Spin Dynamical Properties of the Layered Perovskite La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$

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Inelastic neutron-scattering measurements were performed on a single crystal of the layered colossal magnetoresistance (CMR) material La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ ($T_C$ ∼ 120 K). We found that the spin wave dispersion is almost perfectly two-dimensional with the in-plane spin stiffness constant $D$ ∼ 151 meVÅ. The value is similar to that of similarly doped La$_{1-x}$Sr$_x$MnO$_3$ though its $T_C$ is three times higher, indicating a large renormalization due to low dimensionality. There exist two branches due to a coupling between layers within a double-layer. The out-of-plane coupling is about 30% of the in-plane coupling though the Mn-O bond lengths are similar.

Keywords: A: magnetic materials, B: crystal growth, C: neutron scattering, D: spin waves

I. INTRODUCTION

The layered perovskite Mn oxide La$_{2-2x}$Sr$_{1+2x}$Mn$_2$O$_7$ (LSMO327), in which MnO$_2$ double layers and (La,Sr)$_2$O$_2$ blocking layers are stacked alternatively, attracts much attention as another class of colossal magnetoresistance (CMR) system. Possibly due to the reduced dimensionality, this system exhibits an extremely large MR at the hole concentration $x = 0.4$. Neutron-scattering study on LSMO327 single crystals ($x = 0.40 - 0.48$) by Hirota et al. has revealed that the low-temperature magnetic structure consists of planar ferromagnetic (FM) and A-type antiferromagnetic (AFM) components, indicating a canted AF structure, where the canting angle between neighboring planes changes from 6.3° at $x = 0.40$ (nearly planar FM) to 180° at $x = 0.48$ (A-type AF). The existence of the canted AFM structure is consistent with previous studies focusing upon the structural properties. Kubota et al. carried out a comprehensive powder neutron-scattering work and established the magnetic phase diagram for $x = 0.30 - 0.50$: there is a planar FM phase between $x = 0.32$ and 0.38, which is smoothly connected to the canted AFM region. To understand the magnetic properties of LSMO327 in more detail, it is necessary to study the excitation spectra, from which one can determine magnetic interaction strengths and spin-spin correlation lengths.

II. THEORETICAL MODEL

Figure shows the magnetic spin arrangement on Mn ions in the tetragonal $I4/mmm$ cell of La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$. Although there is a small canting between neighboring layers within a double-layer at $x = 0.40$, we assume a simple planar ferromagnet, which is sufficient to consider the magnetic interactions between $x = 0.40$. We expect that the dominant spin-spin interactions should occur between nearest-neighbor (NN) Mn atoms, though the in-plane interaction $J_\|$, and the intra-bilayer interaction $J_\perp$ might be different. Although there is no super-exchange coupling between layers belonging to different double-layers, there will be the inter-bilayer interaction $J'$ through a direct exchange. However, it is supposed to be much weaker than $J_\|$ and $J_\perp$, thus we neglect it in our simple model calculation.

FIG. 1. The magnetic spin arrangement on Mn ions in the $I4/mmm$ tetragonal cell of La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$. The lattice parameters are $a = b = 3.87$ and $c = 20.1$ Å at 10 K. The spin Hamiltonian can then be
written in the Heisenberg form as
\[
\mathcal{H} = \frac{1}{2} \sum_i \sum_l \sum_{\delta} J_{il\delta} S_i^l \cdot S_{\delta}^l
\]

and
\[
\mathcal{H} = \frac{1}{2} \sum_i \sum_l \left[ S_i^A \{ J_{il\delta} S_{\delta}^A + J_{\perp} S_{\delta}^A \} + S_i^B \{ J_{il\delta} S_{\delta}^B + J_{\perp} S_{\delta}^B \} \right] + S_i^C \{ J_{il\delta} S_{\delta}^C + J_{\perp} S_{\delta}^C \} ,
\]

where \( i \) denotes a unit cell and \( \delta \) indicates NN sites corresponding to a particular interaction, \( J_{il\delta} \) or \( J_{\perp} \). Following the standard approach, we make the Holstein-Primakoff transformation \( \hat{J} \) to boson creation and annihilation operators \( a_i^\dagger, a_i, b_i^\dagger, b_i, c_i^\dagger, c_i, d_i^\dagger, d_i \), which correspond to \( A, B, C, D \) layers. By Fourier transforming to reciprocal space and performing diagonalization, we obtain the following dispersion relations as the eigenvalues:

\[
\hbar \omega(q) = -2J_{\parallel} S(2 - \cos a q_x - \cos a q_y) - J_{\perp} S \{1 \mp \exp(-i2zcq_z)\}
\]

\[
= -2J_{\parallel} S(2 - \cos a q_x - \cos a q_y) - J_{\perp} S (1 \mp 1) ,
\]

where \( a \) and \( c \) are the lattice constants and \( 2zc \) is the distance between layers within a double-layer. Although there should be four different modes, these are classified to two modes, i.e, acoustic (A) and optical (O), when the inter-bilayer coupling \( J' \) is neglected. Note that both \( J_{\parallel} \) and \( J_{\perp} \) are negative because they are FM interactions.

By using a unitary matrix diagonalizing the Hamiltonian Eq. \( \mathcal{H} \), we obtain the differential scattering cross section for spin waves in LSMO327 with \( x = 0.40 \)

\[
\frac{d^2\sigma}{d\Omega dE_f} = \left( \frac{\gamma e^2}{m c^2} \right)^2 \frac{k_f}{k_i} \left\{ 1 \mp \exp(-i2zcq_z) \right\}^2 (1 + \hat{Q}_x^2) e^{-2W(Q)}
\]

\[
\times \frac{S}{2} \frac{(2\pi)^3}{v_0} \frac{4}{N} \sum_m \sum_{qG} \left( n_{qy}^{(m)} + \frac{1}{2} \pm \frac{1}{2} \right)
\]

\[
\times \delta(\hbar \omega_{qy}^{(m)} + \hbar \omega_q) \delta(Q \mp q - G) \{ 1 \pm \cos(2zc \cdot Q_z) \}
\]

where \( \gamma \) is the gyromagnetic ratio of the neutron, \( f(Q) \) is the magnetic form factor for a Mn ion, \( \exp[-2W(Q)] \) is a Debye-Waller factor, \( \hat{Q}_x = Q_x/Q, n_{qy}^{(m)} \) is the bose factor and \( m \) denotes a mode. Since \( 2zc \) is very close to \( a \approx c/5 \), the A-branch has maximum intensity at \( l = 5m \) \( (m: \text{integer}) \), while the phase of O-branch is shifted by \( \pi \).

**III. EXPERIMENTAL**

La\(_{1.2}\)Sr\(_{1.8}\)Mn\(_2\)O\(_7\) powder was prepared by solid-state reaction using prescribed amounts of pre-dried La\(_2\)O\(_3\) (99.9%), M\(_n\)O\(_4\) (99.9%), and SrCO\(_3\) (99.9%). The powder mixture was calcined in the air for 3 days at 1250°C - 1400°C with frequent grindings. The calcined powder was then pressed into a rod and heated at 1450°C for 24 h. Single crystals were melt-grown in flowing 100% O\(_2\) in a floating zone optical image furnace with a traveling speed of 15 mm/h. To check the sample homogeneity, we powderized a part of single crystal and performed x-ray diffraction, which shows no indication of impurities.

Neutron-scattering measurements were carried out on the triple-axis spectrometer TOPAN located in the JRR-3M reactor of the Japan Atomic Energy Research Institute (JAERI). The (0 0 2) reflection of pyrolytic graphite (PG) was used to monochromate and analyze the neutron beam, together with a PG filter to eliminate higher order contamination. The spectrometer was set up in two conditions in the standard triple-axis mode, typically with the fixed final energy at 13.5 meV and the horizontal collimation of B-100’-S-100’-B. The sample was mounted in an Al can as so to give the \( (h 0 l) \) zone in the tetragonal \( 14/mmm \) notation. We studied the same crystal used in Ref. \( \text{[3]} \) (F-40), which is a single grain with mosaic spread of \( \sim 0.3^\circ \) full width at half maximum (FWHM).

**IV. RESULTS AND DISCUSSIONS**

The spin-wave dispersions along \( [h 0 0] \) were measured at 10 K around \( (1 0 0) \) and \( (1 0 5) \) for the A-branch, and \( (1 0 2.5) \) for the O-branch, as shown in Fig. \( \text{[2]} \). Error bars correspond to the FWHM of peak profiles. Open circle and square indicate the acoustic branch, and solid triangle indicates the optical branch. Solid and dotted curves are obtained by fitting to Eq. \( \text{[6]} \) for \( 0 < q \leq 0.25 \) r.l.u.

**FIG. 2.** The dispersion relations of spin waves at 10 K. Error bars correspond to the FWHM of peak profiles. Open circle and square indicate the acoustic branch, and solid triangle indicates the optical branch. Solid and dotted curves are obtained by fitting to Eq. \( \text{[6]} \) for \( 0 < q \leq 0.25 \) r.l.u.
the data points for $0 < q \leq 0.25$ r.l.u. simultaneously, we obtain $-J_\parallel S = 10.1$ meV and $-J_L S = 3.1$ meV.

To quantitatively examine the present model, we measured the $l$-dependence of the spin-wave intensities of A and O branches at a fixed transfer energy $\Delta E = E_i - E_f = 5$ meV. As shown in Fig. 3, the differential scattering crosssection Eq. 3 is in an excellent agreement with both the A (1 0 0) and O (1 0 0) branches. Note that we do not use any fitting parameters except for intensity scaling.

The results show that spin-spin correlations are significantly anisotropic. The inter-bilayer interaction is as small as we can not detect. The intra-bilayer interaction compared with in-plane interaction, $J_L / J_\parallel$ is about 0.31. We speculate that $x^2 - y^2$ orbital is dominant in the Mn $e_g$ band, which enhances the double-exchange, i.e., ferromagnetic, interactions within a plane. A close relation between the magnetism and the Mn $e_g$ orbital degree of freedom has been also pointed out by recent studies. The in-plane spin wave stiffness constant $D = -J_\parallel S a^2$ is about 151 meVÅ$^2$, which is corresponding to the nearly cubic perovskite La$_{1-x}$Sr$_x$MnO$_3$ ($x = 0.2 - 0.3$) whose $D$ are 188 meV($x = 0.3, T_C = 370$ K) and 120 meV($x = 0.2, T_C = 310$ K) in our material is very reduced, indicating a large renormalization due to low dimensionality.

We noticed that the energy-width of constant-$Q$ scan profile becomes broad at large $q$, particularly $q > 0.25$ r.l.u. In the same high $q$ range, the peak position starts deviating from the dispersion curve obtained from small $q$ data using a conventional Heisenberg model Eq. 4. Similar kind of broadening and deviation are seen in other CMR systems, such as Nd$_{0.7}$Sr$_{0.3}$MnO$_3$, which has a narrower electronic band-width than La$_{0.7}$Sr$_{0.3}$MnO$_3$. Although it is not clear that electron-phonon coupling plays a significant role in such anomalies in LSMO327 as suggested in Nd$_{0.7}$Sr$_{0.3}$MnO$_3$, it would be interesting to study the relation between structural and magnetic properties, particularly in their dynamics.

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La$_{2-2x}$Sr$_{1+2x}$Mn$_2$O$_7$ (x=0.4)