributed to orbitally ordered spin clusters based on the neutron diffraction study of Kawano et al. [4] on NSMO 0.5. A similar increase was seen in $Nd_{0.5}Ca_{0.5}MnO_3$ whose charge ordering temperature is higher than the antiferromagnetic transition [5]. Thus orbital degrees of freedom have an appreciable influence on the nature of these spin correlations in manganites even in the paramagnetic state.

The paramagnetic linewidth ΔH_{pp} is found to increase in a quasilinear manner with increase of temperature

*Corresponding author. Tel.: +91-044-22578663; fax: +91-044-22570509.
E-mail addresses: angappan@physics.iitm.ernet.in (S. Angappane), rajan@acer.iitm.ernet.in (G. Rangarajan).

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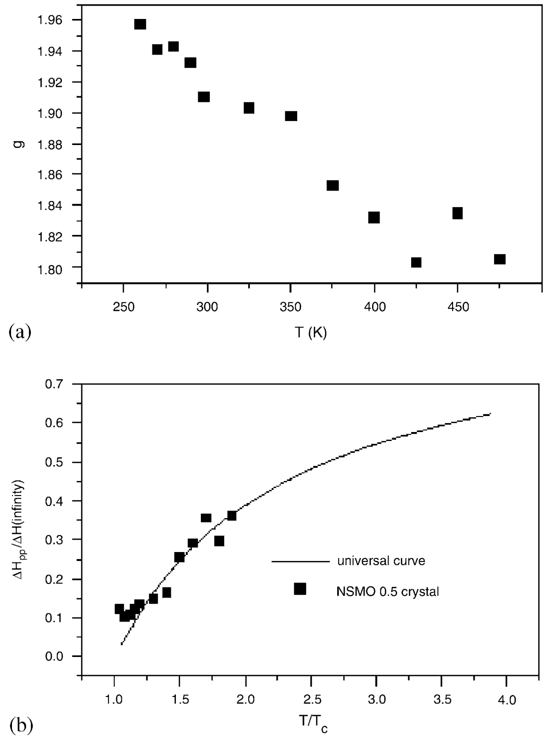

Fig. 1. (a) Plot of g vs. temperature in the paramagnetic state. (b) Plot of $\Delta H_{pp}/\Delta H_{pp}(\infty)$ vs. temperature in the paramagnetic state. The continuous line shows the calculated values.

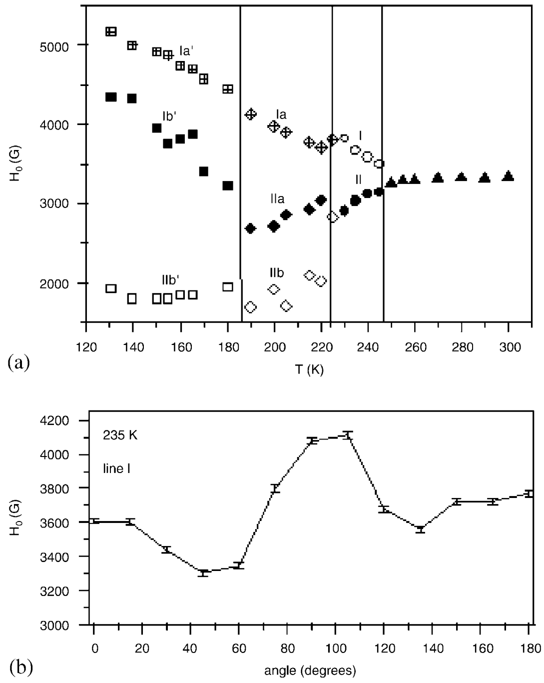

Fig. 2. (a) Plot of resonance field vs. Temperature. (b) Angular dependence of ESR spectra at 235 K.

and is described by an equation of the form [6]

$$\Delta H_{pp}(T) = \Delta H_{pp}(\infty)[C/T\chi],$$

where C/T is the single ion (Curie) susceptibility, χ is the measured paramagnetic susceptibility and ΔH_{pp} (∞) is the linewidth expected at temperatures high enough for the DC susceptibility to follow a Curie–Weiss law. This dependence of ΔH_{pp} (T) in manganites, which results in a *universal* temperature dependence of the ESR linewidth in a scale normalized to T_C, is attributed to the existence of magnetic clusters well above T_C. Thus the observed temperature dependence of the linewidth supports the existence of orbitally ordered spin clusters inferred from the temperature dependence of the paramagnetic g-value. Fig. 1b shows the linewidth of NSMO 0.5 single crystal plotted on this universal curve against the reduced temperature, T/T_C.

Between 250 and 230 K the spectra split into two lines (Fig. 2a). This line splitting could be due to the magnetocrystalline anisotropy of the ferromagnetic sample, when the field is not applied along the easy direction of magnetization. The presence of magnetocrystalline anisotropy was confirmed by the approximately sinusoidal angular variation of the resonance field of line I as shown in Fig. 2b. The asymmetry could be due to (temperature dependent) differences in the lattice parameters from their room temperature values. The single line, which is observed in the Q-band ESR measurements, shows that the magnetic field of 12500 G is sufficient to magnetize the sample completely in the field direction and that this field is greater than the anisotropy field.

Below 225 K, both lines I and II split further into two lines each (Fig. 2a). One of the split components of line I (Ib) was too weak to be analysed and its temperature dependence is not discussed any further. However, it is prominently observed when the angular variation of the lineshape was recorded at 180 K. This splitting could be due to the A-type antiferromagnetic state, which is found to coexist with the ferromagnetic state below 225 K as observed by Ritter et al. [7] from neutron diffraction. The sinusoidal angular dependence which was also found in the resonance fields of lines Ia and IIa indicates the existence of two different values for the anisotropy field in this temperature region.

Below 180 K, the resolution between lines Ia and IIa decreases markedly and a single asymmetric line is observed, line IIb still being present at low fields. The intensity of the lines is nearly constant down to the charge ordering temperature T_{CO} (150 K). This change in line shape coincides with the onset of the CE state below 180 K as shown by Ritter et al. [7].

In conclusion, the ESR measurements on NSMO 0.5 single crystal show the existence of orbitally ordered spin clusters in the paramagnetic state. Thus, orbital degrees of freedom have an appreciable influence on the dynamical spin correlations in manganites even in the paramagnetic state. The temperature evolution of coexisting ferromagnetic, A-type antiferromagnetic phases in the interval 250–180 K can be explained from variations in magnetocrystalline anisotropy. A further transition from A-type to CE-type of an ordering is also observed below 150 K.

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