Low Energy Spin-Wave Excitations in the Bilayer Manganite La$_{2/3}$Sr$_{1/3}$MnO$_3$

S. Rosenkranz, R. Osborn, and J. F. Mitchell
Materials Science Division, Argonne National Laboratory, Argonne, IL 60439 USA

L. Vasiliu-Doloc and J. W. Lynn
NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg MD 20899 USA

S. K. Sinha
Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439 USA

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Low-energy spin wave excitations in the bilayer manganite \( \text{La}_{1,2}\text{Sr}_{1,8}\text{Mn}_2\text{O}_7 \)

S. Rosenkranz, a) R. Osborn, and J. F. Mitchell

Materials Science Division, Argonne National Laboratory, Argonne, IL 60439

L. Vasiliu-Doloc and J. W. Lynn

NIST Center for Neutron Research, National Institute for Standards and Technology, Gaithersburg, MD 20899 and

Department of Physics, University of Maryland, College Park, MD 20742

S. K. Sinha

— Advanced Photon Source, Argonne National Laboratory Argonne, IL 60439

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Inelastic neutron scattering experiments were performed on a single crystal of the bilayer manganite \( \text{La}_{1,2}\text{Sr}_{1,8}\text{Mn}_2\text{O}_7 \). Low energy spin-wave excitations were observed along the c direction with a maximum energy of \( \sim 0.5 \text{ meV} \) at the zone boundary. The dispersion of these acoustic spin wave modes is modeled by a nearest-neighbor Heisenberg model with an inter-bilayer exchange interaction between neighboring spins in different bilayers of 0.048(1) \text{ meV} \) and an anisotropy gap of \( \Delta = 0.077(3) \text{ meV} \). These results confirm the two-dimensional nature of the spin-correlations in the bilayer manganites, with a ratio of the in-plane to inter-bilayer interaction of \( \sim 200 \). The temperature dependence of the energies and intensities of the spin wave excitations are in agreement with our earlier conclusion that the ferromagnetic transition is second-order.

I. INTRODUCTION

The double-layer manganites \( \text{La}_{2-z}\text{Sr}_{z+2}\text{Mn}_2\text{O}_7 \), where \( z \) is the hole-doping in the \( \text{MnO}_2 \) layers, have provided important insights into the mechanisms of colossal magnetoresistance (CMR).\(^1\)\textsuperscript{-11} The lower dimensionality of these systems enhance the magnetic fluctuations in the paramagnetic phase, making it easier to study the interplay of spin, charge, and lattice correlations involved in producing the CMR effect.\(^2\)\textsuperscript{-4} In previous neutron scattering investigations of the spin correlations in the 40\% hole-doped bilayer compound, quasi-two-dimensional ferromagnetic spin-fluctuations above \( T_C = 113 \text{ K} \) were observed.\(^2\)\textsuperscript{-6} The reduced dimensionality was attributed to a strong anisotropy in the nearest neighbor exchange constants \( J_1 \approx J_2 \gg J_3 \) (see Fig. 1) caused by the \( \text{(La,Sr)O} \) layer separating the bilayers. More recently, we showed that these spin correlations develop conventionally according to the quasi-2D model for XY magnets and are ultimately responsible for the magnetic and concomitant metal-insulator phase transitions.\(^4\)

Inelastic neutron scattering investigations of the spin dynamics in the layered compounds have so far focused on determining the exchange constants at low temperature.\(^7\)\textsuperscript{-9} In view of the unconventional behavior observed in some three-dimensional compounds,\(^12\) it is important to investigate the behavior of the spin dynamics close to \( T_C \). We have therefore started an investigation of the low energy spin-wave excitations in the 40\% hole-doped bilayer manganite \( \text{La}_{1,2}\text{Sr}_{1,8}\text{Mn}_2\text{O}_7 \), using high resolution neutron spectroscopy.

\begin{figure}
\centering
\includegraphics[width=0.8\textwidth]{fig1.png}
\caption{Mn spin arrangement of \( \text{La}_{1,2}\text{Sr}_{1,8}\text{Mn}_2\text{O}_7 \) in the tetragonal unit cell with space group \( I4/mmm \). The exchange interactions \( J_1 \) (solid lines), \( J_2 \) (dotted line), and \( J_3 \) (dashed-dotted lines) included in the spin wave calculation are shown for layer \( C \).}
\end{figure}

II. THEORETICAL MODEL

In the doping range \( 0.34 \leq z \leq 0.4 \), the layered manganites \( \text{La}_{2-z}\text{Sr}_{z+2}\text{Mn}_2\text{O}_7 \) order ferromagnetically with the Mn spins aligned within the planes as shown in Fig. 1. For the spin wave calculation, we solve the Heisenberg Hamiltonian

\[ \mathcal{H} = -\frac{1}{2} \sum_{jj'} \sum_{kk'} J \left( \frac{\mathbf{S}_j \cdot \mathbf{S}_{j'}}{\mathbf{K}_k \cdot \mathbf{K}_{k'}} \right), \]

(1)
including the in-plane exchange interaction $J_1$ between neighboring spins within each layer, the intra-bilayer exchange $J_2$ between neighboring spins in different layers within each bilayer, and the inter-bilayer exchange $J_3$ between spins in different bilayers, as defined in Fig. 1. This Hamiltonian is easily diagonalized in reciprocal space using the standard Holstein-Primakoff approach. Since the primitive unit-cell contains a two spin basis, we obtain two spin-wave branches

$$\hbar \omega_{\lambda}(q)/S = -2J_1[\cos(qz_0a) + \cos(qy_0b)] + J_2 + 4J_3$$

$$-2J_1[\cos(qz_0a) + \cos(qy_0b)] \mp |\gamma|,$$  

(2a)

where $\lambda$ labels the acoustic (–) and optic (+) spin waves, respectively, and

$$\gamma = |\gamma| e^{i\phi}$$

$$= J_2 + 4J_3 \cos \left( \frac{qz_0a}{2} \right) \cos \left( \frac{qy_0b}{2} \right) \exp \left( -i \frac{qz_0c}{2} \right).$$  

(2b)

In terms of these excitations, the one-magnon cross section for unpolarized neutron scattering takes the form

$$\frac{\partial^2 \sigma}{\partial Q \partial \omega} \sim \frac{k_f}{k_i} \left( 1 + \frac{Q^2}{Q_s^2} \right) \left[ \frac{1}{2} g f(Q) \right]^2 e^{-2W(Q)}$$

$$\times \sum_{q\neq q_0} \left[ n_1(\bar{q}) + \frac{1}{2} \pm \frac{1}{2} \right] \delta \left[ \hbar \omega \mp \hbar \omega_{\lambda}(q) \right]$$

$$\times \delta(\bar{Q} \mp \bar{q} - \bar{r}) [1 \pm \cos(2zcQ_s + \phi)],$$  

(3)

where $Q = k_i - k_f$ is the momentum transfer, $k_i$ and $k_f$ are the wavevectors of the incoming and scattered neutrons, respectively, $\hbar \omega$ is the energy transfer, $\bar{r}$ is a reciprocal lattice vector, $W(Q)$ is the Debye-Waller factor, $\bar{q}_0$ is the position of the Mn spins within the unit cell, and $n_1(\bar{q})$ is the population factor of the spin-wave excitation $\hbar \omega_{\lambda}(q)$. The sign in the last factor of Eq. (3) refers to the acoustic (+) and optic (–) spin wave modes, whereas the other signs denote the creation and annihilation of a spin wave, respectively. For a strong exchange anisotropy $J_2 \gg J_3$, $\phi \approx 0$ and the neutron scattering intensity is modulated according to $1 \pm \cos(2zcQ_s)$, where $z$ defines the relative position $\bar{r}_A = (0,0,z)$ of the Mn spin within the I4/mmm unit cell. Since $z = 0.096$ in the 40% doped bilayer compound, the intensity of the acoustic mode peaks at $Q_z = 0$ and falls to zero at $l = Q_z c/2\pi = 2.59$, and vice versa for the optic mode.

III. EXPERIMENTAL RESULTS

A single crystal of dimensions 1x4x6 mm$^3$ of the 40% bilayer compound La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ was grown using the floating zone technique. This crystal was subsequently characterized and used in various different experiments. Neutron scattering measurements were conducted on the triple-axis spectrometer SPINS, installed on a cold neutron guide at NIST. Energy spectra were measured in constant-Q scans with fixed final energies $E_f = 3.7$ meV and 2.8 meV. A cooled Beryllium filter was used to suppress higher order contamination. The spin-wave dispersion along $\Gamma - Z$, corresponding to $q = [001]$, was measured at different temperatures in scans around (001) and (101). Figure 2 shows examples of spin wave excitations, both creation ($\hbar \omega > 0$) and annihilation ($\hbar \omega < 0$), measured at $Q = (1,0,0.2)$, i.e., at a reduced wavevector $q = (0,0.8)$ away from $\bar{r} = (101)$, at temperatures $T = 50$ K and 100 K, respectively. The resulting dispersion curve for the acoustic branch at $T = 50$ K is shown in Fig. 3. For $J_3/J_2 << 1$, the dispersion relation Eq. (2) for the acoustic mode along the $\Gamma-Z$ direction is well approximated by

$$\hbar \omega_{ac}(q_z) = 4J_3S \left[ 1 - \cos \left( \frac{q_z c}{2} \right) \right] + \Delta,$$  

(4)

where $\Delta$ accounts in first order for a possible anisotropy gap. With this approximation, we can obtain the interbilayer exchange constant from a least squares fit to the observed dispersion without needing to know the values of the intra-bilayer constant $J_2$, although the latter has been determined from thermal neutron scattering experiments to be $J_2 \approx 3$ meV.$^{7,8}$ As is seen in Fig. 3, allowing for an anisotropy gap gives much better agreement with the observed dispersion. The best fit value for the interbilayer coupling in the anisotropic case is $J_3S = 0.048(1)$ meV at $T = 50$ K with a gap $\Delta = 0.077(3)$ meV, whereas $J_3S = 0.064(2)$ meV when the gap is fixed at zero.

These values are in agreement with the recent investigation of a nominally equal composition by Chatterji et al.$^9$ However, the anisotropy gap $\Delta$ that we derive is nearly twice as large, either because we were able to mea-
FIG. 3. Spin wave dispersion along $q = (00q_z)$ in La$_{1.3}$Sr$_{1.8}$Mn$_2$O$_7$ measured at $Q = (101-q_z)$. The solid (dashed) line is the result of a least squares fit to Eq. (4) including (without) an anisotropy gap $\Delta$.

sure closer to the zone boundary or because of differences in the two sample compositions.$^{14}$

Previous inelastic neutron scattering investigations have determined the in-plane and inter-bilayer exchange interactions to be $J_1S = 10.1$ meV and $J_2S = 3.1$ meV, respectively.$^{7,8}$ The resulting anisotropy in the exchange constants $J_1/J_2 \approx 200$ is similar to the anisotropy observed in transport properties.$^{10}$ This directly verifies the quasi two-dimensionality of the spin correlations below $T_C$, which has previously been observed in the paramagnetic diffuse scattering above $T_C.$$^{2,6}$ In a detailed analysis of the correlation length and static susceptibility, both above and below $T_C$, we found that the spin correlations are in quantitative agreement with the conventional model for quasi-2D XY magnets with a crossover to three-dimensional scaling close to $T_C$. We therefore concluded that the phase transition is driven by conventional magnetic correlations rather than the growth of magnetic polarons.$^4$ However, the strong dependence of the transition temperature on hole-doping$^{11}$ leads to a smeared transition in the form of a Gaussian $T_C$-distribution with a standard deviation of 1.5 K for 0.4% variation in the hole doping, which makes the investigation of the true critical behavior difficult.

Figure 4 shows the temperature dependence of the spin-wave excitation energy at the zone boundary (Z-point) together with the bulk magnetization measured by elastic neutron scattering.$^2$ In the random phase approximation, the spin wave energy is proportional to the magnetization in reasonable agreement with our observations. There is also no evidence of an anomalous loss of intensity as observed in some perovskite compounds$^{12}$, confirming that the transition is a conventional second-order one in the present compound.

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$^{a)}$ Electronic mail: SRosenkranz@anl.gov

$^{b)}$ Present address: Department of Physics, Northern Illinois University, De Kalb, IL 60115 and Argonne National Laboratory, Argonne, IL 60439.


$^3$ L. Vasiliu-Doloc et al., preprint, cond-mat/9907304 (1999).


$^8$ G. Chaboussant et al., unpublished.


$^{13}$ We operate in the reciprocal lattice of the body centered $I4/mmm$ chemical unit cell. A reciprocal lattice vector $\mathbf{q} = (hkl)$ therefore satisfies $h + k + l = \text{even}$.

$^{14}$ Differences in sample composition are suggested by the larger value of $T_C = 128$ K reported in Ref. 9. This would be consistent with a lower hole concentration in their sample, see Ref. 11.