Polarized Neutron Determination of the Magnetic Excitations in $YBa_2Cu_3O_7$

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Polarization analysis has been used to identify the magnetic excitations in $YBa_2Cu_3O_7$. The dominant feature in the spectra is a peak at the (π,π) reciprocal lattice position and centered at 41 meV. The behavior of the peak is shown to change dramatically at T_c, so that the magnetic excitations responsible for the peak must be strongly coupled to the superconductivity. Below an energy of about 35 meV, superconductivity suppresses the continuum scattering found in addition to the 41-meV peak.

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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 correlations may be responsible for some of the unusual physical properties of the material. One of the best studied superconductors is nearly stoichiometric $YBa_2Cu_3O_7$, which we will denote by $(123)O_7$. This compound has a high transition temperature and a relatively simple structure which makes it easier to deal with for theoretical calculations, particularly those involving the electronic band structure.

There have been a number of interesting neutron scattering measurements on the $(123)O_{7-\delta}$ system [1-7]. Most of these have concentrated on highly oxygen-deficient compositions which are near to the antiferromagnetic phase boundary. For the most oxygen-rich compound studied to date, $(123)O_{6.92}$, Rossat-Mignod *et al.* [5-7] concluded that the magnetic response consisted of several peaks, one of which is located at 41 meV and became visible below T_c. In this paper we describe the first polarized neutron scattering measurements on $(123)O_7$ single crystals. Our data yield important new results. First they show that for the oxygen-rich compounds it is impossible to isolate the magnetic scattering with unpolarized neutrons. Second, in marked contrast to the earlier unpolarized measurements, they show that the magnetic excitations are quite simple, consisting of a continuum and a sharp resonance at 41 meV that is present at all measured temperatures (below 150 K). Furthermore, we find that the 41-meV resonance and the continuum are both affected strongly by the superconductivity, but in qualitatively different ways.

Most of the measurements were made using a 7.8-g single crystal that had a mosaic spread of 0.6° as measured from the (006) reflection and yielded 150000 counts/sec for the (001) reflection in the unpolarized triple-axis mode with the analyzer in place. Some of the polarized beam measurements were made with a 9.3-g sample that had a mosaic spread of 2.6°. Within experimental error, both samples gave the same results.

Pieces cut from both crystals were found by ac susceptibility to have a superconducting transition temperature of 92.4 K with a transition width less than 1 K. Also, the 7.8-g crystal was used for neutron measurements of the fluxoid lattice. These small-angle scattering measurements sample the bulk superconductivity of the material and indicate that the entire crystal superconducts below the transition temperature found for the small piece. Observed room-temperature lattice constants for the crystals are $a=3.823$ Å, $b=3.887$ Å, and $c=11.68$ Å, which are those expected for the $(123)O_7$ composition [8]. The crystals contain Y_2BaCuO_5 as an impurity phase which constitutes up to 15% of the crystal volume. Electron microscopy shows that this phase is found in isolated regions in the crystal, while most of the crystal consists of large regions of very high quality (123). Powder diffraction shows the impurity grains to be oriented randomly so they do not affect our measurements of the magnetic excitations which display the symmetry of the (123) lattice; in fact, the boundaries between these grains and the (123) material are important in providing pathways through the large sample to achieve the full oxygen content.

Measurements were made at the High-Flux Isotope Reactor at Oak Ridge National Laboratory using the HB-1 and the HB-3 triple-axis spectrometers. For the unpolarized measurements, pyrolytic graphite was used as an analyzer while Be or pyrolytic graphite was used as a monochromator. For the polarized beam measurements we used a Heusler analyzing crystal and a ^{57}Fe monochromator. Collimations of 40′ were used typically before and after the monochromator with 60′ and 120′ collimators before and after the analyzer. The final neutron energy was fixed at 30 meV, and a graphite filter was placed before the analyzer in the unpolarized measurements.

Searches for the magnetic scattering were made mostly at the $(0.5,0.5,z)$ reciprocal lattice positions since previous investigations had found the magnetic scattering to peak at these Q positions. Since our crystal is twinned, we do not distinguish between the a and b direction and, thus, these Q's may be thought of as the (π,π) point in square lattice notation. The greatest difficulty in the present experiment is in isolating the magnetic scattering from the phonon scattering and other nonmagnetic processes (e.g., unwanted Bragg scattering in the sample crystal). In fact, we found that the only certain method of determining the energy dependence of the magnetic scattering was to use polarization analysis to isolate the spin-flip scattering. Once the energy dependence of the scattering is determined above and below T_c, it is more efficient to use unpolarized neutrons to establish the temperature- and wave-vector-dependent scattering. Our measurements covered the temperatures between 10 and 150 K. This is the most interesting temperature range, and higher temperatures increase difficulties with phonon contamination.

The first and most important part of the experiment was to establish the energy dependence of the scattering in the normal and superconducting states. As shown below, this can only be accomplished by using polarization analysis. Our measurements were made with a 3×10^{-3}-T guide field applied along the scattering vector. The small H_{c1} of $YBa_2Cu_3O_7$ permits polarized measurements below the superconducting transition. The flipping ratio was 11 in both the normal and superconducting state at zero energy transfer. The flipping ratio was checked at the higher incident energies used in the experiment and was found to be energy independent within the accuracy of the experiment. A measurement of the spin-flip cross section gives the magnetic scattering from the electrons, $\frac{2}{3}$ of the nuclear spin incoherent scattering, room background, plus a contribution from incomplete polarization. The spin incoherent cross sections for the nuclei involved are negligible, while the room background can be determined by turning the analyzer crystal by 2° and repeating the scan. The imaginary part of the generalized susceptibility, $\chi''(\omega)$, is given by subtracting the background from the spin-flip scattering, correcting for incomplete polarization, and dividing by the Bose population factor $n(\omega)+1$. Figure 1(a) shows $\chi''(\omega)$ determined for $Q=(0.5,0.5,-5.2)$ for 100 K. The observed $\chi''(\omega)$ consists of a peak at 41 meV superposed on a flat contribution of about 25 counts per 5000 monitor. There is an energy region around 25 meV where measurements are impossible because of the (006) and (114) reflection

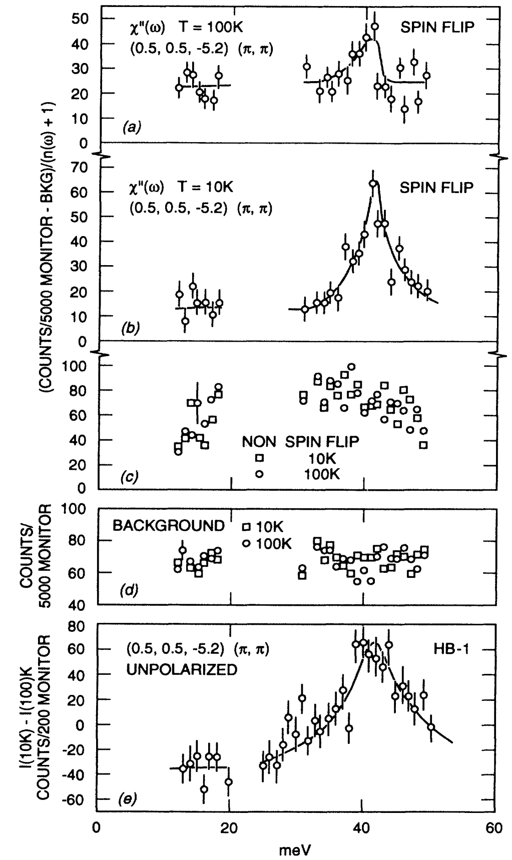

FIG. 1. (a) A polarized beam measurement of χ'' for the (π,π) position for $T=100$ K. (b) The same measurement but for 10 K. (c) The phonon scattering normalized to 5000 monitor counts. The actual counting time was half that for the magnetic scattering resulting in larger error bars. (d) The analyzer-turned background. (e) The result of an unpolarized measurement in which data obtained at 100 K are subtracted from 10-K data.

which are not fully rejected by the analyzer. Figure 1(b) shows $\chi''(\omega)$ determined at 10 K in the superconducting state. $\chi''(\omega)$ is now dominated by a larger peak at 41 meV. The 41-meV peaks are energy resolution limited (5.4 meV). There is still an energy-independent contribution, but it is noticeably smaller, about 15 counts per 5000 monitor. Figure 1(c) shows the non-spin-flip scattering with the background subtracted and scaled by $n(\omega)+1$. This is essentially the phonon scattering, and we see that its intensity is unchanged between 100 and 10 K [9]. The analyzer-turned background is given in Fig. 1(d); this is known to be structureless from prior experience.

Our results for $\chi''(\omega)$ are very different than those obtained by Rossat-Mignod *et al.* [5-7] for their $(123)O_{6.92}$ sample in that their 100-K susceptibility is dominated by a large contribution at 30 meV. We see no such contribution. We note, however, from Fig. 1(c), that the phonon scattering is large at this energy and is about 7 times the magnetic scattering at 10 K. Properly identifying the energy dependence of the magnetic scattering without using polarization analysis would be extraordinarily difficult.

Since we have determined that the intensity of the phonon scattering does not change between 100 and 10 K, we can use unpolarized neutrons to examine temperature-dependent effects. Figure 1(e) shows the difference obtained between an energy scan at 10 K and the same scan taken at 100 K. We see clearly the increase in the large peak near 41 meV at low temperatures, and that the lower energy scattering decreases at 10 K. Figure 2 shows scans performed using unpolarized neutrons at 41 meV across the (π,π) position made for 150, 100, and 10 K. The scattering is independent of temperature between 150 K and the superconducting transition T_c but narrows and increases in peak intensity as the temperature is lowered below T_c. Figure 3(a) shows the temperature dependence of the peak intensity which parallels, to a remarkable extent, that which is expected for the superfluid density. Obviously, while it appears to be controlled by the superconducting order, this intensity does not measure the superfluid density directly because it does not vanish at T_c. Figure 3(b) shows the temperature dependence of the peak widths (FWHM) in Q. These are derived from the Gaussian functions indicated by the solid lines shown in Fig. 1, which best fit the data.

Figure 3(c) shows the dependence of the subtracted 41-meV scattering on the momentum transfer along the (0,0,1) direction. We see that the intensity modulation follows the behavior expected for an acoustic mode for the antiferromagnetically coupled bilayers in $(123)O_6$; also, it is essentially the same as that shown by Tranquada *et al.* [4] for the 27-meV excitation in $(123)O_{6.6}$. The presence of the modulation shows that the excitation at 41 meV involves a correlated motion of the electron spins in the near-neighbor Cu-O planes. Figure 3(d) shows the strength of the difference in scattering measured at different Q points in the zone where the scattering is expected to be identical except for the magnetic form factor

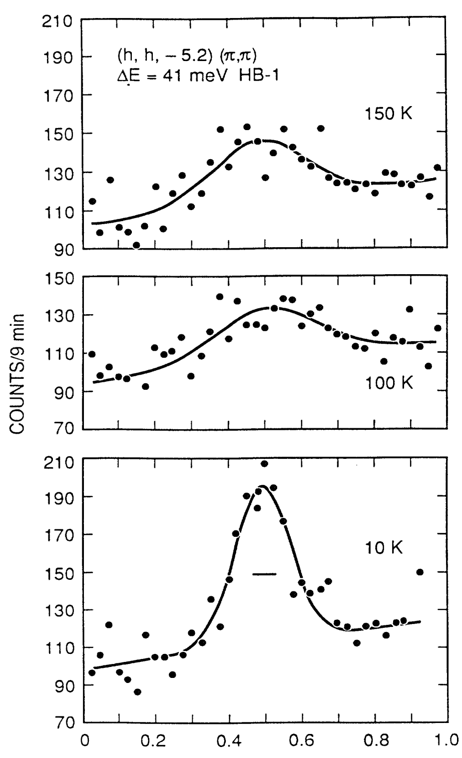

FIG. 2. Constant-energy scans of the 41-meV peak at temperatures above and below T_c. The bar on the graph shows the spectrometer resolution (FWHM).

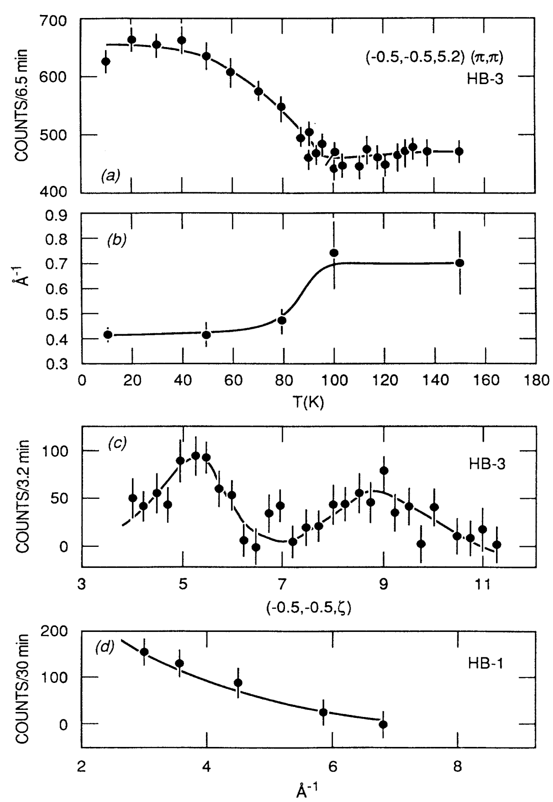

FIG. 3. (a) The temperature dependence of the peak intensity of the 41-meV feature. A number of counts were made at the same spectrometer position for each temperature and averaged together to obtain the count rates and errors shown on the figure. (b) The full width at half maximum obtained by least-squares analysis of the peak shape. In (c) and (d), results are given for the intensity at 41 meV and 100 K subtracted from the intensity at 41 meV and 10 K. The intensity modulation along $(0.5,0.5,z)$ is plotted in (c). The intensities at $(0.5, 0.5,-5.2)$, $(1.5,1.5,-1.8)$, $(1.5,1.5,-5.2)$, $(2.5,2.5,-1.8)$, and $(2.5,2.5,-5.2)$ are given in (d). The solid line is the Cu^{++} form factor squared, normalized at the first data point.

dependence. The solid line is the Cu^{++} form factor squared, normalized to the first point. The data points fall on the line, confirming again the magnetic nature of the scattering.

It is tempting to attribute the intensity reduction of $\chi''(\omega)$ at energies below 41 meV, seen in both the polarized data [Figs. 1(a) and 1(b)] and the unpolarized temperature difference [Fig. 1(e)], to the establishment of a gap in the spin fluctuation spectra. The zero crossing in the difference spectrum [Fig. 1(e)] would then represent the gap which would be about 35 meV ($4.1kT_c$), not far from analogous zero crossings in electronic Raman scattering [10]. Work is in progress to further characterize this effect. Our view of the temperature evolution of the spin fluctuations is again different from that proposed by Rossat-Mignod *et al.* [5–7] in that we find that the 41-meV feature remains fixed in energy at all temperatures rather than shifting to lower energies at the higher temperatures.

The origin of the large 41-meV feature is not clear. Its energy is close to that of a Raman active phonon with substantial coupling to the carriers [10] and somewhat less than the threshold (54 meV) for strong infrared absorption [11] in $(123)O_{7-\delta}$. Because of its modulation along c^*, the feature is due to an excitation where nearest-neighbor moments in adjacent planes are strongly correlated. One might argue that the resonance arises from constant-E surfaces for excited quasiparticles that happen to be most parallel for an energy difference of 41 meV. This description, as well as that where we ascribe the excitation to an interband transition, has the difficulty that unless we abandon the notion that quasiparticles have strong temperature- and energy-dependent mean-free paths in the high-T_c oxides [11,12], the feature would be very temperature dependent, if even resolvable, in the normal state. We thus turn from the obvious band descriptions to a picture where the 41-meV excitation would correspond to a singlet-triplet transition of pairs formed between neighboring spins in adjacent layers. Such a resonance could exist for a pair of Cu-O planes even though local moments do not appear in the individual planes. The magnetic correlation length ξ manifested in the widths of Q scans, such as those in Fig. 2, then reflects the effective in-plane exchange coupling J_{ab} at the singlet-triplet transition energy. In the normal state, incoherent motion of the carriers in the planes would make J_{ab} small and ξ short. On the other hand, in the superconducting state, fewer carriers are available to reduce J_{ab} and ξ would grow correspondingly.

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