HIGH ENERGY INELASTIC NEUTRON SCATTERING FROM La$_2$CuO$_4$

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We have used high energy inelastic neutron scattering to study the magnetic fluctuation spectrum of La$_2$CuO$_4$ in the energy and momentum ranges 30 ≤ ω ≤ 140 meV and |q| ≤ 0.2 Å$^{-1}$, respectively. Our results are consistent, within the experimental error, with conventional spin wave theory the low temperature spin wave velocity, being 850 ± 30 meV Å$^{-1}$.

The magnetic dynamics of La$_2$CuO$_4$ have recently attracted considerable attention. Interest has been motivated by the discovery of superconductivity in the La$_{2-x}$(Ba, Sr)$_x$CuO$_{4-y}$ system and suggestions that La$_2$CuO$_4$ itself is a model system for the two-dimensional (2D) S = 1/2 Heisenberg antiferromagnet [1]. We report here an inelastic neutron scattering study of the magnetic dynamics of La$_2$CuO$_4$ 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.

Points in reciprocal space are labeled such that the basal planes are perpendicular to [0 1 0]. The room temperature lattice parameters (determined from X-ray diffraction) were $a = 5.375(2)$, $b = 13.156(4)$ and $c = 5.409(2)$ Å 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 Cu(2 0 0) monochromator was used with vertically curved pyrolitic graphite (0 0 2) and Cu(2 0 0) 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 [0 1 0] 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 Cu(2 0 0) 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 ω being within the resolution volume, as has also been the case for thermal neutron scattering studies of La$_2$CuO$_4$ [2]. We have analyzed our data in terms of a conventional...
Fig. 1. Transverse constant energy scans through the antiferromagnetic zone center for $h\omega = 30, 50, 100, 140$ meV. Due to kinematic restrictions, scans must be made under different conditions, thus the centers of the scans in momentum transfer were $Q_z = (100), (201), (201)$ and $(300)$, respectively. The deviation of the scattering vector $Q$ from $Q_z$ 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 $h\omega = 140$ 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

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

$$+ n(cq)\delta(\omega + cq)$$

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

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 $T = 296 K$. 

$La_2CuO_4$ $\gamma_{296K}$ $\gamma_{5K}$
prising, even at $T = 296$ K, since we are probing energies larger than $kT$ and length scales less than the correlation length $\xi$ (from data taken for lower energy transfers we estimate $\xi \approx 200$ 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 $770 \pm 40$ meV for $T = 5$ and $T = 296$ K, respectively. We can interpret our spin wave velocity using the Heisenberg Hamiltonian,

$$H = \sum_{\langle i, j \rangle} J S_i \cdot S_j,$$

where the sum is over nearest neighbor pairs $\langle i, j \rangle$. From conventional spin wave theory, the spin wave velocity is then $c = V \sqrt{8SJ}$. 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$_2$CuO$_4$ 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.

References

[4] see for example S.W. Lovesey, Theory of Neutron Scattering from Condensed Matter (Clarendon, Oxford, 1984). By “conventional” here we mean that the ground state is taken to be the Néel state.