High-Energy Spin Waves in La_2CuO_4

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Time-of-flight spectroscopy using neutrons produced by a spallation source is used to measure the one-magnon scattering throughout the Brillouin zone for La_2CuO_4. The zone-boundary magnons have an energy $\hbar\omega_{ZB}=0.312\pm0.005$ eV and are good eigenstates of the quantum Heisenberg Hamiltonian in that they possess lifetimes $>10/\omega$. A multiplicative renormalization of the overall frequency scale of classical spin-wave theory accounts for the quantum effects in the one-magnon spectrum.

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The unusual features of Heisenberg antiferromagnets when quantum mechanics is taken into account have been investigated for much of this century. For the one-dimensional case, quantum fluctuations destabilize the Néel state entirely. Even in higher dimensions, quantum fluctuations lead to significant corrections to the ground-state energy [1,2] and the spin-wave excitations [3] which are strongest for low spin. The discovery of high-temperature superconductivity has focused attention on La_2CuO_4, apparently an excellent realization of a two-dimensional (2D) spin-$\frac{1}{2}$ quantum antiferromagnet [4]. The magnetic correlations in La_2CuO_4 have been investigated by several microscopic techniques, most notably light scattering [5], muon spin relaxation [6–8], and neutron scattering [9–13]. Unfortunately, because of experimental limitations and restrictions in momentum and energy transfer, no technique has been able to probe the excitations over the whole Brillouin zone. We describe here a neutron-scattering investigation performed on a spallation source, in which we were able to overcome previous difficulties and observe magnetic excitations over a wide range of wave vector and energy. Well-defined zone-boundary spin waves with an energy of $\hbar\omega_{ZB}=0.312\pm0.005$ eV are found. Also, the dispersion relation shows the conventional spin-wave form.

Following previous practice, we use the orthorhombic nomenclature to label reciprocal space so that the basal planes are parallel to the $(h0l)$ zone. Even though $a\neq c$, in this experiment we do not distinguish between them, for convenience, we define $\mathbf{a}^*$ and $\mathbf{c}^*$ to lie in the horizontal and vertical planes of the instrument, respectively. The sample was an assembly of sixteen single crystals [14] aligned such that their in-plane axes coincided to better than 1.5°. The total mass of the crystals was 0.1 kg. As described elsewhere [12], crystals were grown from CuO-rich melts contained in a large Pt crucible. The room-temperature lattice parameters were $a=5.375(2)$ Å, $b=13.156(4)$ Å, and $c=5.409(2)$ Å. The Néel temperatures of the individual crystals ranged between 260 and 290 K.

Experiments were performed on the High-Energy Transfer Spectrometer (HET) [15] at the United Kingdom spallation neutron source ISIS of the Rutherford Appleton Laboratory. HET is a "direct geometry chopper" spectrometer. High-energy (spallation) neutrons are produced when an 800-MeV pulsed proton beam with a current of 100 μA hits a uranium target. A beam of monochromatic neutrons is produced at the sample by phasing of Fermi chopper spinning at a frequency of $\sim$500 Hz and at 10 m from the spallation target to the proton pulse. Incident energies between 30 and 2000 meV may be selected. We specify the direction of the scattered neutron $\mathbf{k}_f$ using polar coordinates $(2\theta,\phi)$ with respect to $\mathbf{k}_i$ such that $\phi=0$ corresponds to the horizontal plane and 2θ represents the magnitude of the angle through which neutrons are scattered. Scattered neutrons are detected in an array of fifty ^{3}He detectors located in the plane P perpendicular to $\mathbf{k}_i$ and 4 m from the sample. The detectors are 25-mm-diam tubes in a pentagonal arrangement with their long axes tangential to circles of constant 2θ in the plane P and covering a range in 2θ from 3° to 7°. The sample was oriented with its $\mathbf{c}^*$ axis vertical. During the experiment the sample was rotated about this axis. We specify the orientation of the sample by α, the angle between the $\mathbf{b}^*$ axis and $\mathbf{k}_i$ measured in the same sense as 2θ (for $\phi=0$). The energy transfer $\hbar\omega=E_i-E_f$ and the scattering vector $\mathbf{Q}=\mathbf{k}_i-\mathbf{k}_f$ are determined from the time of flight of the detected neutrons.

To avoid background from cryogenic shields, we collected data at ambient temperature ($T=296$ K), i.e., in the paramagnetic phase. In its paramagnetic phase, La_2CuO_4 exhibits strong 2D correlations which were first observed using "energy-integrated" neutron scattering [9]. At $T=296$ K, the antiferromagnetic correlation length is $\xi\approx200$ Å [9,12]. For $q>\xi^{-1}$ we probe the (spin-wave) excitations within these regions. We first established the presence of the 2D magnetic scattering pre-

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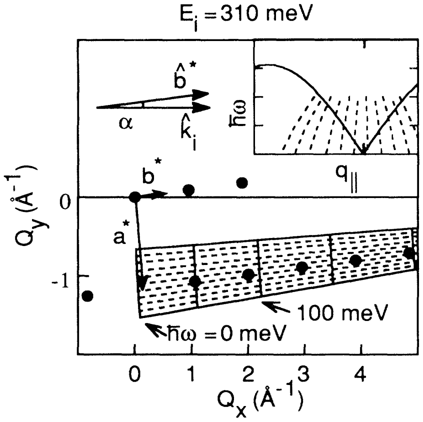

FIG. 1. The geometry of the measurements used to obtain Fig. 2(a). Q_x and Q_y are the parallel and perpendicular components of **Q** with respect to $\mathbf{k}_i$. The solid circles are nuclear Bragg points. Dashed lines in the overlaid net denote boundaries in the reciprocal space sampled by the individual detectors; energy transfers $\hbar\omega$ are represented by the vertical arcs. Inset: The $q_{\parallel}$-$\hbar\omega$ projection of phase space with the dashed lines marking the boundaries of the regions sampled by individual detectors.

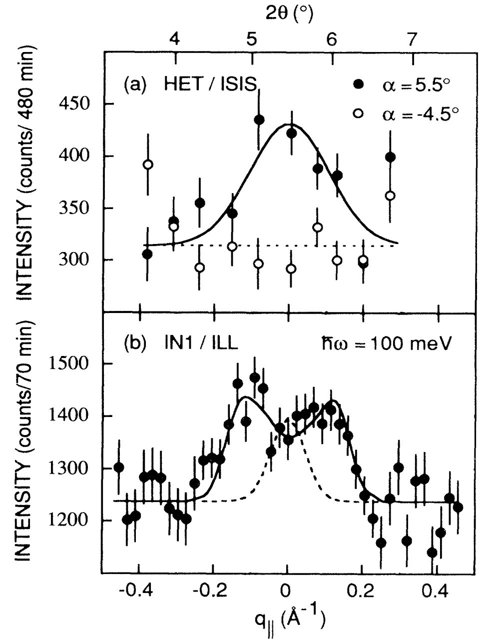

FIG. 2. (a) Measurements (spallation source) of the 2D magnetic scattering in La_2CuO_4 for $E_i=307$ meV and $\hbar\omega=100$ meV. The energy resolution is 50 meV (FWHM) and the sample mass is 100 g. Solid points ($\alpha=5.5°$) correspond to the condition that gives rise to a peak in the magnetic scattering for $2\theta=5.5°$. Open circles are with the crystal rotated ($\alpha=-4.5°$). (b) A transverse scan (reactor source) collected using the triple-axis spectrometer IN1 [11]; the dashed line shows the instrumental resolution. The energy resolution was 20 meV (FWHM) and the sample mass is 15 g.

viously observed in this material: Figure 1 illustrates the geometry of the experiment. The incident energy and the orientation of the crystal were arranged such that the detector with center $2\theta=5.5°$ collected neutrons with scattering vector along the line $\mathbf{Q}=(1k0)$ (the center of the two-dimensional magnetic zone). To fulfill this condition we chose $E_i=307$ meV and $\alpha=5.5°$. Figure 2(a) shows a constant $\hbar\omega=0.1$ eV scan constructed by plotting the number of scattering events within a resolution window with a full width at half maximum (FWHM) of 50 meV as a function of detector angle 2θ, or reduced in-plane momentum transfer $q_{\parallel}=\mathbf{Q}\cdot\hat{\mathbf{a}}^*-|a^*|$, for the detectors in the horizontal plane. Because of the large spin-wave velocity and the poor (≈ 0.2 $Å^{-1}$ FWHM$\parallel\mathbf{a}^*$ and 1.0 $Å^{-1}$ FWHM$\parallel\mathbf{c}^*$) resolution of the instrument, we observe a single peak at $q=0$ ($2\theta=5.5°$), i.e., the two-dimensional Bragg point. Rotation of the sample to $\alpha=-4.5°$ causes the peak to disappear because this changes the relation between the scattering angle 2θ and $q_{\parallel}$ and eliminates the constant-q nature of the scattering collected by individual detectors with $2\theta\cong 5.5°$. To permit comparison between reactor and spallation sources, we show in Fig. 2(b) a constant $\hbar\omega=0.1$ eV scan obtained using the IN1 triple-axis instrument at the Institut Laue-Langevin (ILL) for a sample with one-sixth of the volume of that used in the ISIS experiment. The width of the energy integration is 20 meV, the FWHM of the instrumental resolution function. The HET data are collected simultaneously, while the IN1 data are acquired point by point, with a different instrument setting at each point. This means that per point in the interesting range of momentum transfers, the spectra were collected in similar amounts of time. The signal-to-background ratio is approximately 2 times better for HET than for IN1, while the resolution of IN1 in the configuration employed is clearly superior.

We now turn to the investigation of the higher-energy excitations which were not observable using IN1. For this we used a scattering geometry similar to that shown in Fig. 1, but with a larger incident energy, $E_i=681$ meV. Figure 3(a) shows the difference between events measured by four detectors with azimuthal angles corresponding (for $0.2\leq\hbar\omega\leq 0.4$ eV) to in-plane momentum transfers near the magnetic zone boundaries of La_2CuO_4, and four other detectors with the same 2θ's but different azimuthal angles. Neutron-scattering cross sections generally include elastic incoherent, one-phonon, and multiphonon terms in addition to the magnetic contributions. For the present experiment, the first term is restricted to $|\hbar\omega|\leq 20$ meV (the elastic resolution half-width), while one-phonon scattering exists only below the cutoff frequency of $\sim$90 meV [16]. Finally, multiphonon scattering is not expected to vary with azimuthal angle. Thus, the substantial difference plotted in Fig. 3(a) is due to the zone-boundary magnons in La_2CuO_4. This represents the first evidence that well-defined spin waves exist at the

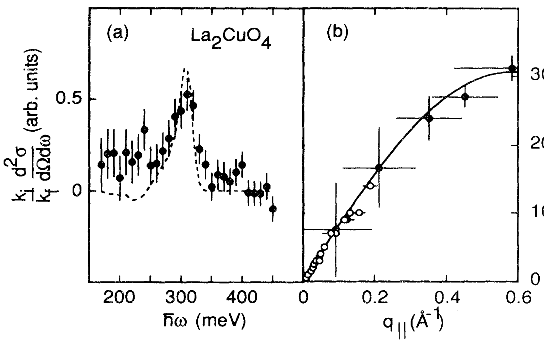

FIG. 3. (a) The difference between a combination of four detectors chosen to emphasize scattering near the magnetic zone boundary, $\mathbf{Q} \approx (1,k,0.5)$, and a background combination with the same 2θ. The dashed line is a resolution-corrected nearest-neighbor spin-wave model described in the text. (b) The dispersion relation deduced from the spectra in (a) and Fig. 4 (closed circles) and previous measurements [11,12] (open circles). Horizontal bars represent the range in wave vector parallel to (1,0,0) over which the detectors integrate. Energies are the fitted values at the centers of the detectors.

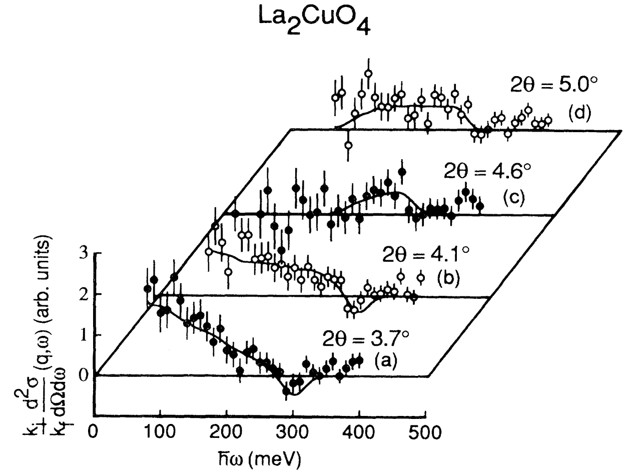

FIG. 4. Spectra from individual detectors in the horizontal plane for $E_i = 685$ meV and $\alpha = 5.5°$ From (a) to (d) the centers of the detectors are $2\theta = 3.7°$, 4.1°, 4.6°, and 5.0° with corresponding elastic reduced in-plane momenta $q_{\parallel} = 0.0$, 0.13, 0.28, and 0.41 Å^{-1}. A measured background has been subtracted (see text). Solid lines are fits by classical spin-wave theory.

zone boundary in La_2CuO_4. Indeed, the peak at $\hbar\omega = 0.312$ eV in Fig. 3(a) has a FWHM of 41 meV which should be compared with a FWHM of 30 meV expected for magnons of infinite lifetime due to the resolution of the instrument. Thus, we conclude that spin-wave lifetime effects in this two-dimensional $S = \frac{1}{2}$ antiferromagnet are small. Specifically, from the measured excess width we obtain a *lower* bound of 9.3×10^{-13} s $\cong 10/\omega_{ZB} \cong \hbar/k_B T$ on the magnon lifetime.

Spectra recorded for individual detectors in the horizontal plane reveal the evolution of the spin-wave scattering through the Brillouin zone. Figure 4 shows a series of such spectra, for $3.5° < 2\theta < 5.2°$. The background has been taken as the scattering found for the detector with center $2\theta = 6.2°$ ($q_{\parallel} = 0.77$ Å^{-1}), where we expect the magnetic scattering to be small but not zero [see Eq. (4) below]. As expected, moving from near the magnetic zone center [spectrum (a)] towards the zone boundary shifts the spectral weight from low to high frequency. The tail extending to high frequencies in spectrum (a) is due to the large detector height. This means that the detector samples scattering along a line parallel to (0,0,1) in reciprocal space of length 1.0 Å$^{-1} \cong c^* \cong a^*$.

Conventional spin-wave theory of the Holstein-Primakoff type [17] for a 2D Heisenberg square lattice antiferromagnet with a Hamiltonian

$$H = \sum_{ij} J\mathbf{S}_i \cdot \mathbf{S}_j , \tag{1}$$

where the sum is over nearest-neighbor spins ($S = \frac{1}{2}$), yields a cross section (for magnon creation)

$$\frac{d^2\sigma}{d\Omega\, d\omega} = \frac{k_f}{k_i} |f(\mathbf{Q})|^2 A(\mathbf{q},\boldsymbol{\tau})[n(\omega)+1]\delta(\omega - \omega_0(\mathbf{q},\boldsymbol{\tau}))\delta(\mathbf{Q} - \boldsymbol{\tau} - \mathbf{q}) . \tag{2}$$

In (2),

$$\hbar\omega_0(\mathbf{q},\boldsymbol{\tau}) = 2J[1 - \cos^2(h\pi)\cos^2(l\pi)]^{1/2} , \tag{3}$$

$$A(\mathbf{q},\boldsymbol{\tau}) \propto \left[\frac{1 - \cos(h\pi)\cos(l\pi)}{1 + \cos(h\pi)\cos(l\pi)} \right]^{1/2} , \tag{4}$$

$n(\omega) = [\exp(\beta\hbar\omega) - 1]^{-1}$ and we have averaged over the spin direction. The lines in Figs. 3(a) and 4 represent resolution-corrected fits of this form to the spectra. Figure 3(b) shows the resulting spin-wave energies $\hbar\omega_0$ together with those obtained from previous reactor-based neutron-scattering experiments [11,12]. From a comparison of the theoretical form and the data, it is clear that the conventional theory with an effective coupling $J_{\rm eff} = 0.153 \pm 0.004$ eV gives an excellent description not only of the dispersion relation but also of the q dependence of the spin-wave amplitude. The theory is successful to the extent that it even accounts for the negative levels in the difference spectra seen near $\hbar\omega = 0.3$ eV in Figs. 4(a) and 4(b), due to the small but finite magnetic scattering present in the subtracted "background."

Neutron-scattering data obtained throughout the Brillouin zone present a unique opportunity to establish the strengths of the interactions $J_2 \mathbf{S}_i \cdot \mathbf{S}_j$ between next-nearest neighbors. Correction of Eqs. (3) and (4) for such interactions does not improve the quality of the fits significantly. From the ratio of the zone-boundary magnon energy to the $T=5$ K zone-center spin-wave velocity, $\hbar c = 0.85 \pm 0.03$ eV [11], we can place an upper bound of 9 meV for $|J_2|$.

It has been known for a long time [1,2] that the Néel state is not the exact ground state of Eq. (1). Furthermore, the energies of the spin-wave excitations show corrections [3] due to interactions between spin waves. The quantum corrections are expressed most appropriately by a multiplicative renormalization of the "classical" values. Thus, the effective coupling in Eq. (3) is actually $J_{\text{eff}} = Z_c J$, where J is the nearest-neighbor coupling in the bare Hamiltonian and Z_c is the renormalization factor. The value of Z_c has been estimated by a number of techniques [18,19]. Using the value $Z_c \approx 1.18$ [18], and assuming $\hbar\omega_{\text{ZB}} = 2Z_c J$, we find an (unrenormalized) coupling constant $J = 0.132 \pm 0.004$ eV. This is in excellent agreement with the value determined from the *low-temperature* (small-q) spin-wave velocity [11,12], $J = 0.136 \pm 0.005$ eV, and two-magnon Raman scattering [5], $J = 0.128 \pm 0.006$ eV. However, we note that the value determined directly from the spin-wave velocity at the temperature of the present experiment [11,12] is $J = 0.120 \pm 0.005$ eV. This result together with the T independence of the Raman cross section [5] for $T < 300$ K suggests that there is a softening of the interactions only at low energies.

In summary, we have performed the first measurements of the one-magnon scattering throughout the Brillouin zone in La_2CuO_4. The classical theory of the square lattice antiferromagnet with nearest-neighbor interactions modified only by a multiplicative renormalization of the frequency scale gives a good description of both the dispersion and the variation of the amplitudes of the spin waves. Furthermore, the zone-boundary magnon scattering is very sharply peaked. We conclude that the classical spin waves correspond very closely to eigenstates of the quantum Hamiltonian for La_2CuO_4.

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