# Magnetic correlations in the bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$

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Neutron scattering has been used to study the magnetic correlations in the double-layer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$, which exhibits colossal magnetoresistance in the vicinity of the ferromagnetic transition at $T_c = 112$ K. Over a large temperature range above $T_c$, there is evidence for two-dimensional magnetic correlations which peak in intensity at the transition. Although the in-plane correlations are predominantly ferromagnetic, a strong canting of spins in neighboring planes within the bilayers, at an angle that is dependent on both the temperature and magnetic field, is observed. There are indications of a crossover to three-dimensional critical scattering very close to $T_c$, but the nature of the phase transition has yet to be determined. © *1998 American Institute of Physics.* [S0021-8979(98)45411-2]

## I. INTRODUCTION

Colossal magnetoresistance can be strongly enhanced in systems with reduced dimensionality and so there has been considerable interest in the two-layer Ruddlesdon–Popper compounds, $La_{2-2x}Sr_{1+2x}Mn_2O_7$, where $x$ is the hole doping in the $MnO_2$ planes.[1–3] In zero field, these systems undergo a ferromagnetic phase transition at around 100 K with a concomitant metal-insulator transition. Although this transition temperature is much lower than in the three-dimensional Perovskite manganites, the reduced dimensionality leads to a significant extension of the temperature range over which magnetic correlations are important, and thereby allows a detailed examination of the link between local spin correlations and the resulting transport properties.

The origin of colossal magnetoresistance (CMR) was first explained by double-exchange interactions,[4–6] but recently it has been proposed that electron-phonon interactions play an important role in enhancing the CMR.[7] However, in the two-layer compounds, it was found that the influence of antiferromagnetic interactions competing with the ferromagnetic double exchange is of equal importance.[3,8] This aspect was first considered nearly 40 years ago by de Gennes,[6] but is only now being investigated further theoretically.[9] As part of a general study to investigate the link between the magnetic and transport properties of naturally layered manganites, we have been measuring the magnetic correlations using neutron diffraction and spectroscopy.

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## II. EXPERIMENTAL RESULTS

A single crystal of the double-layer compound $La_{1.2}Sr_{1.8}Mn_2O_7$ with dimensions $2\times4\times6$ mm$^3$ was grown using the floating-zone technique. Bulk measurements showed a ferromagnetic transition at $T_c \approx 112$ K in correspondence with the metal-insulator transition observed in resistivity measurements, both parallel and perpendicular to the $MnO_2$ planes. The lattice parameters are $a = 3.862$ Å and $c = 20.032$ Å at 125 K in agreement with earlier measurements for this composition.[2] More details on the preparation and characterization of this sample are given by Mitchell *et al.*[2] Neutron scattering experiments were performed on the triple-axis spectrometers BT2 and BT9 at the National Institute for Standards and Technology (NIST), Gaithersburg. Scans were taken in the $(0k0)$ scattering plane with incident neutron energies of 13.7 and 14.7 meV, without analyzing the energy of the scattered neutrons. A superconducting solenoid which provided vertical magnetic fields up to 7 T was employed in the measurements on BT9.

Diffuse scattering studies[10,11] show clear evidence of spin correlations above $T_c$ which have much longer correlation lengths within the $MnO_2$ planes than perpendicular to them. The two-dimensional character of these correlations is manifest in scattering in the form of rods perpendicular to the bilayer planes, i.e., parallel to $[00l]$ for all integer $h$ in the scattering geometry used. Figure 1 shows the temperature dependence of this rod-scattering measured on BT2 at $\mathbf{Q} = [1,0,1.833]$. At this particular value of momentum transfer (for the given incident energy), the wavevector $\mathbf{k}_f$ of the scattered neutrons is parallel to the $c$ axis, leading to an optimal energy integration. The diffuse scattering is strongly

 

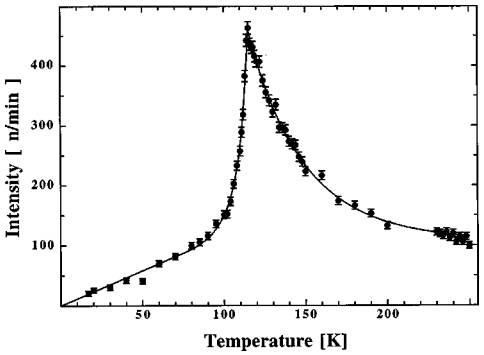


FIG. 1. Temperature dependence of the diffuse scattering measured at $Q=[1,0,1.833]$. A constant background, determined from a scan along $[h,0,1.833]$ at 4 K has been subtracted from the data. The solid line is a guide to the eye.

temperature dependent, becoming stronger in amplitude as the transition temperature $T_c=112$ K is approached and peaking sharply at $T_c$. Below $T_c$, the scattering arises from inelastic spin wave excitations and therefore rapidly decreases with decreasing temperature.

Because of the large separation between different bilayers, we can expect the exchange interaction along the $c$ axis to be small and the dispersion of the spin waves along this direction to be much weaker than in the plane. Recent spin wave measurements carried out on the cold triple-axis spectrometer SPINS at NIST indicated that the spin-wave energy at the zone boundary along the $c$ direction is only $\sim 0.5$ meV, nearly two orders of magnitude smaller than the in-plane zone boundary energy. For our experiments ($T>10$ K), the intensity is proportional to the magnon population, which is given by $n_q \sim k_B T/\hbar\omega_q$ when $k_B T \gg \hbar\omega_q$,[12] where $\omega_q$ is the spin wave energy along the $c$ axis with momentum $q$. The observed diffuse scattering (Fig. 1) follows this linear dependence up to $T\sim 80$ K where the scattering increases more rapidly. This is evidence of a softening of the spin wave energies as $T_C$ is approached.

The temperature and magnetic field dependence of the rods was studied by measuring both parallel and perpendicular to the rods at $[00l]$ and $[10l]$. Because of the large separation between different bilayers, the modulation of the diffuse scattering along the rods is determined by the spin correlations of Mn spins in neighboring layers within each bilayer, i.e., at $\mathbf{r}_i=\pm z\mathbf{c}$. The energy-integrated magnetic neutron scattering cross section for wavevector transfer $\mathbf{Q}$ is then proportional to[10]

$$S(\mathbf{Q}) \approx S^2(1+\langle\cos\,\theta\rangle\cos 4\pi z l). \tag{1}$$

Here $\theta$ is the in-plane cant angle of the two Mn spins. Since $z=0.0965$ in the 40% compound,[2] the rod scattering due to ferromagnetic correlations ($\theta=0$) would peak at $l=0$ and fall to zero at $l=2.59$. In-plane spin correlations produce a broadening of the rods along the $[h00]$ direction with the half-width equal to the inverse correlation length.

Figure 2 shows scans along $[0.95\,0\,l]$, displaced from the $[1\,0\,l]$ rod in order to avoid the nuclear Bragg peaks. For comparison, we have included data from Ref. 10 (upper part of Fig. 2). Clearly, the observed modulation along the rod is

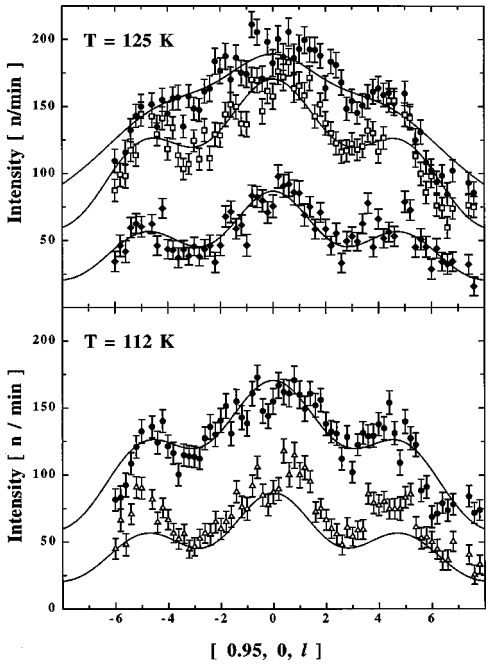


FIG. 2. Diffuse scattering along the $Q=[0.95,0,l]$ direction in a vertical magnetic field of 0 (circles), 0.5 (triangles), 1 (squares), and 2 T (diamonds). The solid lines are fits of Eq. (1) to the $T=125$ K data with $\theta=86.6°$, 74.1°, and 53°, respectively. The upper part of the figure is taken from Osborn *et al.* (Ref. 10).

not very strong, even close to $T_C$, showing that $\langle\cos\theta\rangle \ll 1$. Either the magnetic correlations between the neighboring planes are extremely weak, or the spins are strongly canted with respect to each other. The first possibility seems unlikely since the nearest neighbor distance between the planes is nearly identical to the distance within the planes. The ferromagnetic in-plane correlation length is already 9.7 Å at 125 K,[10] which would give $\langle\cos\theta\rangle\approx 0.67$ for nearest neighbors, whereas the weak modulation is consistent with a value of $\langle\cos\theta\rangle=0.06$. Such a difference would require the interaction between planes to be nearly two orders of magnitude weaker than within the planes. On the other hand, canting is predicted to occur when there is a competition between double exchange and superexchange interactions, as discussed below.

A magnetic field applied vertically along the $b$ axis should lead to a decrease of the cant angle and to a stronger modulation, in agreement with our observation. From least-squares fits of Eq. (1), we determined an average in-plane cant angle of 86.6° at $T=125$ K. This cant angle is reduced to 74.1° at $H=1$ T and 53° at $H=2$ T. Since, at this field strength, the diffuse scattering has become more inelastic and a substantial part of the scattering has been transferred to three-dimensional (3D) Bragg scattering, the diffuse signal has decreased strongly.

Reducing the temperature towards $T_C$ leads again to an increase in the modulation so that the zero field and 0.5 T data at 112 K are very similar to the 1 and 2 T data, respectively, at 125 K. This similarity is seen in the lower part of Fig. 2, where the data measured at 112 K are shown together with the best fits obtained for the 125 K data.

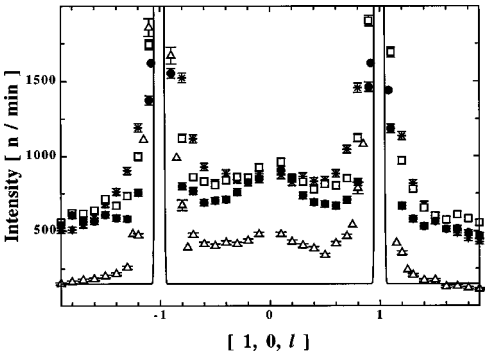


FIG. 3. Diffuse magnetic and nuclear scattering along the $Q=[10l]$ direction at $T=120$ (circles), 116 (squares), 112 (stars), and 4 K (triangles). The solid line gives an indication of the Bragg peaks widths at $[10\pm1]$.

Below 120 K, we observe an increase in the magnetic scattering around the $[1\,0\pm1]$ Bragg peaks along the $[10l]$ rod, which disappears again below $T_C$ (Fig. 3). Since the in-plane correlations have no influence on the modulation along the rod, this scattering is due to the build-up of correlations between different bilayers. In other words, 3D correlations are developing as the transition is approached. However, because of the strength of the nuclear Bragg peaks, it has not been possible to derive out-of-plane correlation lengths.

## III. DISCUSSION

Although measurements of spin wave dispersions, e.g., in $LaMnO_3$,[13] have provided evidence of competing ferromagnetic and antiferromagnetic exchange interactions of the usual Heisenberg form, the observation of canted spin correlations is clear evidence of a competition between double exchange and superexchange. Nearest-neighbor Heisenberg exchange will only produce collinear ferro- or antiferromagnetic ordering. However, because the energy of double exchange interactions for a pair of spins depends on $\cos(\theta/2)$,[5] rather than $\cos\theta$ as in the case of Heisenberg exchange, a minimum energy at $\theta\neq0$ or $\pi$ can result from a competition between the two.[6] The large cant angles observed in our experiments imply that the double exchange and superexchange interactions are of the same order of magnitude in this compound.

Evidently, lowering the temperature towards $T_C$ is equivalent to applying an in-plane magnetic field since both have the effect of reducing the cant angle. This is due to the growth of correlations between different bilayers, seen in Fig. 3, which produce an in-plane molecular field stabilizing ferromagnetic order and eventually lead to the 3D ordering at $T_C$. However, the nature of the transition is still uncertain. Earlier measurements of the in-plane correlation length indicate that it does not diverge at $T_C$[10] and so far no evidence of a crossover to 3D scaling was found. From the strong anisotropy in the spin wave dispersion below $T_C$ we estimate that the correlations between different bilayers are at least two orders of magnitude weaker than the correlations within the plane. Therefore, if there is a crossover to true 3D scaling, it would only occur within a narrow temperature range of about 1 K above $T_C$ and could be smeared by sample inhomogeneity.

A nondivergent correlation length on the other hand would indicate that the transition is first order, although the evidence of spin wave softening below $T_C$ (Fig. 1) and the absence of any hysteresis in this compound[14] show that, if the transition is first order, it is only weakly so. In the case of the three-dimensional CMR perovskites, it was found that the transition in $La_{2/3}Ca_{1/3}MnO_3$ is not a continuous second-order phase transition[15] and it was argued that the nondivergence of the correlation length is related to the formation of magnetic polarons above $T_C$.[16] However, it has not yet been established that this is a universal feature of CMR compounds, and from our present data, we cannot draw such a conclusion on the nature or the mechanism of the phase transition in the layered manganites.

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