RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 60, NUMBER 10 1 SEPTEMBER 1999-II

Spin waves in the quasi-two-dimensional ferromagnetic bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$

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(Received 28 December 1998)

We have used inelastic-neutron-scattering measurements to investigate the magnetic excitations in a layered manganite $La_{1.2}Sr_{1.8}Mn_2O_7$ which shows colossal magnetoresistance behavior. We have measured the dispersion of both the acoustic and the optic modes along [100] and [001]. By analyzing these results within an effective exchange model of weakly coupled ferromagnetic bilayers we have determined the complete set of intra- and interbilayer exchange interactions of an effective localized spin model. We discuss evidence for the quasi-two-dimensional ferromagnetism in this compound. [S0163-1829(99)51034-5]

The recent discovery of colossal magnetoresistance (CMR) in the bilayer manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ (Ref. 1) has generated a lot of investigations focused on understanding the microscopic mechanism of this phenomenon. Due to the reduced dimensionality of this bilayer manganite, its electronic and magnetic properties are expected to be different from those for the well-studied infinite-layer manganite. Indeed, some unusual features have been recently reported.[2–4] The reduced dimensionality in fact enhances the CMR effect, albeit at the cost of decreasing the ferromagnetic transition temperature. Further, some features concern the observation of the coexistence of ferromagnetic and antiferromagnetic short-range-ordered spin correlations[3] above $T_c \approx 126\,\mathrm{K}$. Osborn *et al.*[5] have observed canting of the magnetic moments above T_c. We reported[6] inelastic-neutron-scattering investigations of the dispersive low-energy magnetic excitations in $La_{1.2}Sr_{1.8}Mn_2O_7$ propagating perpendicular to the layers and determined the interbilayer exchange interaction. Here we report a complete investigation of acoustic and optic spin-wave modes of in $La_{1.2}Sr_{1.8}Mn_2O_7$ both in the plane and also perpendicular to it. By analyzing these results within an effective exchange model we are able to determine a complete set of intra- and interlayer effective exchange parameters. This leads us to conclude that $La_{1.2}Sr_{1.8}Mn_2O_7$ is a quasi-two-dimensional ferromagnet with weak coupling between different FM bilayers.

The large cylindrical single crystal of $La_{1.2}Sr_{1.8}Mn_2O_7$ of diameter of 5 mm and length 25 mm already studied in Ref. 6 was used for the present study. Inelastic-neutron-scattering experiments were performed on the thermal triple-axis spectrometer IN22 (CRG) installed at the H25 supermirror guide at the Institut Laue-Langevin. The crystal was put inside a helium cryostat with its [010] crystallographic axis vertical so that the scattering plane was $(h0l)$. We used a vertically curved monochromator and a horizontally-curved analyzer and the vertical and horizontal collimations were 15′-30′-60′-80′ and 15′-120′-120′-120′, respectively. The final momentum transfer was kept fixed to the values $k_f = 2.662$ and 4.1 Å^{-1} for low- and high-energy transfers, respectively.

Figure 1 shows Q scans along a^* at $T = 1.6\,\mathrm{K}$ for different constant energy transfers through the reciprocal lattice point $\mathbf{Q} = (1,0,1)$ which is a zone center. In Fig. 1(a) two almost symmetrically situated peaks are observed on both sides of the zone center which we identify as the acoustic branch of the spin waves propagating along [100]. The spin-wave peaks move to higher values for the momentum transfer q for larger energy transfer. In Figs. 1(b) and 1(c) one observes extra intensity at $\mathbf{Q} = (1,0,1)$ contributed by the optic branch due to the finite energy resolution. Figure 2 shows the energy scan at $\mathbf{Q} = (1,0,3.1)$ for $T = 1.6\,\mathrm{K}$ which has a peak at $E \approx 6$ meV. We identify this as the optic spin wave branch as proved below from the Q_l-dependence of the intensity. The energy width of this scan seems to be much larger than the energy resolution of the instrument which is about 1–1.5 meV in the explored energy range. We have fitted the data by the equation

$$S(\mathbf{q},\omega) = \frac{S_0}{1 - e^{-\hbar\omega/kT}} \left[\frac{\Gamma}{\Gamma^2 + \{\hbar\omega - \hbar\omega(q)\}^2} - \frac{\Gamma}{\Gamma^2 + \{\hbar\omega + \hbar\omega(q)\}^2} \right], \tag{1}$$

where S_0 is the normalization constant, Γ is the damping, and $\omega(q)$ is the energy of the excitation. From a least-squares fit we obtain $\Gamma = 4.0 \pm 0.2\,\mathrm{meV}$, $\hbar\omega(q) = 6.27 \pm 0.08\,\mathrm{meV}$. The result of the fit is shown by the continuous curve of Fig. 2. Figure 3 shows the dispersions of both the acoustic and optic spin-wave branches propagating along

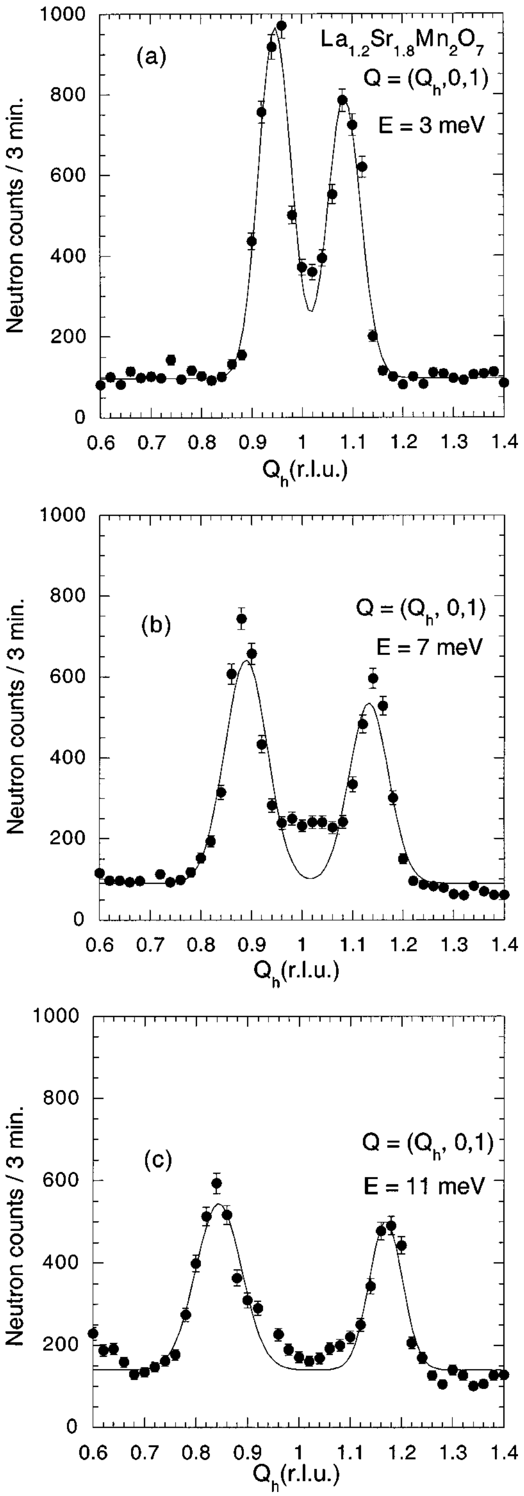

FIG. 1. Q scans along a^* at $T=1.6$ K for different constant energy transfers through the reciprocal lattice point (1,0,1), which is a zone center.

[100]. We have also investigated the dispersion of the optic mode along [001] and found the change in energy to be too small to be measured with a thermal triple axis spectrometer. However, we have reported previously[6] the dispersion of the acoustic mode along [001] as investigated by a cold triple axis spectrometer which was only about 0.4 meV from zone center to zone boundary. The dispersion of the optic branch parallel to [100] is expected to be of the same order.

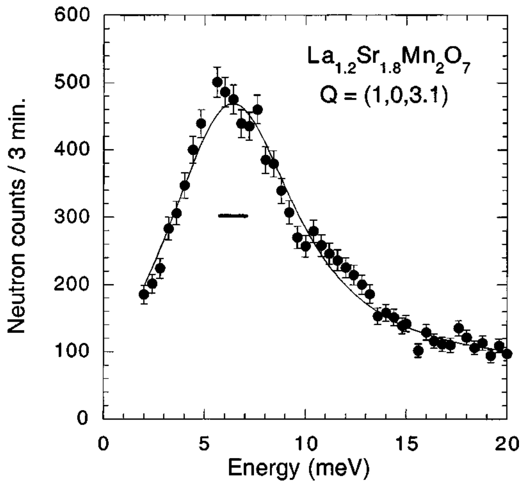

FIG. 2. Energy scan at $\mathbf{Q}=(1,0,3.1)$ at $T=1.6$ K, which shows a peak at $E\approx 6$ meV implying an energy gap of $\Delta\approx 6$ meV of the optic branch at the zone center. The thick line in the middle indicates the energy resolution of the instrument.

In our previous paper[6] we used a simple localized effective exchange model to describe the spin waves of the bilayer manganite. This model was justified by Furukawa[7] on microscopic grounds for the cubic manganites $La_{1-x}A_xMnO_3$ starting from the double exchange Hamiltonian

$$H=-t\sum_{\langle i,j\rangle\sigma}(c_{i\sigma}^{\dagger}c_{j\sigma}+\text{H.c.})-\frac{J_H}{S}\sum_{i}\mathbf{S}_i\cdot\boldsymbol{\sigma}_i\,, \tag{2}$$

which is appropriate for the metallic phase with disordered Mn^{3+}-orbital degrees of freedom. This ferromagnetic (FM) Kondo Hamiltonian describes coupling of itinerant $(c_{i\sigma}^{\dagger})$ e_g electrons of Mn^{3+} which have a hopping energy t and interact with the localized t_{2g} $(S=\frac{3}{2})$ electrons via a Hund's rule coupling of strength $J_H \gg t$. In this limit the model may be mapped to a FM nearest neighbor (NN) Heisenberg model for the localized spin $\mathbf{S}_i$:

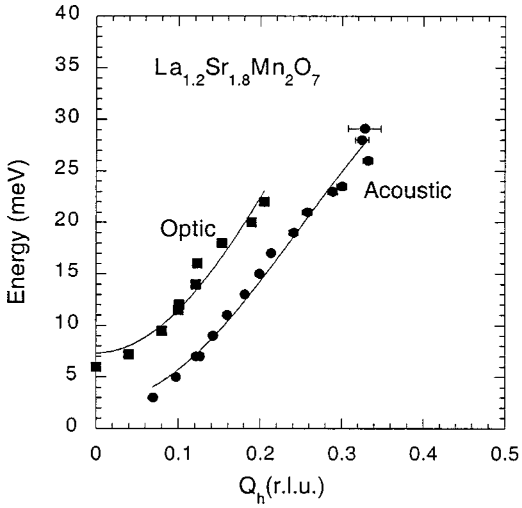

FIG. 3. Spin-wave dispersions of the acoustic and optic branches propagating along [100].

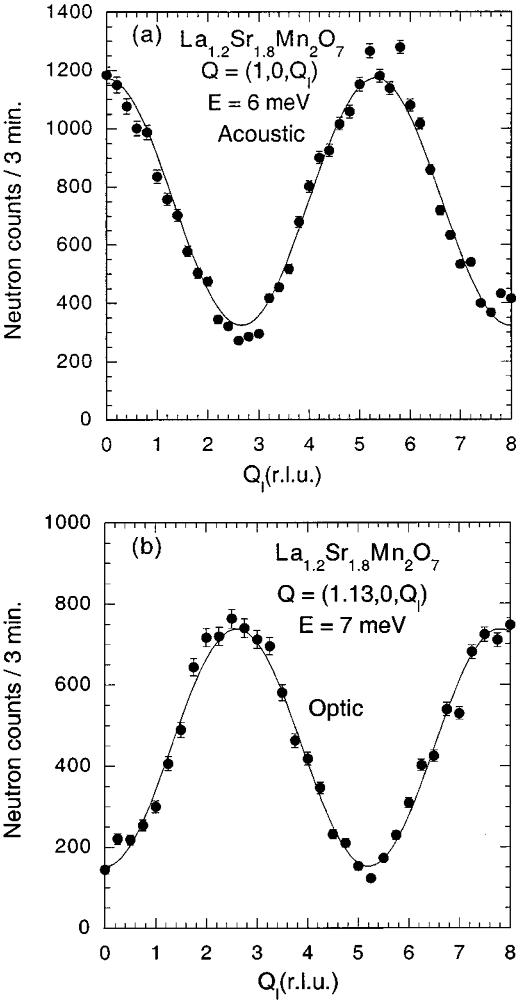

FIG. 4. Q_l variation of the magnon cross section (a) along $\mathbf{Q}=(1,0,Q_l)$ for a constant energy transfer of 6 meV and (b) that along $\mathbf{Q}=(1.13,0,Q_l)$ for a energy transfer of 7 meV.

$$H_{\rm ex}=-J\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_{ij},\qquad J=\frac{t}{S^2N}\sum_{\mathbf{k}} f_{\mathbf{k}\uparrow}\cos k_x. \tag{3}$$

We conjecture that a similar mapping is also possible for the bilayer manganites although this has not yet been shown explicitly. In a more precise treatment, the damping Γ could be related to the hopping energy as in the case of heavy fermion systems. Because of the lower symmetry one now has to invoke three exchange parameters instead of only J: An in-plane exchange $J_a=J_b$, the intrabilayer exchange J_c and the interbilayer exchange J' between adjacent planes of two different bilayers. For this model, general expressions of the acoustical (Ac) and optical (Op) spin wave branches have been derived in Ref. 6. Here we give only the dispersion for $\mathbf{q}=(q_x,0,0)$ and $\mathbf{q}=(0,0,q_z)$ corresponding to our scan directions and assume the limit $J'\ll J_a$, J_c. This is well justified as shown below. In this case we have for the [001] direction[6]

$$\hbar\omega_{\rm Ac}(q_z)=\Delta+4SJ'(1-\cos\tfrac{1}{2}q_z),$$

$$\hbar\omega_{\rm Op}(q_z)=2SJ_c+4SJ'(1+\cos\tfrac{1}{2}q_z), \tag{4}$$

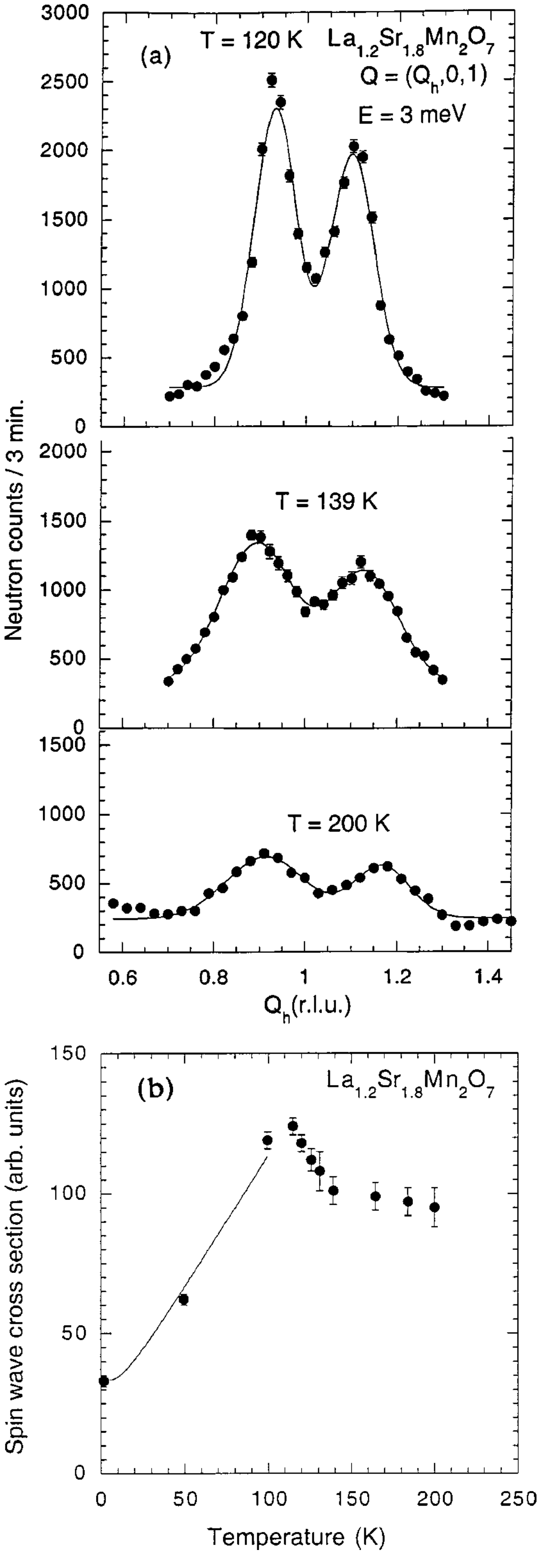

FIG. 5. (a) Q scans along a^* through the reciprocal lattice point (1,0,1) for a constant energy transfer of 3 meV at several temperatures below and above T_c. (b) Temperature variation of the acoustic spin-wave cross section of La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$.

and for the in-plane direction (100)

$$\hbar\omega_{\rm Ac}(q_x)=\Delta+2SJ_a(1-\cos q_x),$$

$$\hbar\omega_{\rm Op}(q_x)=2SJ_c+2SJ_a(1-\cos q_x). \tag{5}$$

Here we added an empirical anisotropy gap Δ for the Ac mode. In our previous low-energy experiments only the (001) acoustical branch $\omega_{\mathrm{Ac}}(q_z)$ was investigated from which we obtained the parameters $\Delta = 0.04$ meV and $J' = 0.026$ meV. However, the other three high-energy branches in the above equations were not found in Ref. 6 and therefore the magnitude of the Ac-Op splitting $2SJ_c$ and the large in-plane dispersion of $4SJ_a$ remained unknown. Our experimental results reported here together with the above Δ and J' allow a complete determination of the exchange model for $La_{1.2}Sr_{1.8}Mn_2O_7$. Fitting our results (Fig. 3) with Eqs. (4) and (5) and using $S = 1.8$ for the effective spin magnitude[6] leads to the exchange constants $J_a = 4.8 \pm 0.1$ meV and $J_c = 1.7 \pm 0.1$ meV. Because $J'/J_c = 1.5 \times 10^{-2}$, $La_{1.2}Sr_{1.8}Mn_2O_7$ can be viewed as an effectively two-dimensional (2D) ferromagnet consisting of weakly coupled FM bilayers. As argued in Ref. 6, the moments lie in the easy *ab* plane. These results of our quantitative analysis are in agreement with both the observation of the in-plane spin-wave-like modes above $T_c = 126$ K shown below and also with the observed rodlike intensity distribution in diffuse scattering.[5,8] It should be noted that J_a and J_c are considerably different $(J_a/J_c = 2.8)$ although NN distances along a and c are almost equal. This suggests that a certain degree of orbital polarization, which leads to this anisotropy, persists even in the metallic phase. Orbitally polarized metallic phases have also been proposed in infinite-layer manganites.[9] In fact, such a state might even be more natural in the bilayer manganites since the degeneracy of e_g orbitals is already lifted by the point symmetry reduction at the Mn sites.

Figure 4 shows the variation of the magnon intensity after correcting for the magnetic form factor as functions of Q_l for $\mathbf{Q} = (1,0,Q_l)$ and $\mathbf{Q} = (1.13,0,Q_l)$ for a constant energy transfer of 6 and 7 meV, respectively. The periodic variation of the intensity reveals the bilayer nature of $La_{1.2}Sr_{1.8}Mn_2O_7$. Remembering that $La_{1.2}Sr_{1.8}Mn_2O_7$ has two Mn atoms in the primitive unit cell, the dynamical susceptibility may be written as[10]

$$\chi(\mathbf{q}+\mathbf{r},\omega) = \tfrac{1}{2}(1+\cos\phi)\chi_{\mathrm{Ac}}(\mathbf{q},\omega) + \tfrac{1}{2}(1-\cos\phi)\chi_{\mathrm{Op}}(\mathbf{q},\omega), \tag{6}$$

where $\phi = 2\pi\mathbf{q}\cdot\boldsymbol{\rho}$ is the phase and $\boldsymbol{\rho} = (0,0,0.19c)$ is the distance vector within a bilayer. The continuous curves in Fig. 4 are obtained by fitting the experimental data with Eq. (6). The excellent agreement clearly proves the quasi-2D nature of interbilayer interactions in $La_{1.2}Sr_{1.8}Mn_2O_7$.

We have also investigated the temperature variation of the acoustic spin-wave intensity by scanning along $\mathbf{Q} = (Q_h,0,1)$ for a constant energy transfer of 3 meV. Figure 5(a) shows such Q scans at several temperatures below and above T_c whereas Fig. 5(b) shows the temperature variation of the integrated intensity of the spin-wave peak. The spin-wave cross section increases with increasing temperature in accordance with the equation

$$S = \frac{S_0}{1-e^{-E/kT}}, \tag{7}$$

where S is the spin-wave cross section at temperature T, S_0 is that at $T=0$, E is the spin-wave energy, and k is the Boltzmann constant. The factor in the denominator is the Bose factor. The spin-wave cross section passes through a maximum at $T_c \approx 120$ K and then decreases slowly and finally tends to become constant at higher temperatures. One clearly observes the spin-wave-like peaks above T_c although they are gradually broadened. This behavior provides additional proof of the quasi-2D ferromagnetic nature of $La_{1.2}Sr_{1.8}Mn_2O_7$. We conclude that the acoustic spin wave propagating along [100] still persists above T_c which is direct evidence for quasi-2D magnetic behavior. This observation contradicts the conclusion of the μSR investigation of Heffner *et al.*,[11] admittedly on a sample with different Sr doping level. However, it is highly unlikely that the quasi-2D nature of $La_{1.2}Sr_{1.8}Mn_2O_7$ would be changed drastically by a slight change in Sr doping, because the 2D magnetic properties originate essentially from the layered crystal structure which does not change abruptly with doping.

In conclusion, we have determined the complete set of exchange interactions of bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$ using inelastic neutron scattering. We have established that $La_{1.2}Sr_{1.8}Mn_2O_7$ is a quasi-2D ferromagnet. We found surprisingly that the in-plane exchange interaction J_a along [100] is about three times bigger than the intrabilayer exchange J_c along [001] although nearest-neighbor distances along [100] and [001] are almost equal. As yet we have no quantitative explanation for this, but the difference in J_a and J_c may be related to a possible orbital polarization persisting in the metallic phase and its influence on the spin exchange.

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