Two-dimensional zone-center spin-wave excitations in La_2CuO_4

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We have studied the spin-wave excitations near the two-dimensional zone center at 80 K in a single crystal of La_2CuO_4 which orders at $T_N = 195$ K. In-plane and out-of-plane modes are observed at energies of 1.0 ± 0.25 meV and 2.5 ± 0.5 meV, respectively, The in-plane mode energy is determined primarily by the antisymmetric exchange and it measures this term directly. The out-of-plane mode energy corresponds to a planar anisotropy energy of ~ 0.016 meV. We observe, in addition, the appearance of significant $E \simeq 0$ scattering below 30 K.

I. INTRODUCTION

The magnetic behavior of La_2CuO_4 is of interest for two distinct reasons. First, recent experiments[1] suggest that stoichiometric La_2CuO_4 is a very good approximation to the two-dimensional (2D) $S = \frac{1}{2}$ Heisenberg antiferromagnet, a system of fundamental importance in statistical physics.[2] Second, an understanding of the magnetism in pure La_2CuO_4 should lead to insights into the mechanism for the high-temperature superconductivity in the doped material.[3] La_2CuO_4 has a lamellar crystal structure closely related to the well-studied 2D magnetic compounds[4] K_2NiF_4 and Rb_2MnF_4. Recent neutron scattering experiments on La_2CuO_4 have indeed revealed the anticipated 2D character of the magnetism.[1,5] Above the Néel temperature (T_N), the spins are correlated antiferromagnetically within the CuO_2 planes over distances greater than 200 Å; however, the scattering is entirely dynamical in character with no significant low-energy quasielastic component. The correlations decrease only slowly with increasing temperature, reaching ~ 50 Å at $T = 500$ K.

Below $T \simeq 500$ K the crystal structure is orthorhombic, space group *Cmca*;[6] at 10 K the unit-cell dimensions in the crystal studied here are $a = 5.34$ Å, $b = 13.10$ Å, and $c = 5.42$ Å. Below T_N (=195 K in this crystal) the Cu^{2+} spins align in the *b*-*c* plane canted $\sim 0.17°$ away from the *c* axis; they form a simple nearest-neighbor antiferromagnetic array within the rectangular CuO_2 planes.[7-9] As we shall discuss below, this symmetry leads to two antiferromagnetic spin-wave branches with the lower (upper) mode at $q = 0$ corresponding to motion of the spins predominantly within (out of) the plane.

In this paper we report neutron scattering measurements of the spin dynamics near the two-dimensional zone center in the three-dimensionally ordered phase. The experimental measurements and analysis are given in Sec. II while Sec. III contains the discussion and conclusions.

II. EXPERIMENTAL RESULTS AND ANALYSIS

The experiments were carried out at the Brookhaven High Flux Beam Reactor using the H4M triple axis spectrometer. In the measurements, the final neutron energy was held fixed at 14.7 meV; all collimators were 40′. The consequent energy resolution was 1.0 meV while the longitudinal and out-of-plane Q resolutions were 0.02 and 0.11 $Å^{-1}$, respectively; all widths are full widths at half maxima. The La_2CuO_4 crystal, which was the same one studied in Ref. 1, was mounted in a displex cryostat with an $\mathbf{a}^*$($\mathbf{c}^*$) axis vertical. The crystal structure and the scattering geometry relevant to this experiment are shown in Fig. 1. Because of twinning, the $\mathbf{a}^*$-$\mathbf{b}^*$ and $\mathbf{c}^*$-$\mathbf{b}^*$ reciprocal lattice planes are superimposed in the experiment. The experiment involved fixing the in-plane wave vector at $\frac{1}{2}|\mathbf{a}^*|+|\mathbf{c}^*|$ and varying ($\mathbf{Q}_\perp$) the component along $\mathbf{b}^*$. If ϕ is the angle between $\mathbf{Q}$ and $\mathbf{a}^*$ then the spin-wave cross section is proportional to

$$I(\mathbf{Q}) = f^2(\mathbf{Q})[\tfrac{1}{2}S^{aa}(1+\sin^2\phi)+S^{bb}\cos^2\phi] , \quad (1)$$

where S^{aa} and S^{bb} are the appropriate components of the Van Hove scattering function; they contain the in-plane and out-of-plane modes, respectively. $f(\mathbf{Q})$ is the form factor[8] and $S_i^a = \mathbf{S}_i \cdot \mathbf{a}$, etc.

The spin-wave dispersion relations for the La_2CuO_4 structure may be deduced from problems already treated in the literature.[10] Assuming nearest-neighbor interactions alone the spin Hamiltonian may be written

$$H = \sum_{\langle NN \rangle} \mathbf{S}_i \cdot \bar{\bar{J}}_{NN} \cdot \mathbf{S}_{i+\delta} , \quad (2)$$

where

$$\bar{\bar{J}}_{NN} = \begin{pmatrix} J^{aa} & 0 & 0 \\ 0 & J^{bb} & J^{bc} \\ 0 & -J^{bc} & J^{cc} \end{pmatrix} .$$

Because of the antisymmetric term, the spins are canted

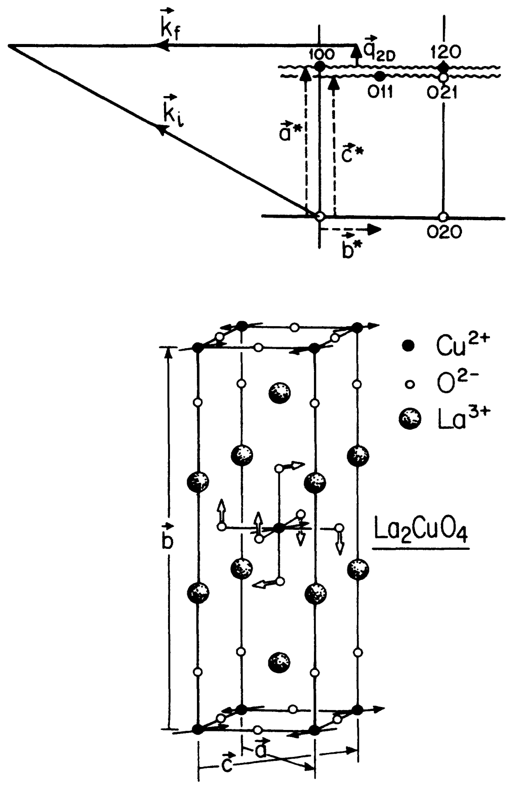

FIG. 1. Top: superposition of **a***-**b*** and **c***-**b*** reciprocal lattice planes of La_2CuO_4 together with a representative scattering diagram for $E=14.7$-meV neutron and $\mathbf{Q}_\perp = 0.57\mathbf{b}^*$; for general $\mathbf{Q}_\perp$, $\mathbf{k}_f$ will not be parallel to $-\mathbf{b}^*$. Bottom: crystal and magnetic structure of La_2CuO_4; the arrows associated with the center oxygens indicate the direction of rotation of the CuO_6 octahedra in the orthorhombic phase.

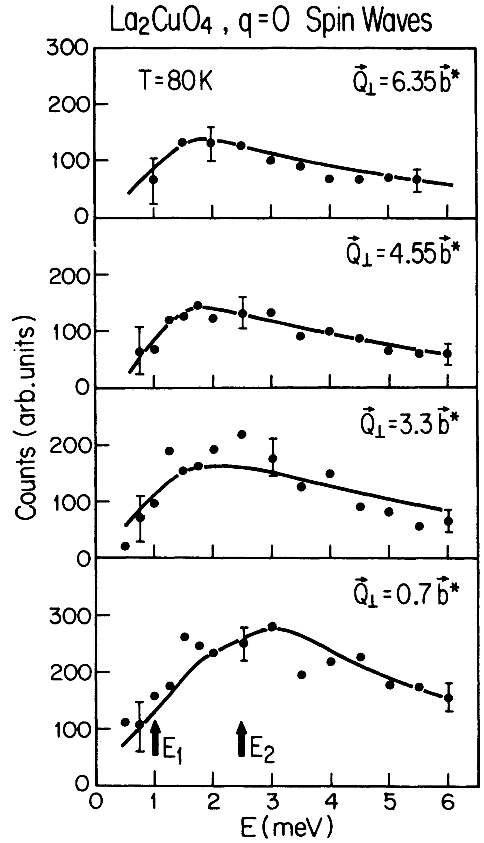

FIG. 2. Inelastic scans for the in-plane wave vector fixed at $\frac{1}{2}(\mathbf{a}^*+\mathbf{c}^*)$ and the between-plane wave vector varied from $\mathbf{Q}^*=0.7\mathbf{b}^*$ to $6.35\mathbf{b}^*$. The background has been subtracted and the intensities have been corrected for $\lambda/2$ contamination in the monitor. A typical counting time at each point was 10 min. The solid lines are the results of the spin-wave theory convoluted with the instrumental resolution function as discussed in the text.

by an angle θ in the **b** direction away from **c**; θ is given by $\theta = J^{bc}/2J_{NN}$ where $J_{NN} = \frac{1}{3}(J^{aa}+J^{bb}+J^{cc})$. Here $|J^{cc}| > |J^{aa}| > |J^{bb}|$. As discussed in Ref. 10, for such canted structures the zone-center spin-wave energies are

$$E_a(0) \simeq g\mu_B\sqrt{H_M^2+2H_EH_A^a}\ ,$$
$$E_b(0) \simeq g\mu_B\sqrt{2H_EH_A^b}\ , \tag{3}$$

where

$$H_M = -4J^{bc}S/g\mu_B\ ,\quad H_E = -4J_{NN}S/g\mu_B\ ,$$
$$H_A^a = 4(J^{aa}-J^{cc})S/g\mu_B\ ,\quad H_A^b = 4(J^{bb}-J^{cc})/g\mu_B\ .$$

As discussed in Ref. 9, one expects $H_M \gg H_A^b \gg H_A^a$ based on Moriya's theory for symmetric and antisymmetric anisotropic exchange.[11] The relative value of H_M and $(2H_EH_A^b)^{1/2}$ is, however, not obvious.

As noted above, experiments were carried out for a series of different $\mathbf{Q}_\perp = Q_\perp\mathbf{b}^*$ values. Background was measured at a number of Q values displaced from the 2D zone center. Away from $E \simeq 0$ there was a Q- and energy-independent background of 35 counts/12 min. The data, corrected for $\lambda/2$ contamination in the monitor and with background subtracted, are shown in Fig. 2. A well-defined peak just above 2 meV is observed for $\mathbf{Q}_\perp = 0.7\mathbf{b}^*$. With increasing $\mathbf{Q}_\perp$ the peak diminishes in intensity and decreases in energy towards ~ 1.5 meV. Far-infrared measurements by Collins, Schlesinger, Schafer, and McGuire[12] indicate a mode at 1.1 meV. From neutron scans at $(\mathbf{a}^*, 0.45\mathbf{b}^*, 0)$ with $E_f = 5.2$ meV we find a mode at ~ 1.0 meV in agreement with Collins *et al.*;[12] we also observe the higher energy mode at ~ 2.5 meV.

To analyze the data shown in Fig. 2 we fix $E_a(0) = 1.0$ meV and $E_b(0) = 2.5$ meV. We then calculate the spin-wave energies at general $\mathbf{q} = \mathbf{Q} - \mathbf{G}$ where **G** is a magnetic reciprocal lattice vector, using the full spin-wave dispersion relation assuming nearest-neighbor interactions only.[10] This 2D approximation is justified by the measurements and analysis in Ref. 9 where it is shown that the net between-plane exchange is of order 0.003 meV, far below our resolution. This is consistent with our direct measurements which show no measurable b-axis dispersion. Calculations have been carried out for J_{NN} varying between 75 and 150 meV, the range of values allowed by the current literature.[5,13] The spin-wave response for each branch is then taken as a Lorentzian $\Gamma/\{\Gamma^2+[E-E_{a,b}(\mathbf{q})]^2\}$ multiplied by the Boson factor $(e^{E/k_BT}-1)^{-1}$ and $1/E$ structure factor; as noted above, the 1 meV mode is assumed to derive from S^{aa} and the 2.5 meV mode from S^{bb} in Eq. (1). This spin-wave cross section is then convoluted with the measured instrumental resolution function to yield the predicted experimental profile. For computational convenience the Lorentzian width Γ was fixed at 0.5 meV, well below the resolution of 1.0 meV. Calculations using a Gaussian line shape for the spin waves give essentially identical results. From Ref. 8 it is known that the form factor $f(\mathbf{Q})$ is approximately constant along the rod. Thus for given J_{NN} the only adjustable parameter in the calculation is an overall intensity factor. We have chosen this to give the optimal average fit to the peak intensities. The solid lines in Fig. 2 are the results of this spin-wave model calculation for $J_{NN}=75$ meV. The long tails extending to high energies are due to the broad vertical resolution combined with the very steep excitation dispersion. We found the shape of the calculated curves to be insensitive to an increase of J_{NN} to values as high as 150 meV; that is, the curves in Fig. 2 are dominated by resolution effects. It is evident that the agreement between theory and experiment is excellent. Thus the spin dynamics near the zone center in La_2CuO_4 in the 3D ordered state are well described by simple two-dimensional spin-wave theory. The zone-center energy gaps are 1.0 ± 0.25 meV and 2.5 ± 0.5 meV for modes in and out of the CuO_2 planes, respectively.

The temperature dependence of the zone-center response for $\mathbf{Q}_\perp=0.7\mathbf{b}^*$ is shown in Fig. 3. Above $T_N=195$ K the modes are overdamped as expected. At 80 K the response is as shown and analyzed in Fig. 2. The most notable new feature is that below ~30 K intense scattering appears near $E=0$. This is shown in more detail in Figs. 4(a) and 4(b) which give, respectively, constant $E=0$ scans at various temperatures and the $E=0$ peak intensity along the rod (0.99,0.7,0) and away from $\mathbf{q}=0$ at (0.9,0.7,0); the latter is presumed to represent the background. This extra scattering led the authors of Ref. 1 to conclude incorrectly that at 5 K the gap was less than 1 meV. Similar, but more dramatic effects have been observed by Endoh *et al.*[5] in a sample with $T_N\simeq100$ K. Aharony *et al.*[14] have suggested that this is indicative of reentrant spin-glass behavior. This is an enticing explanation although further work will be required to establish their explanation definitively.

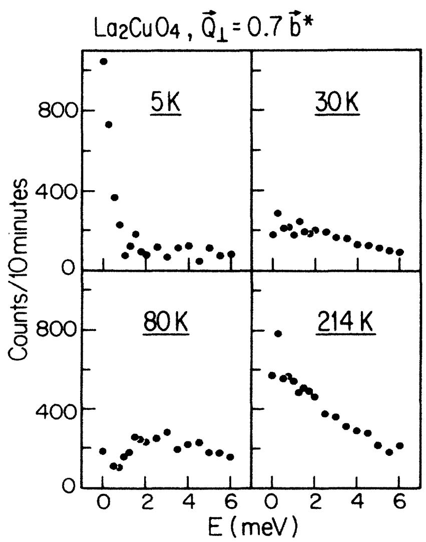

FIG. 3. Inelastic scans at $[\frac{1}{2}(\mathbf{a}^*+\mathbf{c}^*),0.7\mathbf{b}^*]$ at a series of temperatures. The background has been subtracted.

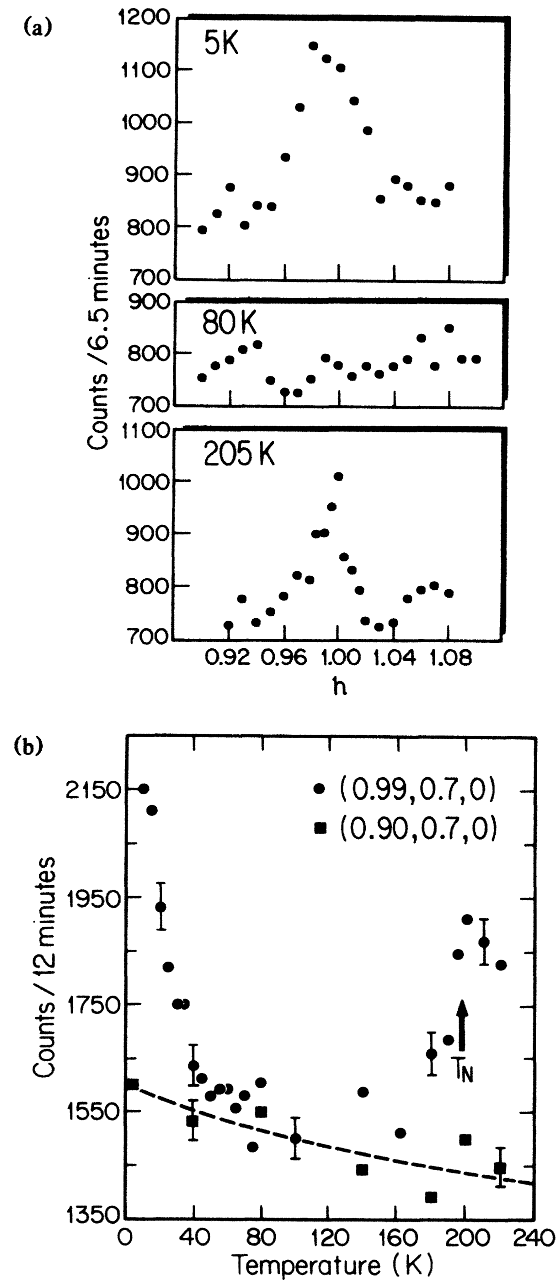

FIG. 4. (a). Constant $E=0$ scans along $(h,0.7,0)$ at various temperatures. Note the absence of an $E=0$ feature at 80 K. (b) $E=0$ peak intensity vs temperature of a position along the rod (0.99, 0.7, 0) and away from the rod (0.9, 0.7, 0). The dashed line is a guide to the eye, through the (0.9, 0.7, 0) points which represents the presumed background.

III. CONCLUSIONS

As discussed above, the salient result of this work is that simple spin-wave theory describes the low-energy response in La_2CuO_4 very well. We observe in-plane and out-of-plane zone-center spin-wave modes at energies of 1.0 ± 0.25 meV and 2.5 ± 0.5 meV, respectively. The in-plane zone-center spin-wave mode energy is almost certainly dominated by the antisymmetric term $g\mu_B H_M$, so to leading order [using the more precise far-infrared value for $E_a(0)$] we have $J^{bc}=0.55\pm0.06$ meV. Thio *et al.*[9] from their measured canted moment, deduce a canting angle $\theta=H_M/2H_E=0.0029\pm0.0006$ radians, thence giving $H_E=190\pm50$ meV. This translates into a spin velocity $V_{sw}=(1.18)H_E(a/2)=0.60\pm0.15$ eV Å in good agreement with the Lyons *et al.*[13] estimate of 0.7 eV Å. From Eq. (3) and $E_b(0)=2.5\pm0.5$ meV we deduce for the out-of-plane anisotropy field $g\mu_B H_A^b=0.016$ meV. From the Néel temperature and the 2D correlation length measurements of Ref. 5 and assuming $kT_N \sim J_\perp(\text{eff})\xi_{2D}^2$ one may estimate an effective between-plane exchange $J_\perp(\text{eff})$ of $\sim$0.003 meV. Thus pure La_2CuO_4 does indeed correspond quite closely to an ideal two-dimensional $S=\frac{1}{2}$ rectangular lattice Heisenberg model. This is a model system which has been long sought after by physicists interested in quantum magnets. It cannot be a coincidence that La_2CuO_4 and related materials, which are realizations of the 2D $S=\frac{1}{2}$ Heisenberg antiferromagnet, when doped with mobile carriers exhibit superconductivity at remarkably high temperatures.[3,15] We hope that these measurements are of value in developing models for the microscopic mechanism responsible for the superconductivity.

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$$E_a(\mathbf{q})^2=g^2\mu_B^2\{H_A^b\cos\theta+H_E\cos2\theta[1+\gamma(\mathbf{q})]+H_0\sin\theta\}\times\{H_A^a+H_M\sin\theta+H_E[1-\gamma(\mathbf{q})]\}\ ,$$

$$E_b(\mathbf{q})^2=g^2\mu_B^2\{H_A^b\cos\theta+H_E[1-\gamma(\mathbf{q})]\cos2\theta+H_0\sin\theta\}\times\{H_A^a+H_M\sin\theta+H_E[1+\gamma(\mathbf{q})]\}\ ,$$

where $\gamma(\mathbf{q})=\cos(\mathbf{q}\cdot\mathbf{a}/2)\cos(\mathbf{q}\cdot\mathbf{c}/2)$, the external field H_0 is along $\mathbf{b}$, and $\theta=(H_0+H_M)/2H_E$.

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