PHYSICAL REVIEW B **66**, 100403(R) (2002)

Spin excitations in La_2CuO_4: Consistent description by inclusion of ring exchange

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(Received 29 April 2002; published 9 September 2002)

We consider the square lattice Heisenberg antiferromagnet with plaquette ring exchange and a finite interlayer coupling leading to a consistent description of the spin-wave excitation spectrum in La_2CuO_4. The values of the in-plane exchange parameters, including ring exchange $J_\Box$, are obtained consistently by an accurate fit to the experimentally observed in-plane spin-wave dispersion, while the out-of-plane exchange interaction is found from the temperature dependence of the sublattice magnetization at low temperatures. The fitted exchange interactions $J=151.9$ meV and $J_\Box=0.24J$ give values for the spin stiffness and the Néel temperature in excellent agreement with the experimental data.

DOI: 10.1103/PhysRevB.66.100403 PACS number(s): 75.10.Jm, 75.40.Gb, 76.60.Es

The magnetic properties of La_2CuO_4 have been the subject of many detailed investigations over the last decade. Understanding this undoped parent compound of high-temperature superconducting cuprates is a precondition for the many theories which describe metallic cuprates by doping carriers into a layered antiferromagnet. The conventional starting point for undoped cuprates is the two-dimensional (2D) spin-1/2 Heisenberg model with only the nearest-neighbor exchange interaction J.[1] This exchange interaction thereby provides the important magnetic energy scale needed as input to theories for the metallic and superconducting properties of doped cuprates. Despite the substantial progress on the theory of the 2D Heisenberg antiferromagnet,[2] which includes such physical properties as the temperature dependence of the magnetic correlation length,[3,4] some of the experimental facts for La_2CuO_4 have clearly demonstrated that a complete description of the magnetic excitations requires additional physics not contained in the 2D Heisenberg model with J only. Examples include the asymmetric line shape of the two-magnon Raman intensity[5] or the infrared optical absorption,[6] which have led to proposals that spin-phonon interactions,[7] resonant phenomena,[8,9] or cyclic ring exchange[11–13] need to be included. In particular, the importance of ring (plaquette) exchange has very recently found direct experimental support from the observed dispersion of the spin waves along the magnetic Brillouin-zone boundary.[14] An alternative theory of excitations in the Heisenberg model, based on a resonating valence bond (RVB) approximation to the ground state, has also been discussed in this connection.[10]

The four-spin plaquette ring-exchange interaction $J_\Box$ was considered rather early as a possible non-negligible correction to the nearest-neighbor Heisenberg model.[15,16] This higher-order spin coupling arises naturally in a strong coupling t/U expansion to fourth order for the single-band Hubbard model at half filling.[17,18] Its quantitative significance was recently demonstrated in high-order perturbation expansions[19] and *ab initio* cluster calculations in realistic three-band Hubbard models for the CuO_2 planes.[20] These derivations of effective spin models for the low-energy magnetic properties of the undoped CuO_2 planes led to the estimate $J_\Box/J=0.11$. A linear spin-wave analysis of the spectrum in La_2CuO_4 at 10 K in Ref. 14 has deduced a considerably larger value $J_\Box/J=0.41$. The necessity for a sizable $J_\Box$ has recently also been conjectured for the spin ladder compound $La_2Ca_8Cu_{24}O_{41}$.[21]

The spin-wave theory and the quasiclassical phase diagram of the frustrated Heisenberg model with ring exchange was investigated in Ref. 22. However, the quantum and thermal renormalizations have not so far been taken into account in the spin-wave theory. It is well known for the quasi-2D Heisenberg model without the ring-exchange term (see Ref. 23 and references therein) that such renormalizations can substantially change the excitation spectrum of the system. The authors of Ref. 14 considered the simplest renormalization of the spectrum by allowing for an overall quantum renormalization factor, which was obtained for the 2D Heisenberg model with nearest-neighbor exchange within a $1/S$ expansion to order $1/S^2$ (Ref. 24) and by series expansion from the Ising limit.[25] However, in the presence of ring exchange or a next-nearest neighbor coupling, it is not possible to provide an accurate determination of exchange parameters without consistent analysis of these terms. Indeed, as we show below, the effects of quantum and thermal fluctuations are not simply captured by a single renormalization factor, and a consistent treatment of the spin-wave spectrum with $J_\Box\neq 0$ to order $1/S$ reveals that the previous early estimate $J=136$ meV from high-energy neutron scattering[26] or two-magnon Raman scattering[27] requires a correction at least as large as 10%. Also, we show that the recent estimate $J_\Box=0.41J$ in Ref. 14 appears to be twice as large as the value calculated by accounting systematically for $1/S$ renormalizations.

In this paper we consider the corrections to the spin-wave spectrum to first order in $1/S$ for finite $J_\Box$ using a self-consistent spin-wave theory.[23] We obtain values of the in-plane and interplane exchange interactions of La_2CuO_4 allowing an accurate fit of the dispersion. We verify that the obtained exchange interactions correctly reproduce the measured values for the spin stiffness and the Néel temperature.

We start from the Heisenberg model with ring exchange,[15,17,18]

$$H=\frac{J}{2}\sum_{i,\delta}\mathbf{S}_i\cdot\mathbf{S}_{i+\delta}+\frac{J'}{2}\sum_{i,\delta'}\mathbf{S}_i\cdot\mathbf{S}_{i+\delta'}+\frac{J''}{2}\sum_{i,\delta''}\mathbf{S}_i\cdot\mathbf{S}_{i+\delta''}+\frac{J_\perp}{2}\sum_{i,\delta_\perp}\mathbf{S}_i\cdot\mathbf{S}_{i+\delta_\perp}+J_\square\sum_{\langle ijkl\rangle}[(\mathbf{S}_i\cdot\mathbf{S}_j)(\mathbf{S}_k\cdot\mathbf{S}_l)+(\mathbf{S}_i\cdot\mathbf{S}_l)(\mathbf{S}_k\cdot\mathbf{S}_j)-(\mathbf{S}_i\cdot\mathbf{S}_k)(\mathbf{S}_j\cdot\mathbf{S}_l)], \tag{1}$$

where J, J', and J'' are the first (δ), second (δ'), and third (δ'') nearest-neighbor in-plane exchanges, $\delta_\perp$ connects to the nearest-neighbor sites in the adjacent planes with interplane exchange $J_\perp$, and $\langle ijkl\rangle$ denotes the four sites of a planar plaquette involved in the four-spin ring exchange.[28] We use the Dyson-Maleev representation for the spin operators,

$$\left.\begin{aligned} S_i^+&=\sqrt{2S}a_i,\; S_i^z=S-a_i^\dagger a_i\\ S_i^-&=\sqrt{2S}\left(a_i^\dagger-\frac{1}{2S}a_i^\dagger a_i^\dagger a_i\right)\end{aligned}\right\}\; i\in A, \tag{2}$$

$$\left.\begin{aligned} S_j^+&=\sqrt{2S}b_j^\dagger,\; S_j^z=-S+b_j^\dagger b_j\\ S_j^-&=\sqrt{2S}\left(b_j-\frac{1}{2S}b_j^\dagger b_j b_j\right)\end{aligned}\right\}\; j\in B, \tag{3}$$

where A and B denote the magnetic sublattices; $a_i^\dagger, a_i$, and $b_j^\dagger, b_j$ are Bose operators. After substituting Eqs. (2) and (3) into the Hamiltonian (1), we decouple quartic terms into quadratic ones according to the procedure described in Ref. 23. Keeping consistently all terms to order $1/S$, we obtain

$$H=S\sum_i\sum_{d=\delta,\delta_\perp}J_d\gamma_d(D_i^\dagger D_i+D_i^\dagger D_{i+d})-S\sum_i\sum_{d=\delta',\delta''}J_d\gamma_d(D_i^\dagger D_i-D_i^\dagger D_{i+d})-J_\square S^3\sum_i\sum_{d=\delta,\delta'}(\gamma_0^\square D_i^\dagger D_i-\gamma_d^\square D_i^\dagger D_{i+d}), \tag{4}$$

where $D_i=a_i$ for $i\in A$ and $D_i=b_i^\dagger$ for $i\in B$; also we use the notation $J_\delta=J$, $J_{\delta'}=J'$, etc. The renormalization of the bare exchange parameters due to quantum and thermal fluctuations is described by the coefficients

$$\gamma_d=\begin{cases}1-[\langle a_i^\dagger a_i\rangle+\langle a_i b_{i+d}\rangle]/S, & d=\delta,\delta_\perp\\ 1-[\langle a_i^\dagger a_i\rangle-\langle a_i^\dagger a_{i+d}\rangle]/S, & d=\delta',\delta'',\end{cases} \tag{5}$$

$$\gamma_0^\square=1-[3\langle a_i^\dagger a_i\rangle+6\langle a_i b_{i+\delta}\rangle+3\langle a_i^\dagger a_{i+\delta'}\rangle]/S,$$

$$\gamma_\delta^\square=1-[3\langle a_i^\dagger a_i\rangle+5\langle a_i b_{i+\delta}\rangle+2\langle a_i^\dagger a_{i+\delta'}\rangle]/S,$$

$$\gamma_{\delta'}^\square=1-[3\langle a_i^\dagger a_i\rangle+4\langle a_i b_{i+\delta}\rangle+\langle a_i^\dagger a_{i+\delta'}\rangle]/S. \tag{6}$$

Diagonalization of this Hamiltonian yields the spin-wave spectrum

$$E_{\mathbf{k}}=\sqrt{A_{\mathbf{k}}^2-B_{\mathbf{k}}^2}, \tag{7}$$

$$A_{\mathbf{k}}=4S[J\gamma-J'\gamma'(1-\nu_{\mathbf{k}}^{\delta'})-J_\square S^2(\gamma_0^\square+\gamma_{\delta'}^\square\nu_{\mathbf{k}}^{\delta'})]-4J''S\gamma''(1-\nu_{\mathbf{k}}^{\delta''})+2J_\perp S\gamma_\perp, \tag{8}$$

$$B_{\mathbf{k}}=4S(J\gamma-J_\square S^2\gamma_\delta^\square)\nu_{\mathbf{k}}^\delta+2J_\perp S\gamma_\perp\nu_{\mathbf{k}}^{\delta_\perp}, \tag{9}$$

where

$$\nu_{\mathbf{k}}^\delta=(\cos k_x+\cos k_y)/2,\quad \nu_{\mathbf{k}}^{\delta'}=\cos k_x\cos k_y,$$

$$\nu_{\mathbf{k}}^{\delta''}=(\cos 2k_x+\cos 2k_y)/2,\quad \nu_{\mathbf{k}}^{\delta_\perp}=\cos k_z, \tag{10}$$

and $\gamma=\gamma_\delta$, $\gamma'=\gamma_{\delta'}$, etc.; the lattice constants are set to unity. Since the equality $\gamma_0^\square=2\gamma_\delta^\square-\gamma_{\delta'}^\square$ is satisfied, the spectrum given by Eq. (7) is necessarily gapless. It is apparent from this result for the dispersion that the renormalization coefficients $\{\gamma_d,\gamma_d^\square\}$ cannot be combined into a single overall renormalization factor. The averages of the bosonic operators which enter in Eqs. (5) and (6) are

$$\langle a_i^\dagger a_i\rangle=\sum_{\mathbf{k}}\frac{A_{\mathbf{k}}}{2E_{\mathbf{k}}}\coth\frac{E_{\mathbf{k}}}{2T}-\frac{1}{2}, \tag{11}$$

$$\langle a_i^\dagger a_{i+d}\rangle=\sum_{\mathbf{k}}\frac{A_{\mathbf{k}}\nu_{\mathbf{k}}^d}{2E_{\mathbf{k}}}\coth\frac{E_{\mathbf{k}}}{2T},$$

$$\langle a_i b_{i+d}\rangle=-\sum_{\mathbf{k}}\frac{B_{\mathbf{k}}\nu_{\mathbf{k}}^d}{2E_{\mathbf{k}}}\coth\frac{E_{\mathbf{k}}}{2T}.$$

The expression for the sublattice magnetization reads

$$\overline{S}=S-\langle a_i^\dagger a_i\rangle. \tag{12}$$

As previously discussed for quasi-2D magnets,[23] Eqs. (5), (6), and (11) must be solved self-consistently. The spin-wave velocity c is obtained by expanding the dispersion at small wave vector $k=\sqrt{k_x^2+k_y^2}$, leading to

$$E_{\mathbf{k}}\simeq ck,\quad c=2\sqrt{2}Z_cJS, \tag{13}$$

where

$$Z_c=(A_{\mathbf{0}}/4JS)^{1/2}[\gamma-2(J'/J)\gamma'-4(J''/J)\gamma''+2(J_\square/J)S^2(\gamma_{\delta'}^\square-\gamma_\delta^\square)]^{1/2} \tag{14}$$

is the spin-wave velocity renormalization factor. For the spin stiffness we obtain the result

$$\rho_s=JS^2Z_\rho, \tag{15}$$

$$Z_\rho=(4J\overline{S}_0/A_{\mathbf{0}})Z_c^2. \tag{16}$$

Given c and ρ_s, the transverse susceptibility follows immediately as

$$\chi_\perp=\rho_s/c^2=\overline{S}_0/(2SA_{\mathbf{0}})=Z_\chi/(8J), \tag{17}$$

where $Z_\chi=Z_\rho/Z_c^2$. For $J'=J''=J_\square=J_\perp=0$, i.e., the 2D Heisenberg model with only nearest-neighbor exchange, the self-consistent numerical solution of Eqs. (5) and (11) at zero

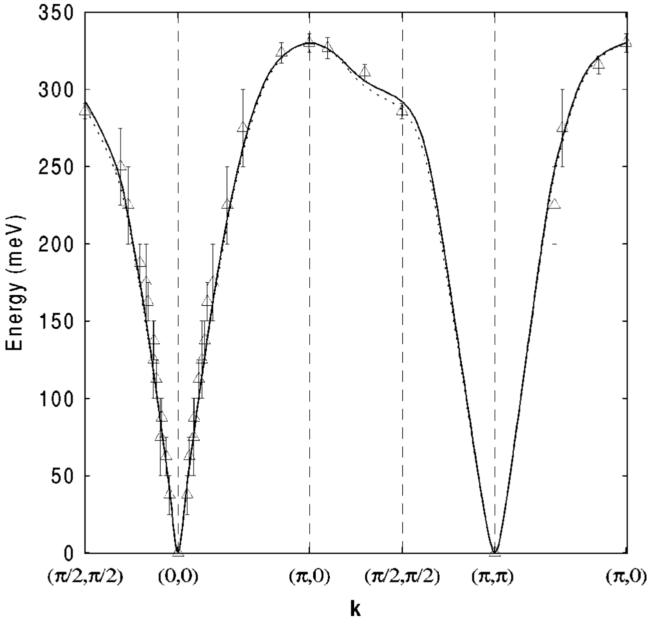

FIG. 1. Spin-wave dispersion along high-symmetry directions in the 2D Brillouin zone. The triangles are the experimental results of Ref. 14 for La_2CuO_4 at 10 K. The solid line is the result of a fit to the spin-wave dispersion result (7) leading to the exchange couplings as listed in Eq. (18). The dashed line is the fit of Ref. 14.

temperature gives[29,30,23] $\gamma = 1.158$ and $\overline{S}_0 = 0.303$, which corresponds to $Z_c = 1.158$, $Z_\rho = 0.702$, and $Z_\chi = 0.524$. We note that due to the relations (16) and (17), Z_ρ and Z_χ contain partially contributions of order $1/S^2$. The above numbers are close to those found for the 2D nearest-neighbor Heisenberg model in a systematic $1/S$ expansion to order $1/S^2$: $Z_c = 1.179$, $Z_\rho = 0.724$, and $Z_\chi = 0.514$.[24] However, we show below, the presence of next-nearest neighbor and plaquette ring-exchange terms substantially alters these numbers.

We use Eqs. (5), (6), and (11) at zero temperature to fit the experimentally determined planar spin-wave dispersion using the data at $T = 10$ K from Ref. 14. To restrict the number of fitting parameters, we suppose $J' = J''$. This restriction is well justified by the perturbation expansions of the half filled one- and three-band Hubbard models.[17–19] The inelastic neutron scattering data together with our fit result along a selected path in the Brillouin zone at $T = 10$ K are shown in Fig. 1. The best fit is obtained for the following parameter set:

$$J = 151.9 \text{ meV}, \quad J' = J'' = 0.025J, \quad J_\square = 0.24J. \tag{18}$$

While the values of J' and J'' are practically indistinguishable from those in Ref. 14 and J in Eq. (18) is only 3% larger, our extracted value of $J_\square$ is 50% lower.

For the corresponding ground state sublattice magnetization $\overline{S}_0$ and for the renormalization parameters $\{\gamma_d, \gamma_d^\square\}$ we obtain

$$\overline{S}_0 = 0.319, \quad \gamma = 1.158, \quad \gamma' = 0.909, \quad \gamma'' = 0.852, \tag{19}$$

$$\gamma_0^\square = 2.220, \quad \gamma_\delta^\square = 1.971, \quad \gamma_{\delta'}^\square = 1.721.$$

In our notation, the spectrum used in Ref. 14 corresponds to equal renormalization factors: $\gamma = \gamma' = \gamma'' = \gamma_i^\square \simeq 1.18$; the parameter values obtained with this spectrum were $J'/J = J''/J = 0.020$, and $J_\square/J = 0.41$. However, some of the γ coefficients show a remarkable deviation from the value 1.18 found for the 2D system with nearest-neighbor exchange only.[24] In particular, the renormalization coefficients $\{\gamma_d^\square\}$ for the ring exchange deviate very strongly. We emphasize again, although some fitting parameters are very similar to those in Ref. 14, it is the self-consitently renormalized parameters $\{\gamma_d, \gamma_d^\square\}$ which allow us to obtain an accurate and reliable set of *bare* superexchange couplings.

In the self-consistent spin-wave theory presented here the in-plane magnon spectrum varies only weakly with temperature at $T \ll J$. Although the spectrum changes qualitatively in the same way as found experimentally, it accounts only for a few percent of the observed changes in the zone boundary dispersion of the data at $T = 295$ K in Ref. 14.

From the parameter values obtained above we deduce the spin stiffness, the spin-wave velocity, and the transverse susceptibility: $\rho_s = 23.8$ meV, $c = 206$ meV, and $\chi_\perp = 4.8 \times 10^{-5}$ K^{-1}. The corresponding values of the renormalization factors as calculated from Eqs. (14), (16), and (17) are $Z_c = 0.96$, $Z_\rho = 0.63$, and $Z_\chi = 0.68$. These values differ substantially from those for the 2D nearest-neighbor Heisenberg model. We note that our value for the spin stiffness is in very good agreement with the earlier estimate[31] $\rho_s = 23.9$ meV found from fitting the spin-spin correlation length $\xi(T)$ at $T > T_N$ to the nonlinear σ model result[2] with the correct preexponential factor.[3] This agreement strongly supports the validity of the self-consistent renormalized spin-wave theory.

As discussed in Refs. 23,32, it is difficult to fit the value $J_\perp$ from measurements of the out-of-plane spin-wave spectrum. Instead, we use an alternative procedure and fit the temperature dependence of the sublattice magnetization at temperatures $T < T_N/2$ where the above theory is reliable (cf. Ref. 23 and references therein) employing the exchange parameters listed in Eq. (18). In this way we obtain $J_\perp/J = 1.0 \times 10^{-3}$ and $\gamma_\perp/\gamma = 5.67 \times 10^{-4}$ which practically coincides with the previous estimate in Refs. 23,32.

As another test of the above results, we also calculate the Néel temperature for the obtained exchange parameter values. On the basis of a renormalization-group approach and a $1/N$ expansion in the $O(N)$ quantum nonlinear σ model, the result for the Néel temperature of a quasi-2D isotropic Heisenberg antiferromagnet has the form[23,32]

$$T_N = 4\pi\rho_s \left[\ln\frac{T_N^2}{c^2\alpha_r} + 3\ln\frac{4\pi\rho_s}{T_N} - 0.0660 \right]^{-1}, \tag{20}$$

where $\alpha_r = (\gamma_\perp/\gamma)_{T=0}$, and c and ρ_s are the respective ground-state spin-wave velocity and spin stiffness given by Eqs. (15) and (13). With the above parameter values (18) we obtain $T_N = 328$ K, in almost perfect agreement with the experimental value $T_N = 325$ K.[31]

In conclusion, we have considered the renormalization of the spin-wave spectrum to order $1/S$ for the Heisenberg antiferromagnet in the presence of plaquette ring exchange. The results allow for an accurate fit of the magnon dispersion in La_2CuO_4 and a consistent determination of the exchange coupling parameters for this material. As an independent check of the parameter set, the spin stiffness and the Néel temperature are correctly reproduced. With $J=151.9$ meV the obtained value for the bare ring-exchange coupling $J_\square/J=0.24$ in La_2CuO_4 is significant although due to four-spin order of this term its effect in the Hamiltonian will be reduced by a factor S^2 with respect to the J term. The magnitude of $J_\square$ suggests that for hole doped cuprates ring exchange might be relevant too, and we may postulate that it is connected to recent proposals of staggered circulating currents in underdoped materials.[33,34] The role of ring exchange for the spectral line shape of the B_{1g} shift in Raman experiments and for infrared absorption remains to be reexplored.

It is a pleasure to thank B. Normand and T. Kopp for useful discussions. We are indebted to G. Aeppli for sending us experimental data for the spin-wave dispersion. We acknowledge support through Sonderforschungsbereich 484 of the Deutsche Forschungsgemeinschaft.

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