Magnetic correlations and energy gap in superconducting $YBa_2Cu_3O_{6.6}$ with T_c=53 K

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The dynamic magnetic correlations have been characterized in a large, orthorhombic single crystal of $YBa_2Cu_3O_{6+x}$ with x=0.6, having a T_c of 53 K. Inelastic-neutron-scattering measurements reveal a gap (zero magnetic cross section) of 5 meV in the spin-excitation spectrum at 10 K. The size of the gap fits well between those reported by Rossat-Mignod *et al.* for x=0.51 and 0.69. However, the rapid change in the gap size with a relatively small change in T_c is not understood.

Over the past two years, inelastic-neutron-scattering studies have demonstrated the existence of dynamic magnetic correlations in superconducting single crystals of $YBa_2Cu_3O_{6+x}$.[1-4] The data show that the imaginary part of the spin susceptibility, $\chi''(\mathbf{Q},\omega)$, is characterized in reciprocal space by a broad peak centered about the two-dimensional (2D) antiferromagnetic (AF) zone center, which in three dimensions forms the tetragonal rod $\mathbf{Q}_{AF}$ $(\frac{1}{2},\frac{1}{2},l)$ measured in reciprocal lattice units (r.l.u.). For $x \geq 0.5$ this peak width corresponds to a spin-spin correlation length on the order of twice the lattice spacing. As for the frequency dependence, the usual $1/\omega$ behavior observed for spin waves in the antiferromagnetic phase is strongly suppressed at low frequencies. It has been shown that the observed magnetic cross section is consistent with a model in which antiferromagnetic spin fluctuations are heavily overdamped due to the short correlation length.

In addition to the overdamping of the low-frequency excitations, Rossat-Mignod *et al.*[5] have reported a temperature-dependent energy gap in superconducting $YBa_2Cu_3O_{6+x}$ crystals with x=0.51 and 0.69 (T_c=47 and 59 K, respectively). They observed the gap in measurements of the frequency dependence of χ'' at $\mathbf{Q}_{AF}$. Recently, Bulut and Scalapino[6] have pointed out that they obtain a pseudogap in χ'' precisely at $\mathbf{Q}_{AF}$ from a random-phase approximation (RPA) analysis of the normal-state properties of a 2D Hubbard model; the gap occurs for kinematic reasons and does not appear at the incommensurate positions where their theoretical χ'' peaks. In this paper, measurements are presented on a single crystal with x=0.6 and T_c=53 K which convincingly demonstrate that the energy gap exists not only at $\mathbf{Q}_{AF}$, but all along the zone diagonal. At 10 K the $\mathbf{Q}$-dependent magnetic peak vanishes (within experimental error) for energies $\hbar\omega \leq 5$ meV.

The measurements described here were performed on the BT9 triple-axis spectrometer at the National Institute of Standards and Technology. The pyrolytic graphite (002) reflection was used to monochromatize and analyze the incident and scattered neutron beam. Collimations were 40′-40′-80′-100′ from reactor to detector. Most data were taken using a fixed incident neutron energy E_i of 30.5 meV. A fixed incident neutron energy of 14.7 meV was used for measurements at an energy transfer of 3 meV in order to avoid the tail of elastic scattering which is present due to the finite energy resolution of the spectrometer. A pyrolytic graphite filter was positioned before the sample to minimize higher-order neutron contamination of the beam.

The large (~1 cm^3) single crystal of $YBa_2Cu_3O_{6+x}$ described here was grown at the Institute for Molecular Science in Japan with an oxygen content of x=0.45 and a T_c of 45 K. The crystal was labeled IMS No. 30 in prior studies.[1-3] For the purposes of the present study, the crystal has since undergone an annealing treatment at Nagoya University in order to increase its oxygen content to x=0.6. An ac-susceptibility measurement of the superconducting transition has its midpoint at 53 K and a width of ~5 K. Details of the sample growth and preparation have been described elsewhere.[8] For the neutron study, the $YBa_2Cu_3O_{6.6}$ sample was placed in a small Al box and mounted inside an Al can filled with He gas for exchange purposes. A Displex closed-cycle refrigerator was used to vary the sample temperature between 10 and 300 K using a Ge resistor as sensor.

In order to study the tetragonal (hhl) scattering zone, the crystal was oriented with the $[1\bar{1}0]$ direction vertical. Because of the bilayer structure of antiferromagnetically coupled CuO_2 planes in $YBa_2Cu_3O_{6+x}$, constant-E scans along the 2D rod $(\frac{1}{2},\frac{1}{2},l)$ are modulated by the magnetic inelastic structure factor $\sin^2(\pi z l)$, where zc is the spacing, measured along the c axis, between the individual CuO_2 planes. This modulation is quite useful inasmuch as it provides a signature of the magnetic nature of the scattering. Scans across the rod were constrained to $l=1.8$ where the modulated intensity was a maximum.

Figure 1 shows several representative constant-E scans as a function of the in-plane momentum transfer taken along the zone diagonal $(h,h,1.8)$ at energy transfers of 5, 9, and 12 meV at 10 K. Each scan was fit to a Gaussian line shape plus a constant sloping background, the sum of which is indicated by a solid line. Clearly the most striking feature is the absence of a measurable **Q**-dependent magnetic cross section at 5 meV. An identical scan at 3 meV also revealed no sign of a peak. In spite of the low counting rates associated with the magnetic signal, a clear and well-defined cross section is seen to emerge at 9 meV, becoming even more prominent at 12 meV. The difficulty inherent in these measurements becomes apparent when one realizes that the magnetic signal at 9 meV, counting 15 minutes per point, is only $\sim$ 30 counts above a background of $\sim$ 60 counts. The low counting rate stems in part from the very short spin-spin correlation lengths ξ present in superconducting samples. For $x=0.60$, the constant-E scans at 10 K have full widths at half maximum (FWHM) of order $\simeq 0.12 \pm 0.02$ r.l.u. which implies a correlation length of only $\xi \simeq$ 7–9 Å.

The complete set of measurements at 10 K is summarized in Fig. 2 where the one-dimensional q-integrated intensity of constant-E $(h,h,1.8)$ scans is plotted as a function of energy transfer. The magnetic intensity is zero within experimental error up to 5 meV at 10 K. Above this energy, the spectral weight of the spin fluctuations begins to increase until it appears to saturate between 12 and 15 meV. The size of the gap is therefore taken to be $\simeq$ 5 meV ($\simeq$ 60 K), which is of order T_c. In order to show the consistency between these results and those of Rossat-Mignod *et al.*,[5] the measured low-temperature frequency dependence of the magnetic cross section for $x=0.6$ is replotted in Fig. 3 alongside those reported for two samples corresponding to $x=0.51$ and 0.69. The gap energy of 5 meV for $x=0.6$ is nicely bracketed by these two samples having both higher and lower oxygen contents. The data for $x=0.51$ and 0.69 represent the difference between two constant-**Q** scans taken on and off the 2D AF rod, whereas the data for $x=0.6$ are integrated intensities derived from constant-E scans. Figure 3 also shows data for an $x=0.5$ sample which exhibits a finite magnetic cross section down to an energy transfer of 3 meV.[1-3] The size of the gap observed at low temperature in these samples appears to change rapidly

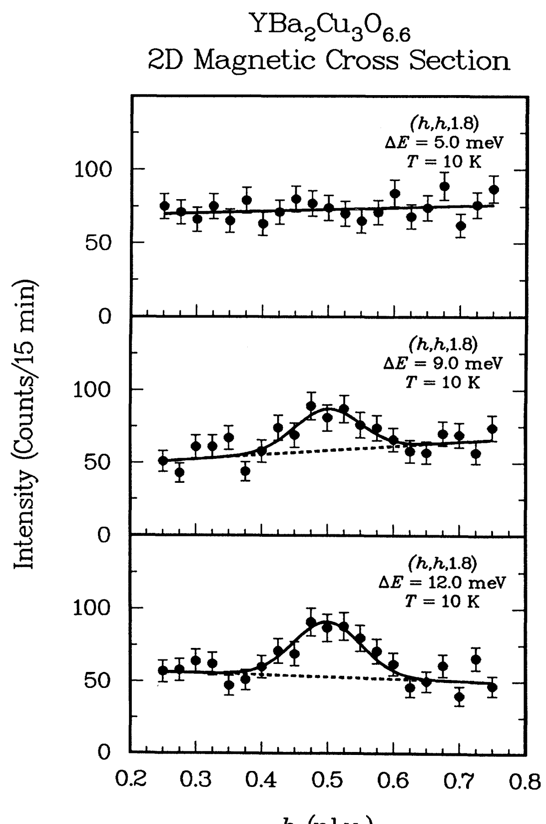

FIG. 1. Constant-E scans measured along $(h,h,1.8)$ at 5, 9, and 12 meV at 10 K, using a fixed incident neutron energy of 30.5 meV. Solid lines represent best fits to a Gaussian line shape. h is measured in reciprocal lattice units (r.l.u.), where 1 r.l.u.$=2.30$ Å^{-1}.

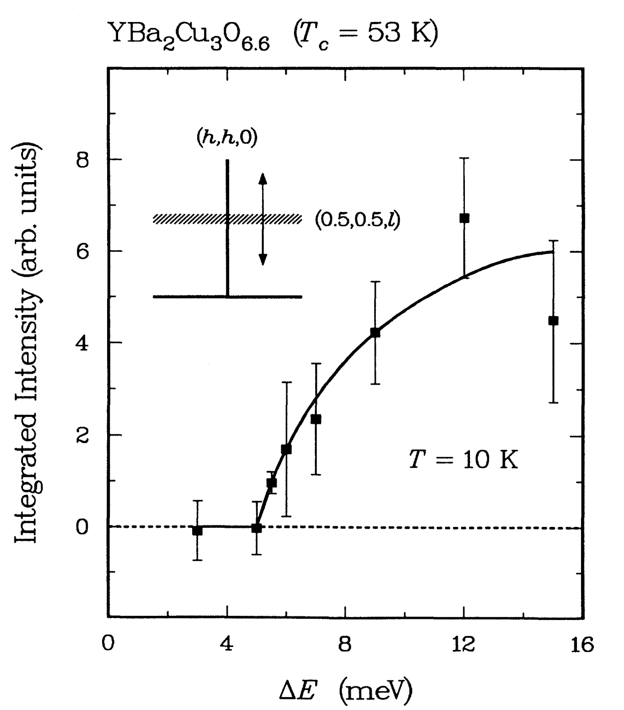

FIG. 2. Frequency dependence of the magnetic cross section. Data points are the integrated intensities from Gaussian fits to $(h,h,1.8)$ constant-E scans as shown schematically in the inset. Each scan was corrected for a constant, sloping background and corrected for the spectrometer resolution. The solid line is a guide to the eye.

with oxygen content, but shows little correlation with T_c.

Because of the time-consuming nature of these measurements, only limited results at higher temperatures have been obtained so far. The temperature dependence of the magnetic cross section at two energies is shown in Fig. 4. The data for 3 meV are constant-E scans along $(h,h,1.8)$, identical to those presented in Fig. 1. The 12-meV data represent the intensity measured on the 2D rod at $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},1.8)$ from which was subtracted a background signal measured at $\mathbf{Q}=(\frac{1}{4},\frac{1}{4},1.8)$. As a check, constant-$E$ scans at 12 meV were taken between 10 and 150 K which showed no change in intensity for $h=\frac{1}{4}$. Compared to the 10 K data, there is a large increase in intensity at 80 K for 3 meV. The data at 12 meV, on the other hand, are essentially temperature independent. A weak temperature dependence at higher frequencies is consistent with measurements on an x=0.5 crystal[3,7] where a slight decrease in magnetic scattering was observed between 10 and 300 K at energies of 6, 8, and 33 meV. The temperature dependence of the low-frequency scattering in the x=0.6 crystal is clearly quite different.

Rossat-Mignod *et al.*[5] have reported more extensive results on the temperature dependence for their x=0.69 sample. Plotting their data in the form of $\chi''(\mathbf{Q}_{\rm AF},\omega)$, they find that the gap smears out gradually with increasing temperature, but does not completely vanish until well above T_c. They have also pointed out the similarity between the temperature dependence of $\chi''(\mathbf{Q}_{\rm AF},\omega)$ for small ω and the unusual behavior of the nuclear spin-lattice relaxation rate $1/T_1$ for Cu measured by nuclear magnetic resonance (NMR) in 60-K material.[9,10] Thus, it appears that the strong decrease observed in $1/T_1T$ as the temperature decreases toward T_c may be due to gaplike behavior in the spin susceptibility, rather than to a uniform change in the spin-fluctuation spectrum. Such a connection between an energy gap and the be-

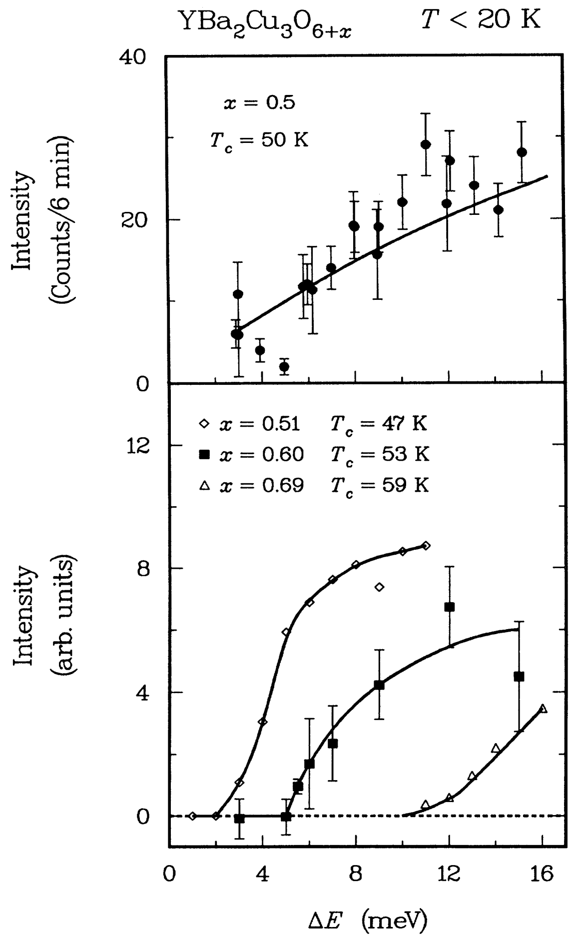

FIG. 3. A composite figure showing the frequency dependence of the magnetic scattering cross section for x=0.6 taken from Fig. 3 relative to those for x=0.51 and 0.69 measured by Rossat-Mignod *et al.* (see Ref. 4). The solid lines are guides to the eye. The frequency dependence for the x=0.5 sample exhibits a finite cross section down to 3 meV. The solid line for x=0.5 is a fit assuming paramagnetic scattering from a system of correlated spins (see Refs. 1 and 2).

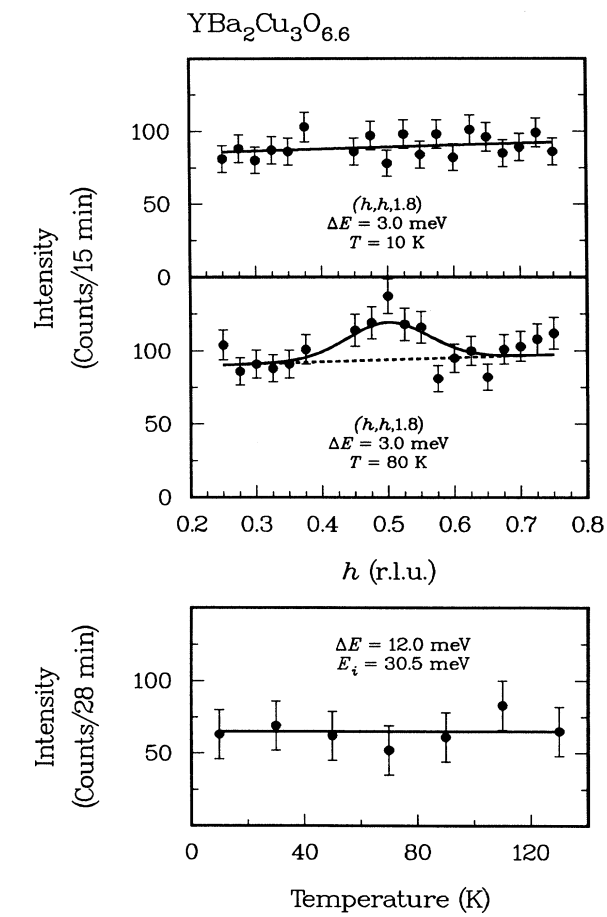

FIG. 4. Temperature dependence of the magnetic correlations at 3 and 12 meV. The 3-meV data are constant-E scans along $(h,h,1.8)$ at 10 and 80 K. The solid lines are best fits to a Gaussian form. Data at 12 meV represent the difference between constant-$\mathbf{Q}$ scans measured at $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},1.8)$ and $(\frac{1}{4},\frac{1}{4},1.8)$. The solid line gives the average intensity.

havior of the spin relaxation rate was first suggested in a neutron-scattering study of $La_{1.85}Sr_{0.15}CuO_4$, however, the low-frequency magnetic cross section in this system was never observed to vanish.[11]

Associating the spin-fluctuation gap with the normal-state behavior of the NMR raises more questions than it answers. If the low-temperature gap develops above T_c, is it related to the superconductivity or is it a normal-state property? Bulut *et al.*[12] found a gaplike depression for integrated inelastic-scattering intensities at low temperature in their RPA analysis of a 2D Hubbard model. In a more recent study, Bulut and Scalapino[6] found a pseudogap in $\chi''(\mathbf{Q},\omega)$ at $\mathbf{Q}=\mathbf{Q}_{\mathrm{AF}}$. By contrast, the data in Fig. 2 appear to indicate a true gap at 10 K in the x=0.6 sample along the entire zone diagonal, not just at the point $\mathbf{Q}=(\frac{1}{2},\frac{1}{2},1.8)$. If the gap observed by neutron scattering is connected to superconductivity, why is it so small and why does it change so rapidly with small changes in T_c? Much more experimental work will be needed before these questions can be resolved.

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