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Authors

Hayden, SM
Aeppli, G
Mook, HA
[et al.](#)

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HIGH ENERGY INELASTIC NEUTRON SCATTERING FROM La_2CuO_4

S.M. HAYDEN^a, G. AEPPLI^b, H.A. MOOK^c, Z. FISK^d, S-W. CHEONG^d, J.P. REMEIK^{b†},
G.P. ESPINOSA^b and A.S. COOPER^b

^a*Institut Laue-Langevin, 38042 Grenoble Cedex, France,*

^b*AT & T Bell Laboratories, Murray Hill, NJ 07974, USA*

^c*Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA*

^d*Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

We have used high energy inelastic neutron scattering to study the magnetic fluctuation spectrum of La_2CuO_4 in the energy and momentum ranges $30 \leq \hbar\omega \leq 140$ meV and $|q| \leq 0.2 \text{ \AA}^{-1}$, respectively. Our results are consistent, within the experimental error, with conventional spin wave theory the low temperature spin wave velocity, being 850 ± 30 meV \AA^{-1} .

The magnetic dynamics of La_2CuO_4 have recently attracted considerable attention. Interest has been motivated by the discovery of superconductivity in the $\text{La}_{2-x}(\text{Ba}, \text{Sr})_x\text{CuO}_{4-y}$ system and suggestions that La_2CuO_4 itself is a model system for the two-dimensional (2D) $S = \frac{1}{2}$ Heisenberg antiferromagnet [1]. We report here an inelastic neutron scattering study of the magnetic dynamics of La_2CuO_4 performed with epithermal neutrons. The use of high energy neutrons allows us to access large energy transfers, thus we probe the high energy magnetic fluctuations. These are inaccessible using thermal neutron scattering [2], which suffers severe resolution problems at the available energy transfers. We describe here a study in the range $30 \leq \hbar\omega \leq 140$ meV, an extension of our previous measurements [3].

Crystals were grown from melts containing excess CuO. The sample used for the inelastic neutron scattering experiment consisted initially of an aligned array of seven La_2CuO_4 crystals with total volume 2 cm^3 , for the scans with the highest transfers eight additional larger crystals were added bringing the total volume up to 8 cm^3 . The crystals were aligned such that their in plane axes coincided to better than 1.5° . Points in reciprocal space are labeled such that the basal planes are perpendicular to $[010]$. The room temperature lattice parameters (determined from X-ray diffraction) were $a = 5.375(2)$, $b = 13.156(4)$ and $c = 5.409(2)$ zone. The Néel temperature for the individual crystals, determined from elastic Bragg scattering, varied from 260 ± 5 to 290 ± 5 K, with saturation of the magnetic order for $T < 200$ K.

Inelastic experiments were performed using the triple-axis spectrometer IN1 on the “hot” neutron source of the Institut Laue-Langevin (ILL), Grenoble, France. In our experiments, incident neutron energies of up to 290 meV were used. A vertically curved $\text{Cu}(200)$ monochromator was used with vertically curved pyrolytic graphite (002) and $\text{Cu}(200)$ as analyzers. Curving the monochromator and analyzer allows a gain in signal at the expense of the vertical resolution of the spectrometer. In our experiment this relaxed resolution was used to integrate along the $[010]$ direction in reciprocal space. Data were collected in two experimental configurations, for lower energy transfers we used a graphite (002) analyzer with $E_i = 80$ meV, at higher energy transfers we used a $\text{Cu}(200)$ analyzer with $E_i = 150$ meV. Higher order contamination (present from higher order reflections of the monochromator) was eliminated using a 0.4 mm thick sheet of Er in front of the sample (for $E_i = 80$ meV) and 0.5 mm of Hf before and 0.4 mm of Er after the sample for $E_i = 150$ meV. Er and Hf are “nuclear filters”, resonances in their neutron absorption cross section allow them to be used as filters.

Transverse constant energy scans through the antiferromagnetic zone center are shown for various energy transfers in fig. 1. For the higher energy scans, two maxima can be observed, corresponding to spin waves propagating in opposite directions. At low energy, the instrumental resolution in wave vector and energy is insufficient to resolve the two branches, all the scattering from magnons of energy $\hbar\omega$ being within the resolution volume, as has also been the case for thermal neutron scattering studies of La_2CuO_4 [2]. We have analyzed our data in terms of a conventional

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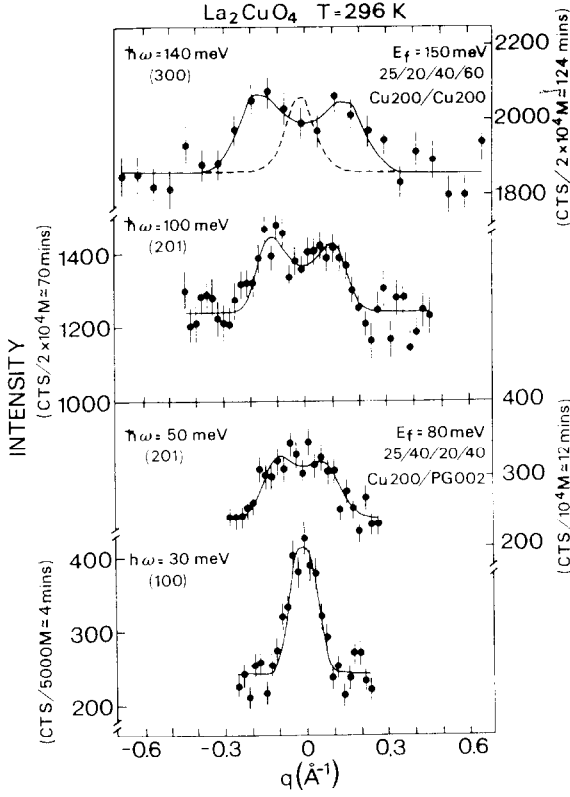


Fig. 1. Transverse constant energy scans through the antiferromagnetic zone center for $\hbar\omega = 30, 50, 100, 140$ meV. Due to kinematic restrictions, scans must be made under different conditions, thus the centers of the scans in momentum transfer were $Q_0 = (100), (201), (201)$ and (300) , respectively. The deviation of the scattering vector Q from Q_0 is q . The solid line represents a full resolution corrected fit of spin wave theory to the data (see text). The dashed line in the upper frame corresponds to the resolution corrected cross section calculated for spin waves with infinite velocity for $\hbar\omega = 140$ meV. Note that the 140 meV scan was performed on a sample of volume 8 cm^3 , other scans were performed using a sample of volume 2 cm^3 .

cross section for undamped antiferromagnetic spin waves [4], which can be written as

$$\frac{\partial^2 \sigma}{\partial \Omega \partial \omega} = \frac{k_i}{k_f} A_q \{ [n(cq) + 1] \delta(\omega - cq) + n(cq) \delta(\omega + cq) \},$$

where k_i and k_f are the initial and final neutron wave vectors, respectively, $n(\omega) = \{\exp(\hbar\omega/kT) - 1\}^{-1}$, the Bose-Einstein occupation factor, and $A_q \sim 1/q$. The

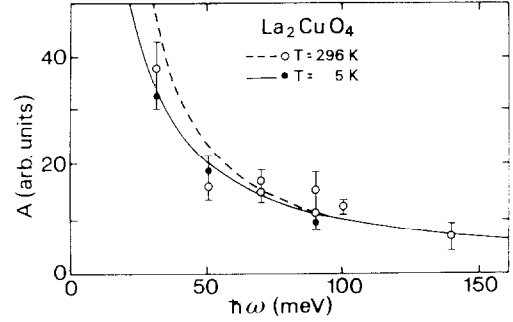


Fig. 2. Fitted spin wave amplitudes for a variety of constant energy scans. The solid and dashed curves are a fit of conventional spin wave theory. Amplitudes measured under different experimental conditions corrected using the known Cu^{2+} form factor. The effect of different analyzers was taken into account by repeating measurements under the different experimental conditions for the same energy transfer.

data were fitted to a convolution of this cross section with instrumental resolution function. The variation of the fitted amplitude and spin wave pole position is shown in figs. 2 and 3. Conventional spin wave theory would predict that the overall amplitude varies as $A \sim (1/\omega)(n(\omega) + 1)$. The solid and dashed lines in fig. 2 show a fit of this form to the data over the energy range investigated. We find that conventional spin wave theory provides a good description of the data at the temperatures investigated. This result is not sur-

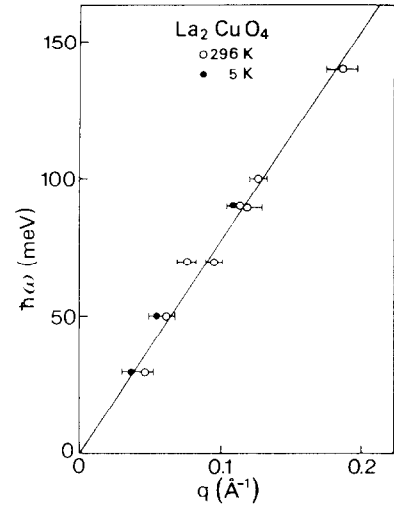


Fig. 3. Wave vectors from fitting conventional spin wave theory to constant energy scans (see caption to fig. 2). The solid line is a fit to the data for $T = 296$ K.

prising, even at $T = 296$ K, since we are probing energies larger than kT and length scales less than the correlation length ξ (from data taken for lower energy transfers we estimate $\xi \cong 200$ Å at $T = 300$ K, for our samples). In this limit, one would expect $\chi(q) \sim q^{-2}$, which is implicit in eq. (1).

From the fitted spin wave pole positions we obtain spin wave velocities $\hbar c = 850 \pm 30$ and 770 ± 40 meV for $T = 5$ and $T = 296$ K, respectively. We can interpret our spin wave velocity using the Heisenberg Hamiltonian,

$$H = \sum_{\langle ij \rangle} JS_i \cdot S_j,$$

where the sum is over nearest neighbor pairs $\langle i, j \rangle$. From conventional spin wave theory, the spin wave velocity is then $c = \sqrt{8SJ}a$. Using our low temperature value for the spin wave velocity, $\hbar c = 850 \pm 30$ meV Å⁻¹, we obtain $J = 0.16$ eV. This is close to the value obtained from two-magnon Raman scattering [5] (0.14 eV). The difference between the two values is probably not significant since there is an uncertainty in the amount by which the peak in the two magnon spectrum is shifted from $8SJ$. In addition, theoretical work [6] shows that c may be renormalized upwards with respect to the conventional value.

In summary, we have investigated the magnetic fluctuation spectrum of La_2CuO_4 in the energy range $30 \leq \hbar\omega \leq 140$ meV. We find that within the experimental resolution, the data can be well described by conventional spin wave theory. Thus at the wave vectors and energies probed the system behaves as an ordered antiferromagnet.

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