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Spectral shift of the magnetic cross section in superconducting $YBa_2Cu_3O_{6+x}$

P. Bourges, P.M. Gehring,* B. Hennion, and A.H. Moudden
Laboratoire Léon Brillouin, Centre d'Etudes Nucléaires de Saclay, 91191 Gif-sur-Yvette, CEDEX France

J.M. Tranquada and G. Shirane
Brookhaven National Laboratory, Upton, New York 11973

S. Shamoto and M. Sato
Nagoya University, Nagoya 464-01, Japan

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A triple-axis neutron-scattering study has been done of the magnetic correlations in three large single crystals of $YBa_2Cu_3O_{6+x}$ with x=0.5, 0.8, and 0.9 having T_c's of 50, 60, and 80 K, respectively. The frequency dependence of the cross section at $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},l)$ was found to peak near 5 meV for x=0.45 in an earlier study. Our data confirm that this peak shifts to $\sim$ 30 meV for x=0.5. Moreover, the low-frequency (<40 meV) spectra for x=0.8 and 0.9 become progressively suppressed with respect to those of the less oxygenated samples. These data are consistent with a picture in which the spectral weight of the magnetic scattering shifts to higher energy as the oxygen concentration x and carrier density increase.

The existence of magnetic correlations in the superconducting region of the $YBa_2Cu_3O_{6+x}$ phase diagram has been a subject of considerable debate in recent years. Well-defined antiferromagnetic Bragg peaks are observed in the insulating phase of $YBa_2Cu_3O_{6+x}$ for $x<0.4$ in neutron-scattering experiments on single crystals.[1-3] For $x\geq 0.4$, antiferromagnetic spin fluctuations have been inferred from NMR measurements of Cu^{2+} nuclear-relaxation rates[4] as well as from two-magnon Raman-scattering experiments.[5] In addition, inelastic neutron-scattering measurements on a large superconducting single crystal of $(La_{1.85}Sr_{0.15})CuO_4$ (T_c=33 K) have documented the presence of spin fluctuations in the superconducting state.[6] However, the absence of any appreciable magnetic cross section in a fully oxygenated (x=1.0) powdered sample of $YBa_2Cu_3O_{6+x}$, as determined in a polarized-neutron study,[7] led to the conclusion that the Cu atoms carried no moment and cast serious doubt on magnetic-pairing models of the high-T_c phenomenon.

Several papers have now shed significant light on how these spin fluctuations evolve with increasing oxygenation.[8-10] The presence of strong two-dimensional (2D) magnetic correlations was established at low (3 meV) frequency in two crystals having oxygen concentrations of x=0.4 and 0.45, which places them just beyond the border with the insulating phase. A more highly concentrated crystal with x=0.5, however, displays a markedly smaller magnetic cross section at identical excitation energies and temperatures.[8] In order to understand this, two more recent studies[9,10] extended these measurements on the same crystals to higher ($\sim$ 15 meV) energies. The results from these studies provided clear evidence of antiferromagnetic spin fluctuations in $YBa_2Cu_3O_{6.5}$. It was found that, while the magnetic cross section for the x=0.5 crystal was indeed small at low energies, the cross section increases with increasing excitation energy. The cross section for the x=0.45 sample, on the other hand, decreases with increasing excitation energy above approximately 5 meV. In this paper we present additional data for x=0.5, and data for two orthorhombic, superconducting single crystals of $YBa_2Cu_3O_{6+x}$ with x= 0.8 and 0.9. The respective T_c's of the samples, determined by ac inductive methods, are 50, 60, and 80 K. All crystals were grown and treated at either the Institute for Molecular Science or at Nagoya University. Details of the sample growth and preparation have been described elsewhere.[1] The neutron-scattering measurements on these samples cover a temperature range from 15 to 300 K and up to 40 meV in energy.

The measurements were performed at the Laboratoire Léon Brillouin Orphée reactor in Saclay on the 1-T triple-axis spectrometer. Pyrolytic-graphite (002) reflections were used for both the monochromator and analyzer. The monochromator was vertically focused at each energy to maximize the available incident neutron flux. Collimations were 40′-40′-60′-60′ from reactor to detector. Higher-order contamination of the beam was minimized using a pyrolytic-graphite filter, the neutron final energy being held fixed at either 14.7 or 30.5 meV for most of the measurements. The x=0.5 sample was held in a small Al box, and the other samples were wrapped in Al foil and affixed to an Al sample holder. Each crystal was then mounted inside a thin-walled Al can filled with

He exchange gas and attached to the end of the cold finger of a Displex closed-cycle refrigerator. The temperature was monitored using either a Pt or Ge resistor as sensor, depending on the temperature range. The [$1\bar{1}0$] direction was oriented vertically to allow access to the tetragonal (hhl) scattering zone. The longitudinal acoustic phonon at (0,0,6.25) was used to determine relative sample volumes and is shown in Fig. 1. An earlier single-crystal study[2] found the sample volumes of both the x=0.45 and 0.5 crystals to be about 1 cm^3. From Fig. 1 it is clear that the x=0.9 crystal has almost the same volume as that of the x=0.45 sample. Therefore all three crystals (x=0.45, 0.5, and 0.9) have roughly equal volumes, whereas that of the x=0.8 sample is smaller by about a factor of 2. The inset displays the room-temperature value of the tetragonal c-axis lattice constant for each sample (solid squares) relative to those measured previously (open circles).[2] The solid line is a guide to the eye.

Because of the bilayer nature of the antiferromagnetically coupled copper oxide planes in $YBa_2Cu_3O_{6+x}$, the two-dimensional magnetic scattering along the $(\frac{1}{2},\frac{1}{2},l)$ rod is modulated by the factor $\sin^2(\pi z l)$, where zc_0 is the spacing between the individual CuO_2 planes measured along the c axis. Scans were thus constrained to values of l where the modulation was a maximum.

Figure 2 is a composite of results obtained at Risø, Chalk River, and Saclay.[8,9] Data for the x=0.45 sample come from constant-Q scans, corrected for a constant background, taken at $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},1.8)$ at a fixed final neutron energy E_f of 14.7 meV. All other data points were derived from constant-E scans. The Risø data were taken using E_f=13.7 meV and those of Saclay using E_f=30.5 meV. The solid lines are fits of the data (solid circles) by Tranquada *et al.*[9] using the standard formula for paramagnetic scattering from a system of correlated spins. The open triangles for x=0.5 and 0.9 are constant-E scans from this work and have also been corrected for a constant background. The x=0.5 data have been scaled to match the fit of Tranquada *et al.* at 12 meV. Data for x=0.9 have been scaled by the same factor. Because of the nearly identical sample volumes shared by all three crystals, it is not unreasonable to put the data on the same relative scale.

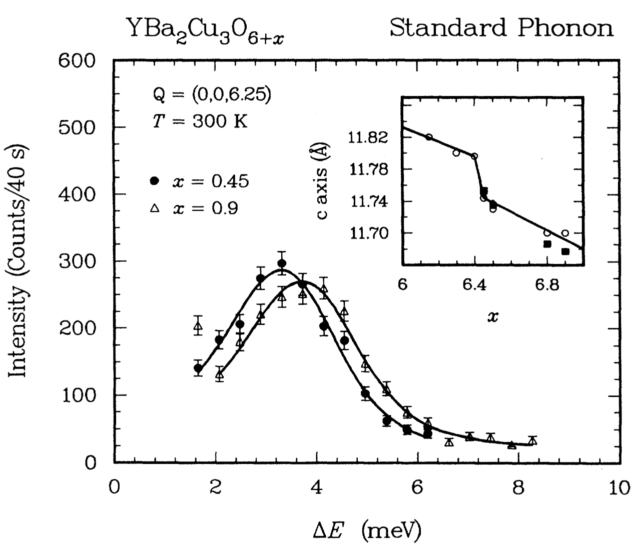

FIG. 1. Comparison of standard phonon scans for x=0.45 and 0.9. The solid lines are fits to a damped harmonic oscillator with a slope of 26.5 meV Å. The inset displays the x dependence of the tetragonal c-axis lattice constant at room temperature. Solid squares are data from this work, and open circles are from a prior single-crystal study by Tranquada *et al.* (Ref. 10). The solid line is a guide to the eye.

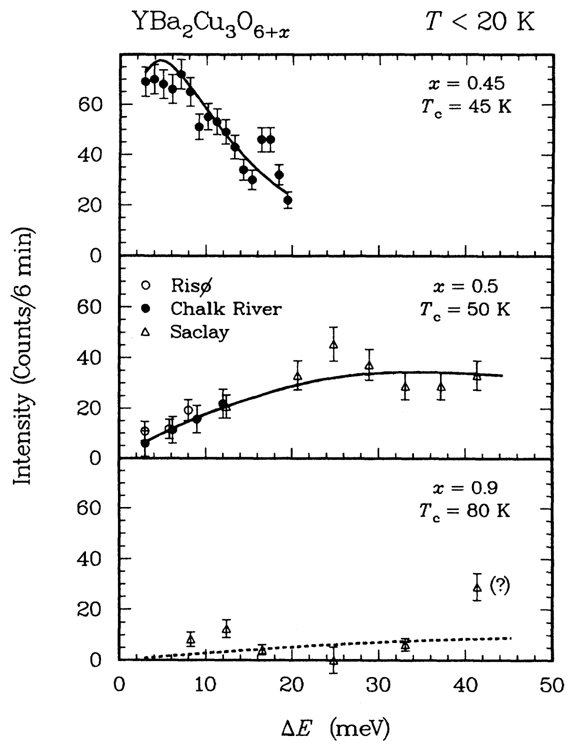

FIG. 2. Constant-Q scans measured at $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},1.8)$ and $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},5.0)$ at $T \leq 20$ K for x=0.45 and 0.5 are fitted with the solid line. Open triangles for x=0.5 and 0.9 represent constant-E scans taken at Saclay. Note that these data have been scaled to match the data taken at Chalk River. The counting times at Saclay were still 6 min per point. A constant background has been subtracted from all of the data at each energy.

A definite trend is immediately evident from these data, which show a progressive suppression of the magnetic scattering at low frequencies in the presence of increasing carrier concentration. The peak in the frequency dependence seen at x=0.45 near 5 meV clearly shifts to higher energy (~30 meV) at x=0.5. This shift is even more pronounced in the case of the x=0.9 crystal where the low frequency (<40 meV) region of the spectrum is greatly diminished in intensity with respect to those of the less oxygenated samples. The dashed line is obtained from the fit to the x=0.50 cross section after doubling the size of Γ from 30 to 60 meV. Although not a fit, this line indicates the difficulty of measuring the magnetic cross section in more highly doped samples.

For x=0.9, a strong increase in scattering is seen above 40 meV centered about $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},l)$, just where magnetic scattering is expected. This suggests the spectral weight of the magnetic scattering has shifted to higher frequencies and is not observable below 40 meV. However, we wish to emphasize that we have not yet demonstrated that this scattering is free from phonon contamination. Further measurements are being carried out to study the scattering cross section in highly doped samples at higher energies. We mention that the low-frequency data for x=0.8 fit well between those for x=0.5 and 0.9, and lend support to the trend of a spectral shift to higher energies. However, the small size of this sample make a reliable determination of its cross section very difficult, and we await the growth of a larger sample before presenting data for this composition.

In Fig. 3 we show typical constant-E scans across the 2D rod at energy transfers of 8.3 and 33.1 meV for x=0.5, and 24.8 meV for x=0.9. The solid lines are best fits to a Gaussian line shape, corrected for a background linear in q. A strong, two-dimensional cross section is seen for the x=0.5 sample at 8.3 meV along $(h,h,-1.8)$ at 52 K, indicating the presence of antiferromagnetic spin fluctuations. At higher excitation energy, the linewidth is essentially the same, although the background increases by more than a factor of 2 upon changing the neutron final energy from 14.7 to 30.5 meV. Between 20 and 300 K, the fitted linewidths show no temperature dependence. At 8.3 meV, the resolution was only 25% of the linewidth. At 33.1 meV, the resolution was 50% of the linewidth. The cross section for x=0.9 at 24.8 meV is greatly reduced by comparison. This appears to be true at higher temperatures as well, since scans at 12.4 meV at 150 K showed no evidence of any magnetic scattering. However, very few scans were performed above T_c for x=0.9, and more data is needed to confirm this result.

Following Tranquada *et al.*,[9] one might want to model the $(h,h,1.8)$ constant-E scans across the magnetic 2D rod with a modified paramagnetic cross section. Although speculative, since propagating spin-waves do not exist, this description seems to be quite appropriate given the 2D character of the antiferromagnetic fluctuations. Thus we suggest that the difference between the resolution function (dashed line) and the measured signal in Fig. 3 results either from the effect of a finite pseudo-spin-wave velocity c or from a significantly large inverse correlation length $\kappa = 1/\xi$. Of course both effects might

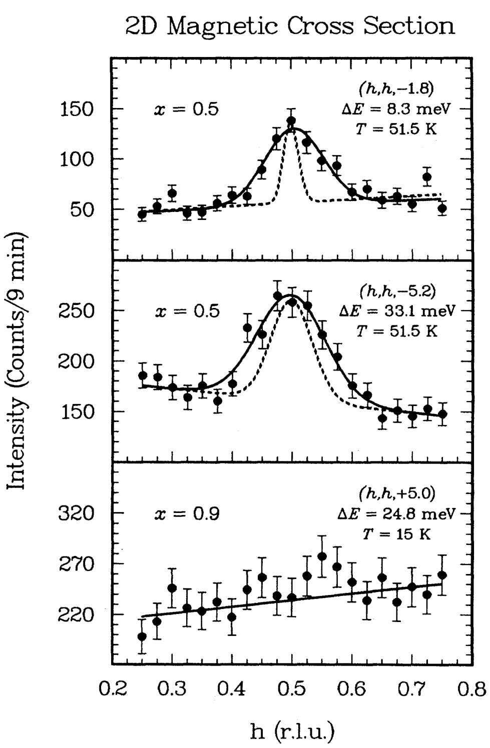

FIG. 3. Constant-E scans at 8.3 and 33.1 meV for x=0.5 and 24.8 meV for x=0.9. The neutron final energy E_f was fixed at 14.7 meV for scans at an energy transfer of 8.3 meV, and at 30.5 meV otherwise. The solid lines are fits to a Gaussian line shape corrected for a sloping background. The dashed lines are simulations for spin waves with infinite velocity. h is measured in reciprocal-lattice units (r.l.u.), where 1 r.l.u.=2.30 Å^{-1}.

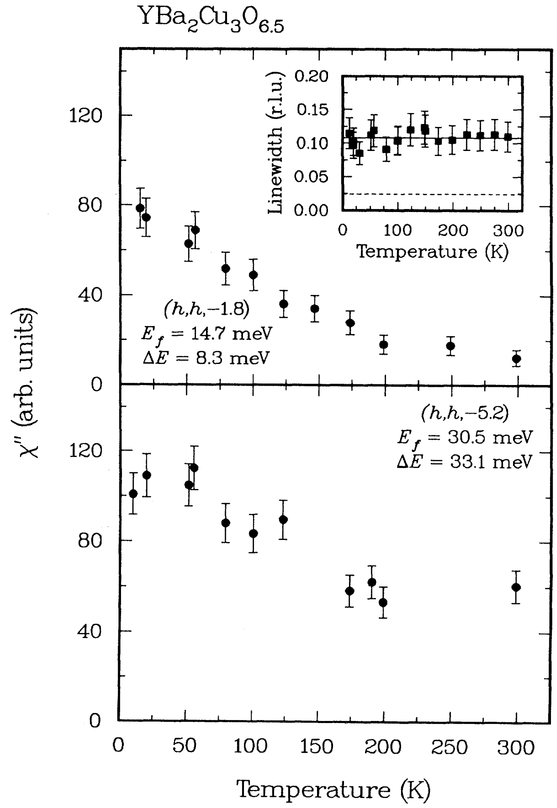

FIG. 4. Temperature dependence of $\chi''(\omega)$ at 8.3 and 33.1 meV. Points represent the peak intensity of constant-E scans determined after fitting to a Gaussian line shape and correcting for background. Data have been corrected by the Bose factor. The temperature dependence of the linewidth (full width at half maximum), measured in reciprocal lattice units (r.l.u.), is shown in the inset.

be present simultaneously. Accordingly we performed fits of the measured cross section using the paramagnetic cross section [see Eq. (1) of Ref. 9] convolved with the instrumental resolution function at a fixed temperature of 30 K under different resolution conditions ($E_f = 14.7$ meV for ΔE=8.3 and 12.4 meV, and $E_f = 30.5$ meV for scans taken at ΔE=24.8, 33.1, and 41.5 meV). We kept the parameters c and κ constant during the fit for all equivalent scans (performed under different resolution conditions). The best fit yields $c = 200 \pm 40$ meV Å and $\kappa = 0.11 \pm 0.01$ Å^{-1}, which translates into a correlation length of $\xi = 9$ Å, in agreement with the measurements made by Rossat-Mignod *et al.*[11] on a sample with the same oxygen concentration. Moreover, the fitted values of κ and c yield a value of $\Gamma = 22 \pm 7$ meV, in reasonable agreement with the fit of Tranquada *et al.*[9]

Figure 4 shows data for $YBa_2Cu_3O_{6.5}$ in which constant-E scans were performed along $(h,h,-1.8)$ and $(h,h,-5.2)$ at 8.3 meV and 33.1 meV, respectively. These scans have been corrected for a temperature dependent background and fit to a Gaussian line shape. The resulting fitted peak intensities are plotted versus temperature. In this figure we have removed the Bose factor so that χ'' is presented. These data represent an improvement upon those of Chou *et al.*[10] In that paper the change in intensity with temperature was attributed to a temperature-dependent correlation length $\xi(T)$. Our observation of a temperature-independent linewidth (see inset to Fig. 4), which is directly related to the correlation length, would seem to rule out this interpretation.

Very recently Rossat-Mignod *et al.*[11] reported on the neutron-scattering cross sections observed in superconducting single crystals of $YBa_2Cu_3O_{6+x}$ with x=0.51 (T_c=47 K) and x=0.69 (T_c=59 K). Two sets of measurements, those of Brookhaven and Grenoble, are in essential agreement on two very important points: (1) magnetic correlations are seen in superconducting samples of $YBa_2Cu_3O_{6+x}$, and (2) the spectral weight of these correlations shifts to higher frequency in the presence of increasing hole concentration. Yet some questions remain concerning the variation of χ'' with temperature at low frequency. In particular, we have not observed a marked change in the scattering intensity at T_c.

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*Present address: Physics Department, Brookhaven National Laboratory, Upton, NY 11973.

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