NEUTRON SCATTERING MEASUREMENTS ON $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ H.A. Mook,¹ P. Dai,¹ F. Doğan,² K. Salama,³ G. Aeppli,⁴ and M.E. Mostoller¹¹Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393²University of Washington, Seattle, Washington 98195³Texas Center for Superconductivity, University of Houston, Houston, Texas 77204⁴NEC, 4 Independence Way, Princeton, NJ 08540

Polarized and unpolarized neutrons have been used to study the excitations in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. The magnetic excitations are found to be centered at (π, π) in the reciprocal lattice and are sharply peaked in energy at 41 meV in the superconducting state and broad in energy in the normal state. The intensity is found to result from coupled bilayers in both the normal and superconducting state. A new set of incommensurate one-dimensional phonon excitations has been discovered in scans that integrate over energy along planes in reciprocal space. These are found to be very sharply defined in momentum space and are thought to stem from $2k_F$ effects for the Cu-O chains.

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1 Magnetic Excitations

Magnetic inelastic neutron scattering is a powerful technique which gives direct information about the behavior of the electrons in materials. This information is of particular value for high-temperature superconductors where electron correlation effects may be responsible for some of the unusual physical properties of the material. One of the best studied superconductors is $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) ¹⁻⁷. Many years ago, Rossat-Mignod *et al.* ¹ reported enhanced scattering in the superconducting state in the region of 41 meV although other features in the scattering were reported as well. Phonon scattering is found to be stronger than the magnetic scattering in many cases for YBCO and thus polarized beams are needed to distinguish the magnetic scattering. ² The polarized measurements showed that the only scattering found in the superconducting state was centered at 41 meV and that lower energy scattering disappeared as the superconducting gap opened. The normal state scattering first appeared to have a small peak at 41 meV as well ² but improved measurements have shown the normal state scattering is generally featureless in energy, but still centered at (π, π) in the reciprocal lattice. Fong *et al.* ⁴ suggested from unpolarized measurements that the normal state scattering is unobservable, but we ⁵ have shown it is readily found if polarized neutrons are used. Fig. 1(a) & (b) show polarized measurements of the magnetic excitations in YBCO at 100 K and 10 K. The open points are the spin flip scattering and thus show the magnetic intensity directly. The filled points were taken with the HF-VF technique ³ which determines the background directly. The HF-VF measurements are very time consuming and were done at selected points to determine the background. We have found from high resolution unpolarized measurements that the 41 meV peak found in the superconducting state is very narrow in energy and we estimate that its width is as small as 2.5 meV.

The polarized measurements shown in Fig. 1 were taken with a wide resolution of 12.5 meV to obtain sufficient signal to observe the scattering in the normal state. A comparison of the height of scattering in the normal and superconducting state thus must take the energy resolution into consideration. The scattering in the normal state is found to be broad in energy and is not affected substantially by the resolution. Assuming 2.5 meV for

Figure 1: Polarized beam measurements at (0.5,1.5,-1.8). The open circles are the spin flip scattering. The closed circles are obtained by the HF-VF method. (a) shows the normal state scattering at 100 K. (b) is the scattering at 10 K. (c) shows the 10 K scattering where the solid line is the 41 meV peak corrected for resolution assuming a 2.5 meV intrinsic width.

Figure 2: (a) shows measurements at (0.5,0.5,5.4) at 100 K which is essentially the phonon scattering. The solid line is the fit from the shell model calculation. (b) is the temperature subtracted scattering which shows the 41 meV scattering. The solid line is a Gaussian fit.

the width of the 41 meV peak would result in a height shown in Fig. 1(c) where the solid line shows a peak of the same area as that given in the polarized measurements.

The 41 meV peak has attracted considerable attention and a number of theoretical attempts⁷ have been made to explain its origin. The absolute value of the integrated intensity of the 41 meV peak can be obtained if the magnetic scattering is compared carefully to the phonon scattering. The cross section for phonon scattering is given by:

$$\frac{d^2\sigma}{d\Omega dE} = 2.09 \frac{k_f}{k_i} \left(\sum_l e^{-i\mathbf{q}\cdot\mathbf{l}} \frac{[\mathbf{Q} \cdot \boldsymbol{\sigma}_l(\mathbf{q})] e^{-W_l(\mathbf{Q})} b_l}{\sqrt{M_l}} \right)^2 \frac{1}{\omega_q} (1 + n(\omega)) \text{ barns/unit cell} \quad (1)$$

where $\mathbf{Q}$ is the momentum transfer, b_l the nuclear scattering amplitudes of the atoms at site l , $\boldsymbol{\sigma}_l$ the polarization vectors for each branch, M_l the mass of the atom at the site l , $n(\omega)$ the population factor, and W_l the Debye-Waller factors for the atoms. If the frequency ω is given in meV and the masses in amu the units are in barns/unit cell. In order to study both magnetic and non-magnetic scattering, we have made our measurements for $\mathbf{Q} = (0.5, 0.5, 5.4)$ where the phonon modes are not pure modes and have a complex displacement pattern. The polarization vectors then must be calculated and we have done this using a shell model for the lattice dynamics. The shell model uses parameters that give a good account of the optical and neutron measurements on the phonons in YBCO.

Fig. 2(a) shows the unpolarized data at 100 K which is largely the phonon scattering as the normal state magnetic scattering is small on the scale of the phonon scattering.

The line shows a fit to the data using the result of the theoretical phonon scattering cross-section convolved with the experimental resolution. There are a number of phonon modes that involve all the atoms in the unit cell in the energy range shown. The contributions from most of the modes are small, with the largest contributions coming from modes that involve c-axis displacements of the oxygen plane atoms and the oxygen chain atoms. The calculation gives a good account of the measurements so that the theoretical phonon cross section can be relied upon to obtain an absolute scale for the magnetic scattering from the same sample.

For unpolarized neutron scattering from a system of unaligned Heisenberg spins, the cross-section is:

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{\pi} \frac{k_f}{k_i} r_0^2 \frac{1}{2} f^2(\mathbf{Q})(1 + n(\omega)) \chi''_{\alpha\alpha}(\mathbf{q}, \omega) / \mu_B^2 \quad (2)$$

where r_0 is 5.4×10^{-13} cm, and $f(\mathbf{Q})$ is the magnetic form factor. Fig. 2(b) shows the 41 meV peak obtained by taking the difference between 10 K and 100 K. Polarized measurements have shown that the phonon scattering does not change much between 100 K and 10 K. The 100 K magnetic scattering is quite small and energy-independent [Fig. 1(a)] so it doesn't affect the peak height determined for the 41 meV excitation. This measurement then gives the magnetic scattering which can be compared to the phonon scattering.

Since as far as we can determine, the $\hbar\omega_0 = 41$ meV peak is arbitrarily narrow we can rewrite the susceptibility at two-dimensional zone-boundary points (0.5, 0.5, Q_c) as:

$$\chi''_p / \pi = \frac{1}{2} \chi' \sin^2(\pi Q_c / d) (\delta(\hbar\omega - \hbar\omega_0) - \delta(\hbar\omega + \hbar\omega_0)) \hbar\omega_0 \quad (3)$$

where the bilayer spacing is $d = 3.43$ Å and Q_c is in $\text{\AA}^{-1}$. The absolute intensity calibration provided by the phonons yields a value of $4.3 \pm 0.13 \mu_B^2/\text{eV/unit cell}$ for the real part χ' of the magnetic susceptibility associated with the 41 meV resonance.

Now that the size of the 41 meV peak is available in absolute units we expect it will be easier to choose between various suggestions for the origin of the 41 meV peak. In choosing between various scenarios for the origin of the peak it must be remembered that the peak results from a coupled bilayer excitation. We find this to be the case as well for the normal state scattering.

2 Incommensurate Fluctuations

We have also performed a series of investigations to examine scattering from the Cu-O chains. These are one-dimensional in nature so that the scattering from them can be distinguished from other sources of scattering as it should appear as sheets in reciprocal space. We have made scans that integrate in energy along planes of scattering that are perpendicular to the $\mathbf{b}^*(\mathbf{a}^*)$ direction. Since the sample is heavily twinned we cannot distinguish $\mathbf{a}^*$ from $\mathbf{b}^*$. Such scans yield very sharp incommensurate peaks that occur between the standard Bragg reflections.

Fig. 3 shows some of these scans, obtained with an incident energy of 18.93 meV. The satellite peaks narrow as the temperature is reduced and also reduce in intensity. The solid

Figure 3: Excitations observed by integrating over planes of scattering perpendicular to $b^*(a^*)$. These are thought to stem from a dynamic charge density wave associated with the Cu-O chains.

line is a least squares fit to the data using a Gaussian and a sloping background. The peaks are found to be centered at 1.767 along b^* and have a width of 0.01 in b^* which is consistent with the size of the resolution. Additional smaller peaks are found at 0.767 and at 1.233 which represent the same points in reciprocal space. We have checked a number of other directions in reciprocal space and have confirmed the one-dimensional origin of the peaks in Fig. 3. Calculations have also been made that simulate the experiment using the shell model for the phonon positions and structure factors. These show that no structure from the normal phonons that can account for the satellites. Likewise, identical measurements on $\text{YBa}_2\text{Cu}_3\text{O}_{6.15}$ show no satellites either.

The position of the satellites is consistent with calculated values⁸ for $2k_F$ for the Cu-O chains. Since the satellite scattering is inelastic in nature and has the temperature and Q dependence of phonon scattering we propose that it stems from an uncondensed charge density wave associated with the Fermi surface of the Cu-O chains. It is perhaps surprising that the scattering is so narrow in momentum space as the chain bands are expected to hybridize with other bands to some degree. We expect magnetic excitations at the chain $2k_F$ position as well and are in the process of studying these excitations.

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