Observation of Kosterlitz-Thouless spin correlations in the colossally magnetoresistive layered manganite La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$

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The spin correlations of the bilayer manganite La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ have been studied using neutron scattering. On cooling within the paramagnetic state, we observe purely two-dimensional behavior with a crossover to three-dimensional scaling close to the ferromagnetic transition. Below $T_C$, an effective finite size behavior is observed. The quantitative agreement of these observations with the conventional quasi two-dimensional Kosterlitz-Thouless model indicates that the phase transition is driven by the growth of magnetic correlations, which are only weakly coupled to polarons above $T_C$.

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It is now generally accepted that colossal magnetoresistance (CMR) in doped manganese oxides involves a strong coupling between spin, charge, and lattice degrees of freedom close to the ferromagnetic transition. However, the nature of the transition and the relative importance of the different interactions in controlling the magnetotransport above the critical temperature $T_C$ have not been clearly established. A number of studies have indicated that the magnetic phase transition may be unconventional in CMR compounds, with the observation of an anomalous spin diffusion component below $T_C$ and a non-divergent correlation length. This has been attributed to the development of magnetic polarons above $T_C$ and their possible persistence below $T_C$, although it can be difficult to distinguish them from standard critical fluctuations. In other studies, conventional critical scaling of bulk properties has been observed. It is important, therefore, to study the nature of the phase transition in CMR compounds, and determine if unconventional magnetic correlations are essential to the mechanism of CMR.

Naturally layered manganites are derived from the perovskite structure of the three-dimensional (3D) compounds by the addition of non-magnetic blocking layers. The bilayer compounds La$_{2-x}$Sr$_{1+x}$Mn$_2$O$_7$, in which $x$ represents the hole concentration on the Mn$_2$O$_2$ planes, have been extensively studied in recent years because of the insights they provide into the mechanisms of CMR. The motivation of the present study is to utilize the low-dimensionality of these compounds to perform a detailed investigation of the spin correlations close to $T_C$ using neutron scattering. The reduced dimensionality of the spin fluctuations extends the temperature region over which critical fluctuations may be studied, and makes them easier to distinguish from other dynamic processes in the sample. This has allowed us to compare the temperature evolution of magnetic correlations with other two-dimensional (2D) systems exhibiting similar magnetic ordering.

In previous work on the 40% hole-doped bilayer system we observed strong 2D in-plane ferromagnetic fluctuations above $T_C$, with evidence of competing ferromagnetic and antiferromagnetic interactions perpendicular to the planes. The in-plane correlation length $\xi$ was measured in scans through the 2D rods of magnetic scattering chosen to optimize the energy integration. However, these measurements are necessarily performed away from the wavevector corresponding to 3D magnetic ordering, and so were not sensitive to a possible crossover to 3D correlations.

We have now investigated the correlations close to the 3D ordering wavevector as a function of temperature, both above and below $T_C$. We show that the spin correlations are quantitatively consistent with a quasi-2D XY model, exhibiting Kosterlitz-Thouless correlations above $T_C$ with a crossover to 3D correlations at a correlation length consistent with the known in-plane and interbilayer exchange constants. This agreement with other 2D systems indicates that the magnetic correlations develop conventionally, and that the transition is a true second-order phase transition, although there is evidence that it is smeared by weak inhomogeneity. This is in contrast to a magnetic polaron model, in which the magnetic correlations are strongly bound to charge degrees of freedom. Recent x-ray and neutron scattering results provide evidence of charge localization and the development of charge correlations above $T_C$. Nevertheless, they do not have a strong influence on the magnetic correlations above $T_C$. Instead, the incipient charge ordering is preempted by the onset of ferromagnetic ordering, and the charge correlations collapse at $T_C$.

The neutron scattering experiments were performed at the NIST Center for Neutron Research, using the same single crystal of the 40% hole-doped bilayer manganite.
La$_{1.2}$Sr$_1.8$Mn$_2$O$_7$ (lattice parameters $a = 3.862$ Å and $c = 20.032$ Å at 125 K) that has already been studied in various other experiments [8–11]. This compound shows a transition to long-range ferromagnetic order with the Mn spins aligned entirely within the $a$-$b$ plane at $T_C \approx 113$ K, coincident with the metal-insulator transition obtained from a Lorentzian profile analysis of the scans [6]. The inset shows the temperature dependence of the (002) Bragg peak from Ref. [8] and the result of a fit to a Lorentzian profile convoluted with the instrumental resolution function. The inset shows the temperature dependence of the (002) Bragg peak from Ref. [8] and the result of a fit to a standard power-law behavior convoluted with a Gaussian $T_C$-distribution.

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The magnetic diffuse scattering was measured on the BT2 triple-axis spectrometer operating in two-axis mode, i.e., without analyzing the energy of the scattered neutrons. Scans were taken around the (002) Bragg position where the nuclear scattering contribution is extremely weak, with a fixed incident energy 13.7 meV and horizontal collimations of $60^\circ$-$20^\circ$-$20^\circ$, full width at half maximum (FWHM). Pyrolitic graphite was used both as monochromator and filter against higher order contamination. In this two-axis mode, the diffuse scattering is, in the quasistatic approximation, proportional to the wavevector-dependent susceptibility $\chi_T(q)$, where $q = Q - \tau$, $Q$ is the momentum transfer and $\tau$ denotes a reciprocal lattice vector of the magnetic structure.

Figure 1 shows the magnetic diffuse scattering observed in scans along $Q = [h02]$. The susceptibility is well described by the usual Lorentzian profile except for temperatures close to $T_C$. In this region, however, there is also a strong tail in the order parameter that was attributed to an inhomogeneous broadening of the transition common to disordered systems like the doped manganites [6]. Because of the strong dependence of $T_C$ on hole doping in the bilayer manganites [12], such sample inhomogeneities would manifest themselves as a distribution of $T_C$. We have therefore reanalyzed the scaling behavior of the order parameter, allowing for a Gaussian distribution of $T_C$‘s. As the inset to Fig. 1 shows, such a distribution is in excellent agreement with the observations. The newly obtained values of the order parameter are $\beta = 0.14(1)$ and $T_C = 113.2(2)$ K, compared to $\beta = 0.13(1)$ and $T_C = 111.7(2)$ K reported in Ref. [8]. The derived standard deviation $\sigma_{T_C} = 1.6$ K of the Gaussian $T_C$-distribution corresponds to less than 0.4% variation in the total hole-doping [12]. This shows that even a very small sample inhomogeneity strongly affects the measurements and has to be included for the analysis of critical quantities.

An analysis of the wavevector dependent susceptibility including the $T_C$-distribution requires a detailed knowledge of the phase transition, i.e., a model for the temperature dependence of the correlation length $\xi$ and the static susceptibility $\chi_T(0)$, which will be derived below. For the following discussion, we use the correlation lengths obtained from a Lorentzian lineshape analysis, which are shown in Fig. 1. For these to be valid, the scans need to integrate over all fluctuation frequencies. In 2D systems,
an optimal energy integration is achieved for a scattering geometry that has the scattered wavevector parallel to the c-axis \([002]\), but this is not kinematically possible in scans along \(Q = [h02]\). However, the agreement with previous measurements performed in the optimal configuration at \(Q = [1 + h,0,1.833]\) (Ref. 3) validates the quasi-static approximation in the present experiments.

In the layered manganites, the separation of the MnO2 bilayers by insulating (La,Sr)O layers leads to a strong anisotropy, both in transport properties and magnetic correlations. In a simple nearest-neighbor exchange Hamiltonian for the magnetic interactions, one therefore expects large differences between the various exchange constants: \(J_1\) between spins within the same plane, \(J_2\) between spins in different layers within a bilayer, and \(J_3\) between spins in different bilayers. This quasi-2D behavior has been verified by spin-wave measurements \([11,13,14]\), which yield \(J_1/J_2 \approx 150\), similar to the anisotropy observed in the resistivity \(\rho_{xx}\).

To our knowledge, the critical behavior of quasi-2D bilayer systems has not yet been investigated thoroughly, but there exist detailed theoretical and experimental studies of single-layer quasi-2D XY magnets (Q2DXY), in which there is a strong exchange anisotropy \(J/J'\), where \(J\) and \(J'\) are the in-plane and interlayer exchange constants, respectively \([17,20]\). In such systems, both above and below \(T_C\), spin fluctuations on length scales that are small compared to the characteristic length \(L_{\text{eff}} \approx \sqrt{J/J'}\) (in units of the nearest neighbor Mn-Mn distance \(a\)) are not affected by the interlayer coupling and are therefore purely 2D. When approaching \(T_C\) from above, one expects the correlation length and the static susceptibility \(\chi_T(q = 0)\) to increase according to the Kosterlitz-Thouless expressions

\[
\xi/a = \xi_0 \exp[b(T/T_{KT} - 1)^{-1/2}] \tag{1}
\]

and

\[
\chi_T(0) = C \exp[B(T/T_{KT} - 1)^{-1/2}], \tag{2}
\]

where \(T_{KT}\) is the topological ordering temperature of the 2D XY model \([21]\). Once the interlayer interaction becomes important, i.e., when the correlation length reaches the order of \(L_{\text{eff}}\), a crossover to 3D scaling is expected. Renormalization group theory estimates a 3D ordering temperature \(T_C\) that is related to \(T_{KT}\) by \([18]\)

\[
T_C = T_{KT}[1 + (b/\ln L_{\text{eff}})^2]. \tag{3}
\]

Below \(T_C\), the correlation length decreases rapidly to \(L_{\text{eff}}\), where spin fluctuations are again unaffected by the interlayer coupling and the correlation length remains constant. From this point on, although the magnetization is 3D, the fluctuations are 2D and the system can be modeled by a 2D system of effective size \(L_{\text{eff}}\) \([19]\). This behavior has been observed, for example, in \(\text{Rb}_2\text{CrCl}_4\) \([20]\).

As is shown in Fig. 3, our observations are in remarkable agreement with the above predictions of the Q2DXY model. From a least squares fit of Eq. (1) to the observed correlation length above 120 K, we obtain \(\xi_0 = 0.3(1)\), \(T_{KT} = 64(5)\) K, and \(b = 2.1(2)\), in excellent agreement with the theoretical value of \(b \approx 1.9\) \([19,21]\). For the static susceptibility we obtain \(B = 3.9(4)\), very close to the theoretical value \(B = b \ast (2 - \eta)\), where the critical exponent \(\eta < 1/4\). Using the fitted values for \(b\) and \(T_{KT}\) and \(J/J' = 150\), we obtain \(T_C \approx 109\) K, consistent with the value \(T_C \approx 113.2\) K derived from the order parameter.

The deviation from the purely 2D behavior above \(T_C\) occurs at a correlation length \(\xi_{2D} \approx 12\) \(\AA\), which also corresponds to the effective finite size observed below \(T_C\). This is in rough agreement with the value of \(L_{\text{eff}} \approx 40\) \(\AA\) predicted from the measured in-plane and interbilayer exchange constants. In the region around \(T_C\), we approximate the behavior of both the correlation length and the static susceptibility with an abrupt crossover to standard 3D scaling, although a more complete description would have to include a crossover scaling function \([22]\).

This model for the correlation length and the static susceptibility (shown as solid lines in Figs. 3 and 4) can now be used for an analysis of the peak lineshapes that includes the \(T_C\)-distribution. Figure 4 shows that this model gives a good description of the lineshape even close to \(T_C\). It is important to note that the model includes a divergence at \(T_C\) of both \(\xi\) and \(\chi_T(0)\), which is not apparent in the observed data. From a simple Lorentzian lineshape analysis, one would conclude erroneously that the correlation length does not diverge at \(T_C\).

Although the inhomogenous broadening prevents a reliable determination of the critical scaling within the 3D regime, it is evident from the above analysis that the magnetic correlations are qualitatively consistent with a conventional model of quasi-2D behavior. To underline
the significance of this conclusion to our understanding of CMR, we compare our results with the recent observation of charge correlations in the same compound \cite{9}. In the paramagnetic phase, we have observed a growth of diffuse x-ray and neutron scattering, arising from the strain field produced by quasistatic polarons. Furthermore, broad incommensurate peaks modulating this diffuse scattering show that these polarons become increasingly correlated with each other, producing short range charge scattering. These results suggest that these polarons become increasingly correlated with each other, producing short range charge ordering on lowering the temperature towards $T_C$. Both the quasistatic polaronic scattering and the charge correlation peaks start to collapse just above $T_C$, and disappear in the ferromagnetic, metallic state (see Fig. 3). It has been argued that these polarons explain the low hole mobility in the paramagnetic state, which cannot be due to double exchange alone \cite{6}.

The present observations are strong evidence that the phase transition in La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ is driven by the growth of magnetic correlations, which are only weakly coupled to the polarons above $T_C$. The polarons are likely to induce some exchange disorder, but it is evidently not sufficient to disrupt the development of critical magnetic fluctuations. This conclusion is not consistent with a magnetic polaron model, in which the spin correlations are strongly coupled to the charge degrees of freedom. In such models, the ferromagnetic phase transition is induced by a transition from small to large polarons \cite{3}, but it is unlikely that such a transition would mimic the scaling behavior observed here. This does not mean that such models cannot be relevant to other CMR compounds, particularly those with much stronger electronphonon coupling, but they are not essential to a description of the CMR process.

Although the spin-charge coupling is weak above $T_C$, the magnetic correlations ultimately drive the metal-insulator transition; once the spin correlations extend over a large enough region, the double exchange interaction can overcome the mechanism responsible for localizing the charges, inducing the polaron collapse \cite{3}. This may affect the critical scaling in the 3D regime and could be responsible for the unusually low value of $\beta$ (in quasi-2D XY systems, $\beta$ is predicted to be approximately 0.23) \cite{19}.

Our results demonstrate that the critical properties of the bilayer CMR manganite La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$, both above and below $T_C$, are in quantitative agreement with an effective finite-size 2D XY model. The magnetic and charge degrees of freedom are therefore only weakly coupled except close to $T_C$, where the growth of magnetic order delocalizes the polaronic charges. It is possible that some form of charge ordering would occur at lower temperature if it were not preempted by the magnetic ordering. From these observations, we conclude that magnetic polaron models are not appropriate to the present bilayer compound and are therefore not universal to CMR.

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