Low-energy spin-wave excitations in the bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$

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Inelastic neutron scattering experiments were performed on a single crystal of the bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$. Low-energy spin-wave excitations were observed along the c direction with a maximum energy of $\sim$ 0.5 meV at the zone boundary. The dispersion of these acoustic spin wave modes is modeled by a nearest-neighbor Heisenberg model with an interbilayer exchange interaction between neighboring spins in different bilayers of 0.048(1) meV and an anisotropy gap of $\Delta = 0.077(3)$ meV. These results confirm the two-dimensional nature of the spin correlations in the bilayer manganites, with a ratio of the in-plane to interbilayer 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. [S0021-8979(00)42508-9]

I. INTRODUCTION

The double-layer manganites $La_{2-2x}Sr_{1+2x}Mn_2O_7$, where x is the hole doping in the MnO_2 layers, have provided important insights into the mechanisms of colossal magnetoresistance (CMR).[1–11] The lower dimensionality of these systems enhances 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.[2–4] In previous neutron scattering investigations of the spin correlations in the 40% hole-doped bilayer compound, quasi two-dimensional (2D) ferromagnetic spin fluctuations above $T_C \approx 113$ K were observed.[2,6] The reduced dimensionality was attributed to a strong anisotropy in the nearest-neighbor exchange constants $J_1 \sim J_2 \gg J_3$ (see Fig. 1) caused by the (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.[7–9] 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 $La_{1.2}Sr_{1.8}Mn_2O_7$, using high resolution neutron spectroscopy.

II. THEORETICAL MODEL

In the doping range $0.34 \leqslant x \leqslant 0.4$, the layered manganites $La_{2-2x}Sr_{1+2x}Mn_2O_7$ 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

$$\mathcal{H} = -\frac{1}{2}\sum_{jj'}\sum_{kk'} J\binom{jj'}{kk'}\mathbf{S}_j^k\mathbf{S}_{j'}^{k'}, \tag{1}$$

including the in-plane exchange interaction J_1 between neighboring spins within each layer, the intrabilayer exchange J_2 between neighboring spins in different layers

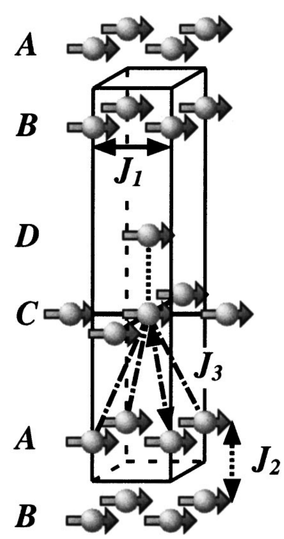

FIG. 1. Mn spin arrangement of $La_{1.2}Sr_{1.8}Mn_2O_7$ in the tetragonal unit cell with space group $I4/mmm$. The exchange interactions J_1 (solid lines), J_2 (dotted line), and J_3 (dashed–dotted lines) included in the spin wave calculation are shown for layer C.

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within each bilayer, and the interbilayer exchange J_3 between spins in different bilayers, as defined in Fig. 1. This Hamiltonian is easily diagonalized in reciprocal space[13] using the standard Holstein–Primakoff approach. Since the primitive unit cell contains a two spin basis, we obtain two spin-wave branches

$$\hbar\omega_\lambda(\mathbf{q})/S = 4J_1 + J_2 + 4J_3 - 2J_1[\cos(q_x a) + \cos(q_y b)] \mp |\gamma|, \tag{2a}$$

where λ labels the acoustic $(-)$ and optic $(+)$ spin waves, respectively, and

$$\gamma = |\gamma| e^{i\phi} = J_2 + 4J_3 \cos\left(\frac{q_x a}{2}\right)\cos\left(\frac{q_y b}{2}\right)\exp\left(-i\frac{q_z c}{2}\right). \tag{2b}$$

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

$$\frac{\partial^2\sigma}{\partial\Omega\,\partial\omega} \sim \frac{k_f}{k_i}\left(1 + \frac{Q_x^2}{Q^2}\right)\left[\frac{1}{2} g f(Q)\right]^2 e^{-2W(Q)} \times \sum_{\mathbf{q}\tau\lambda}\left[n_\lambda(\mathbf{q}) + \frac{1}{2} \pm \frac{1}{2}\right]\delta[\hbar\omega \mp \hbar\omega_\lambda(\mathbf{q})] \times \delta[\mathbf{Q} \mp \mathbf{q} - \boldsymbol{\tau}][1 \pm \cos(2zcQ_z + \phi)], \tag{3}$$

where $\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$ is the momentum transfer, $\mathbf{k}_i$ and $\mathbf{k}_f$ are the wave vectors of the incoming and scattered neutrons, respectively, $\hbar\omega$ is the energy transfer, $\boldsymbol{\tau}$ is a reciprocal lattice vector,[13] $f(Q)$ is the magnetic form factor, $W(Q)$ is the Debye–Waller factor, and $n_\lambda(\mathbf{q})$ is the population factor of the spin-wave excitation $\hbar\omega_\lambda(\mathbf{q})$. 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 $1 \pm \cos(2zcQ_c)$, where z defines the relative position $\mathbf{r}_{\mathrm{Mn}} = (0,0,z)$ of the Mn ion within the $I4/mmm$ unit cell. Since $z = 0.096$ in the 40% doped bilayer compound,[1] the intensity of the acoustic mode peaks at $Q_z = 0$ and falls to zero at $l = Q_z c/2\pi \approx 2.59$, and *vice versa* for the optic mode.

III. EXPERIMENTAL RESULTS

A single crystal of dimensions $1 \times 4 \times 6$ mm^3 of the 40% bilayer compound $La_{1.2}Sr_{1.8}Mn_2O_7$ was grown using the floating zone technique. This crystal was subsequently characterized and used in various different experiments.[2–5] 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 $E_f = 3.7$ and 2.8 meV. A cooled beryllium filter was used to suppress higher order contamination.

The spin-wave dispersion along $\Gamma - Z$, corresponding to $\mathbf{q} = [00q_z]$, was measured at different temperatures in scans around $(00l)$ and $(10l)$. Figure 2 shows examples of spin wave excitations, both in creation ($\hbar\omega > 0$) and annihilation ($\hbar\omega < 0$), measured at $\mathbf{Q} = (1,0,0.2)$, i. e., at a reduced wave vector $\mathbf{q} = (0,0,0.8)$ away from $\boldsymbol{\tau} = (101)$, 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 $J_3/J_2 \ll 1$, the dispersion relation Eq. (2) for the acoustic mode along the Γ-Z direction is well approximated by

$$\hbar\omega_{\mathrm{ac}}(q_z) = 4J_3 S\left[1 - \cos\left(\frac{q_z c}{2}\right)\right] + \Delta, \tag{4}$$

where Δ 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 intrabilayer constant J_2, although the latter has been determined from thermal neutron scattering experiments to be $J_2 S \approx 3$ 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 $J_3 S = 0.048(1)$ meV at $T = 50$ K with a gap $\Delta = 0.077(3)$ meV, whereas $J_3 S = 0.064(2)$ 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 Δ that we derive is nearly twice as large, either because we were able to measure closer to the zone boundary or because of differences in the two sample compositions. Differences in sample composition are sug-

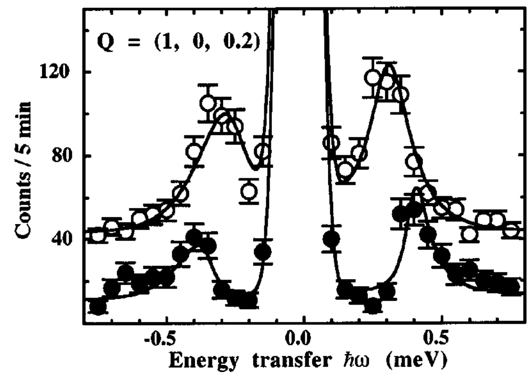

FIG. 2. Spin wave excitations in $La_{1.2}Sr_{1.8}Mn_2O_7$ measured on SPINS with fixed final energy $E_f = 3.7$ meV at $T = 50$ K (solid circles) and $T = 110$ K (open circles, shifted by 30 counts), respectively.

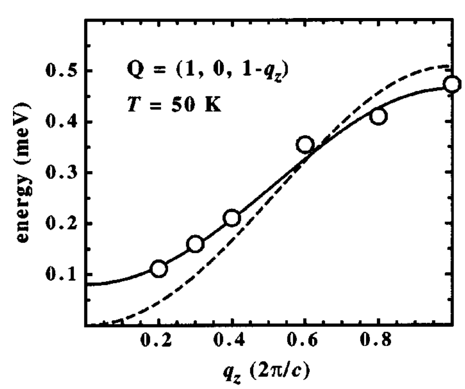

FIG. 3. Spin wave dispersion along $\mathbf{q} = (00q_z)$ in $La_{1.2}Sr_{1.8}Mn_2O_7$ measured at $\mathbf{Q} = (1,0,1-q_z)$. The solid (dashed) line is the result of a least squares fit to Eq. (4) including (without) an anisotropy gap Δ.

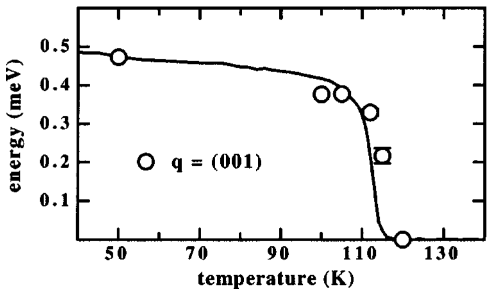

FIG. 4. Temperature dependence of the *Z* point acoustic spin wave excitation in $La_{1.2}Sr_{1.8}Mn_2O_7$. Overplotted (in arbitrary units) is the magnetization (line) obtained from neutron diffraction measurements of the (002) Bragg peak, from Ref. 2.

gested by the larger value of T_C =128 K reported in Ref. 9, which would be consistent with a lower hole concentration in their sample, see Ref. 11.

Previous inelastic neutron scattering investigations have determined the in-plane and interbilayer 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 $J_1/J_3\approx200$ 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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