

58916-00-105
58701-00-105
58701-40-105

ANL/MSD/CP-95884

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*Proceedings of International Conference on Neutron Scattering, Toronto, Canada,
August 17-21, 1997*

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This work supported by the U.S. Department of Energy, Basic Energy Sciences-Material Sciences
under contract #W-31-109-ENG-38 (JFM, RO, JDJ) and #W-7405-ENG-36 (TMK, DNA, RAR).

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197 ICNS⁺ Toronto Aug 17-21, 1997

Short Range Spin Correlations in the CMR Material $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$

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The $(\text{La}_{1-x}\text{Sr}_x)_3\text{Mn}_2\text{O}_7$ compounds are layered materials that exhibit higher magneto-resistance than the corresponding 3D manganite perovskites. Quasi-elastic neutron scattering on a polycrystalline sample of $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$ shows that the spin fluctuation spectrum of the these layered CMR materials is qualitatively similar to those found in the perovskite manganites $(\text{La},\text{Ca})\text{MnO}_3$; their concentration, lifetime, and coherence length increase as T decreases to T_C . Unlike the perovskites we find a lower spin-diffusion constant above T_C of ~ 5 meV \AA^2 .

Keywords: colossal-magneto-resistance, $(\text{La}_{1-x}\text{Sr}_x)_3\text{Mn}_2\text{O}_7$, magnetic polarons

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* Int'l Conf. on Neutron
Scattering¹⁾

The close interplay among charge, spin, and lattice degrees of freedom in the colossal magneto-resistive (CMR) manganite oxides is widely believed to play an important role in the mechanism of transport in these itinerant ferromagnets. Among the current models of transport in the three-dimensional magneto-resistive perovskite materials is that magnetic polarons—mobile lattice distortions carrying spin—play a fundamental role, at least above the Curie temperature (T_C).^{1,2} Supporting evidence for this model have been provided recently by DeTeresa, *et al.* from in-field small angle neutron scattering experiments on the perovskite $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$.² In this paper we report inelastic neutron scattering measurements from the layered CMR material $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$.

Inelastic neutron scattering data were taken from a 15g, single phase polycrystalline sample of $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$ using the time-of-flight chopper spectrometer PHAROS at MLNSC, Los Alamos National Laboratory. Data were measured as a function of temperature, using an incident energy $E_i=12.1$ meV, except for data measured at 115 K where incident energies of 12.1 and 8.1 meV were used to extend the Q-range of the measurement. Neutron powder diffraction data were measured as a function of temperature from 20-300K, using (SEPD) at IPNS at the Argonne National Laboratory. This polycrystalline sample was characterized using a.c. susceptibility and resistivity; a transition from a PI-FM was observed at 116K.

Fig. 1 shows the spectrum at 115 K and 30 K, with a gaussian fit to each: while there is little statistically significant deviation from the gaussian at 30 K, at 115 K the gaussian is clearly unable to fit the data. The addition of a lorentzian term $L(\varepsilon)=B\Gamma/\pi(\varepsilon^2+\Gamma^2)$ to fit the quasi-elastic component gives a much better fit to the data (ε is the energy transfer, B the Bose-Einstein factor, I the integrated intensity, and Γ the half-width at half maximum). This gives an intensity and a lifetime ($\tau = h/\Gamma$) for the spin correlations at each temperature (Fig. 2). The quasi elastic scattering diverges as $Q\rightarrow 0$, suggesting that the nature of the scattering is ferromagnetic. Similar observation have been made by Lynn *et al.*³ for the three dimensional perovskite manganites.

The intensity of the quasi-elastic scattering increases as the sample is cooled from 322 K to T_C , then decreases below T_C , with similar behavior for the lifetime. Fitting the intensity as a function of $|Q|$ gives a correlation length of ~ 12 Å at 128 K, while fitting the energy width as a function of $|Q|$ gives a spin-diffusion constant of ~ 5 meV Å². This value is substantially lower than the 30 meV Å² measured for $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ ³ and may reflect the effect of reduced dimensionality on the dynamics of spin fluctuations.

Neutron powder diffraction measurements from the same sample used for the inelastic scattering measurements, show that a relaxation of the lattice occurs at T_C . For the perovskite materials this has been interpreted as the relaxation of localized lattice distortion

from lattice polarons. The close correlation of the spin-dynamics (i.e. the increase of the quasi elastic scattering as $T \rightarrow T_C$), with the relaxation of the lattice suggests that these two phenomena are coupled. This observation is qualitatively similar to results reported by De Teresa *et al.*² for $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$. However, magnetic scattering from layered $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$ is complicated by the fact that in two-dimensional magnetic systems critical magnetic fluctuations can be enhanced well above T_C (e.g. K_2CuF_4 ⁴). Work is currently underway to decouple the expected 2-D ferromagnetic fluctuations from any additional signal that may emerge from magnetic polarons.

In conclusion, we observe a quasi-elastic magnetic scattering in $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$, between $0.7 < T_C < 2.8 T_C$. The intensity of the scattering and the lifetime of spin fluctuations increases as T decreases to T_C . This phenomenon correlates strongly with the electronic (insulator-metal) and structural transition at the onset of 3D magnetic ordering.

This work was supported by the U.S. Department of Energy, Basic Energy Sciences-Materials Sciences under contract W-7405-ENG-36 (TMK, DNA, RAR) and W-31-109-ENG-38 (JFM, RO, JDJ).

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Figure captions:

Fig. 1: Quasi-elastic spectrum from $\text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$ at 30 K (top) and 115 K (bottom).

The solid curves are gaussian fits.

Fig. 2: (a) Temperature dependent resistivity and magnetization (determined from Rietveld refinement of neutron powder data) (b) Intensity of the quasi-elastic component as a function of temperature. (c) Lifetime ($=\hbar/\Gamma$) as a function of temperature.



