High-energy spin excitations in YBa$_2$Cu$_3$O$_{6.5}$

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Inelastic neutron scattering has been used to obtain a comprehensive description of the absolute dynamical spin susceptibility $\chi''(\mathbf{q},\omega)$ of the underdoped superconducting cuprate YBa$_2$Cu$_3$O$_{6.5}$ ($T_c=52$ K) over a wide range of energies and temperatures (2 meV $\leq \hbar\omega \leq$ 120 meV and 5 K$\leq T \leq$200 K). Spin excitations of two distinct symmetries (even and odd under the exchange of two adjacent CuO$_2$ layers) are observed which exhibit *two different* gaplike features (rather than a single ''spin pseudogap''). The excitations show dispersive behavior at high energies. [S0163-1829(97)51142-8]

The magnetic excitation spectra of doped cuprates contain incisive and highly specific information about theories for high-temperature superconductivity. Many models based on strong Coulomb correlations between charge carriers, for instance, predict that at high energies the spin dynamics should resemble those of the parent antiferromagnetic insulator. Until recently, neutron experiments on metallic and superconducting cuprates were confined to excitation energies below ~ 50 meV, larger than the superconducting energy gap but smaller than the intralayer nearest-neighbor superexchange $J_{||} \sim 100$ meV which sets the energy scale for spin excitations in the undoped antiferromagnetic precursor compounds. Recent pioneering studies of the single-layer superconductor La$_{1.85}$Sr$_{0.15}$CuO$_4$ extended these measurements to higher energies and established the presence of significant spectral weight at energies comparable to $J_{||}$.[1,2] Analogous data for metallic and superconducting YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) have thus far not been reported.

As YBCO is a bilayer system, such experiments can also answer questions about the nature and strength of the coupling between two directly adjacent CuO$_2$ layers, a problem of intense current research. As there are two Cu atoms per unit cell of YBCO, one generally expects the formation of bonding and antibonding electronic states. Conventional band theory predicts the formation of two Fermi surfaces in bands composed of bonding and antibonding states. How-

ever, theoretical[3] as well as experimental[4] evidence indicates that these predictions do not generally hold for low-dimensional systems in the presence of strong correlations. Here we report neutron-scattering measurements that evidence a magnon dispersionlike behavior at energies of the order of $J_{||}$. Moreover, the spin excitation spectrum is characterized by two different gaplike features. These features provide fundamental insights into the ''spin pseudogap'' phenomenon in metallic underdoped cuprates as well as essential information about the interlayer interactions.

Transitions between states of the same type (bonding-to-bonding or antibonding-to-antibonding) and those of opposite types are characterized by even or odd symmetry, respectively, under the exchange of two adjacent CuO$_2$ layers. In the cross section for inelastic magnetic neutron scattering, these transitions can be distinguished according to their distinct dependences on the wave-vector component L perpendicular to the CuO$_2$ sheets:[5,6] The odd component of the spin susceptibility displays a $\sin^2(\pi z_{Cu}L)$ dependence whereas the even component has the complementary L dependence, $\cos^2(\pi z_{Cu}L)$. [Here, $z_{Cu}=0.285$ is the reduced distance between nearest-neighbor Cu spins within one bilayer, and the wave vector $\mathbf{Q}=(H,K,L)$ is measured in units of the reciprocal lattice vectors $2\pi/a \sim 2\pi/b \sim 1.63$ Å^{-1} and $2\pi/c \sim 0.53$ Å^{-1}]. In previous low-energy neutron experiments in the metallic regime,[5-15] *only* spin excitations of odd sym-
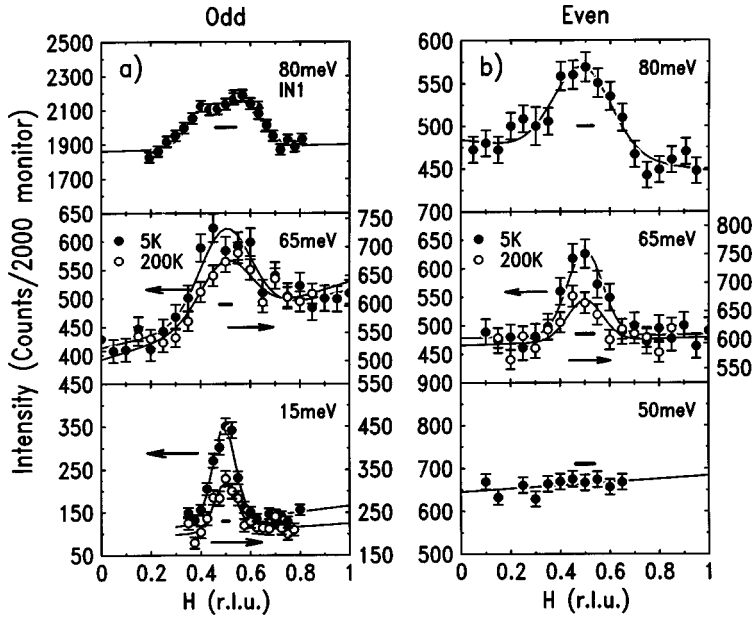
FIG. 1. Constant energy scans of (a) $\chi''_{odd}(\mathbf{Q},\omega)$ through $\mathbf{Q}=\left(\frac{1}{2},\frac{1}{2},L_{odd}\right)$ and (b) $\chi''_{even}(\mathbf{Q},\omega)$ through $\mathbf{Q}=\left(\frac{1}{2},\frac{1}{2},L_{even}\right)$, measured at 5 K (closed circles) and 200 K (open squares). Data have been obtained mostly on IN8 with a final energy of $E_f=35$ meV except at the energy transfer of $\hbar\omega=15$ meV where $E_f=14.7$ meV. The scan for $\hbar\omega=80$ meV has been obtained on IN1 by rocking the sample around $Q=(0.5,0.5,5.4)$ with $E_f=62.6$ meV. Data obtained on IN8 in different counting times have been rescaled to the same monitor (monitor=2000, $\approx$ 6 mn/point). Lines are the results of fits to a single Gaussian on top of a linear background, except at 80 meV where the line corresponds to two identical Gaussian lines displaced symmetrically on each side of $H=0.5$.

metry were found over the entire doping range. Does this reflect a fundamental selection rule, or is there a different (higher) energy scale associated with the first type of transition? In the latter case, a comparison between the energies and susceptibilities associated with the two excitation modes would yield quantitative information on the strength of the bilayer coupling, obviously an important parameter in models of high-temperature superconductivity. This issue also bears directly on the interpretation of other experiments that are sensitive to this coupling. Our measurements of high-energy spin excitations in underdoped metallic $YBa_2Cu_3O_{6.5}$ have much better resolution and counting statistics than the initial studies on $La_{1.85}Sr_{0.15}CuO_4$.[1,2] Excitations of *both* odd *and* even symmetries are observed; the even excitations are characterized by a ~53 meV energy gap, the odd excitations show a gaplike feature at ~23 meV.[16] Both excitations exhibit large temperature dependences, and a dispersionlike behavior at high energies.

Our sample was a high-quality $YBa_2Cu_3O_{6.5}$ single crystal of volume ~2.5 cm^3 and superconducting transition temperature (midpoint) $T_c=52$ K. Its preparation and characterization have been described elsewhere;[13] in particular, susceptibility measurements shown in Fig. 2 of Ref. 13 indicate a sharp superconducting transition (full width 5 K) which rules out significant inhomogeneities in oxygen content. The neutron experiments were performed on the triple axis spectrometers IN8 (installed on a thermal beam) and IN1 (installed on the hot neutron source) at the Institut Laue-Langevin (ILL). The incident beam was monochromated by the (111) reflection of a flat copper crystal on IN8, and by the (200) or (220) reflections of a vertically curved copper crystal on IN1. On both spectrometers, we used a graphite (002) analyzer with fixed vertical and adjustable horizontal curvatures. On IN8, higher-order contamination was eliminated using a pyrolytic graphite (PG) filter on the scattered beam for different fixed final energies, $E_f=14.7$, 30.5, and 35 meV. On IN1, we worked with a fixed final energy of $E_f=62.6$ meV, and a nuclear resonance Er filter was placed on the scattered beam to suppress contaminations from higher energy neutrons. In all the measurements we scanned

the wave vector $\mathbf{Q}$ while keeping the energy transfer constant. Two different scattering geometries were chosen in which wave vectors of the forms $\mathbf{Q}=(H,H,L)$ or $\mathbf{Q}=(3H,H,L)$, respectively, were accessible.[12] The results obtained on both instruments and in both geometries are in good agreement.

Previous experiments[5–15] have established that the low energy magnetic cross section is peaked around the in-plane wave vector $\mathbf{q}_{2D}=(\pi/a,\pi/b)$, or $H=K=\frac{1}{2}$ in reciprocal lattice units (r.l.u.). The cross section for odd spin excitations exhibits maxima at $L_{odd}\approx 1.7+3.5n$ (n = integer), and that for even excitations is maximum for $L_{even}\approx 3.5n$. We therefore performed constant-energy scans along $\mathbf{Q}=(H,H,L_{odd})$ and (H,H,L_{even}), shown in Fig. 1. There are numerous optical phonon modes in the energy range covered by these data. However, scans in different diffraction zones as well as checks against phonon structure factor calculations[12] established the magnetic origin of the peaks. The strong temperature dependence of the peak intensities (see below) is also inconsistent with phonon scattering. This methodology has previously been applied to neutron data at lower energies,[5–7,9–12,14] and the results were found to be consistent with polarized beam experiments wherever such checks proved feasible.[8,12] The temperature dependence of the uniform background presumably arises from multiphonon scattering.

The scans were fitted to Gaussian profiles convoluted with the instrumental resolution function, corrected for the Cu magnetic form factor,[17] and converted to the dynamical spin susceptibility χ'' (Ref. 18) by adjusting for the thermal population factor. Figure 2 shows $\chi''_{odd/even}(\omega)=\int d\mathbf{q}_{2D}\chi''(\mathbf{q}_{2D},L_{odd/even},\omega)/\int d\mathbf{q}_{2D}$, the susceptibility averaged over the two-dimensional Brillouin zone in both odd and even channels. [We assume an isotropic q dependence of the spin susceptibility around (π,π). This assumption is consistent with the data along (H,H) and a more limited data set along $(3H,H)$.] While odd excitations are observed over the entire energy range probed by our experiments, an energy gap of $\Delta_{even}\sim 53$ meV exists for even excitations. Both
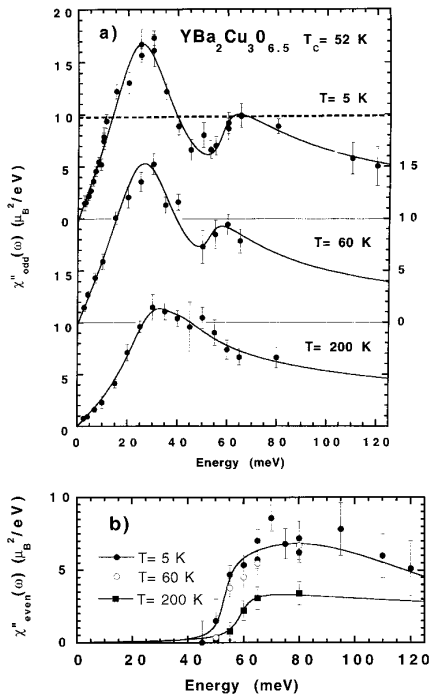
FIG. 2. Odd (a) and even (b) spin susceptibilities at $T=5$ K, $T=60$ K (just above T_c) and $T=200$ K in absolute units (see text). Measurements using different final neutron energies E_f obtained on the two different spectrometers have been rescaled to the same units. The error bars do not include a ~ 30 % normalization error.
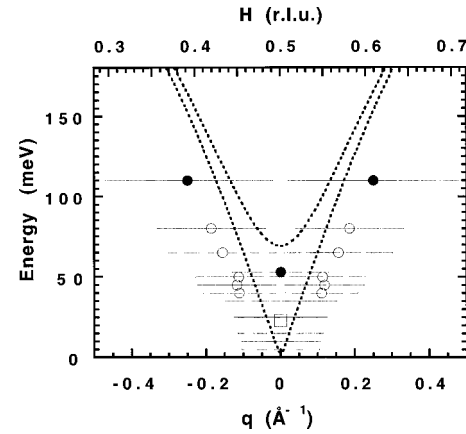

FIG. 3. Spin excitation spectrum for odd (open symbol) and even (closed circles) excitations at 5 K. The open square indicates the energy of the maximum of the odd susceptibility. The horizontal bars represent the full width at half maximum after a Gaussian deconvolution from the spectrometer resolution. The dotted lines correspond to the spin-wave dispersion relation in the insulating antiferromagnetic state with $J_{||}=$ 120 meV and $J_{\perp}/J_{||}=0.08$ (without quantum corrections) (Refs. 21 and 22).

$\chi''_{odd}(\omega)$ at low energies and the gap for $\chi''_{even}(\omega)$ are consistent with previous measurements in this doping regime[5,6,9] which established a lower bound on the gap. Further, previous studies had given the neutron cross section in arbitrary units, but many quantitative models now require an absolute unit scale for χ''. We have therefore calibrated the magnetic intensity against the phonon spectrum, both against acoustic phonons at low energies and against an optical phonon at $\hbar\omega=42.5$ meV according to a procedure discussed elsewhere.[12] Both normalization procedures are in good agreement.

The temperature dependences of both χ''_{odd} and χ''_{even} are striking. At 200 K, $\chi''_{odd}(\omega)$ shows a broad peak around 30 meV which sharpens and shifts to lower energy with decreasing temperature. The ~ 50 meV dip in $\chi''_{odd}(\omega)$ at low temperatures had not been observed before and may be related to the gap in $\chi''_{even}(\omega)$. Much of the temperature evolution of $\chi''_{odd}(\omega)$ takes place in the normal state; however, there is also an additional enhancement of the peak intensity at T_c,[13] which is presumably related to the magnetic resonance peak in the superconducting state dominating the magnetic spectra of more heavily doped samples ($x>0.9$).[6–15] While $\chi''_{odd}(\omega)$ is little affected by temperature above $\hbar\omega \sim 60$ meV, at these energies $\chi''_{even}(\omega)$ increases by almost a factor of 2 between 200 and 5 K. In the same temperature interval Δ_{even} softens from ~ 59 to ~ 53 meV. This parallels the enhancement and softening of the peak in $\chi''_{odd}(\omega)$ at lower energies.

We now turn to the wave-vector dependence of the spin susceptibility. At low energies (below ~ 30 meV), the scans are peaked around $\mathbf{q}_{2D}$ with an intrinsic width of $\Delta\mathbf{q}_{2D}$

~ 0.2 Å^{-1} full width at half maximum. (Previous measurements with better $\mathbf{q}$ resolution had indicated a flat-topped shape of these profiles).[19] Figure 1 shows that at high energies the peak broadens to $\Delta\mathbf{q}_{2D} \sim 0.3$ Å^{-1} and disperses away from $\mathbf{q}_{2D}=\left(\frac{1}{2},\frac{1}{2}\right)$, so that a double-peak structure emerges. For the sake of simplicity, we have fitted the $\mathbf{q}$ scans to the sum of two displaced peaks only when a single peak did not give a satisfactory fit. The positions and intrinsic widths of the peaks in $\chi''_{odd}(\mathbf{q}_{2D},\omega)$ and $\chi''_{even}(\mathbf{q}_{2D},\omega)$ resulting from this procedure are shown in Fig. 3.

It is instructive to compare these data to previous high-energy measurements on the bilayer antiferromagnetic parent compound YBa$_2$Cu$_3$O$_{6.2}$ and on the single-layer, optimally doped compound La$_{1.86}$Sr$_{0.14}$CuO$_4$. Clearly, the basic structure of the spin excitation spectrum of YBa$_2$Cu$_3$O$_{6.5}$ bears some resemblance to the spin-wave spectrum of the YBa$_2$Cu$_3$O$_{6.2}$,[6,17,20–22] with the ungapped odd (gapped even) excitations corresponding to acoustic (optical) spin waves, that is, in-phase (antiphase) precessions of localized spins in adjacent layers. The dynamical susceptibility of the acoustic spin-wave mode is

$$\chi''_{odd}(\mathbf{Q},\omega)=4S\pi Z_{\chi}Z_c\mu_B^2\frac{1+\gamma(\mathbf{q}_{2D})}{\sqrt{1-\gamma^2(\mathbf{q}_{2D})}}\sin^2(\pi z_{Cu}L)$$
$$\times \delta[\hbar\omega-4SZ_cJ_{||}\sqrt{1-\gamma^2(\mathbf{q}_{2D})}], \qquad (1)$$

where $\gamma(\mathbf{q}_{2D})=\frac{1}{2}[\cos(q_x a)+\cos(q_y b)]$, and the quantum corrections for the spin-wave velocity and the spin susceptibility are taken into account, respectively, as $Z_c=1.18$ and $Z_{\chi}=0.51$ following theoretical predictions[23] in agreement with experiment.[12] An analogous equation holds for even excitations, with $\cos^2(\pi z_{Cu}L)$ instead of the $\sin^2$ factor and a dispersion with a gap of 67 meV.[21] The average of Eq. (1) over the 2D Brillouin zone is finite, $4SZ_{\chi}/J_{||}\mu_B^2$, and approximately independent of energy in the energy range probed by our experiment. For comparison this quantity is shown as the dashed line in Fig. 2 (as 10 μ_B^2/eV with $J_{||}=102$ meV after quantum corrections).[17] Clearly, the spectral weights of spin

excitations in antiferromagnetic $YBa_2Cu_3O_{6.2}$ and superconducting $YBa_2Cu_3O_{6.5}$ are comparable. Finally, the spin-wave dispersions of $YBa_2Cu_3O_{6.2}$ are superimposed on the data of Fig. 3. Heuristically, the odd excitation spectrum of $YBa_2Cu_3O_{6.5}$ can be fitted to a dispersive mode with a pseudogap of 23 meV,[16] and a dispersion of ~ 420 meV Å, smaller than the antiferromagnetic spin wave velocity of 650 meV Å.[17,22] However, this analysis requires a very large damping parameter comparable to the gap. This pseudo-gap is characteristic of the normal state; at this doping level, the dynamical susceptibility is only weakly modified by superconductivity. Other features of the spin excitations of $YBa_2Cu_3O_{6.5}$, such as the strong temperature dependence, the pronounced peak-dip structure at low temperatures, and the energy-dependent broadening of the **q** width are also very different from spin waves in $YBa_2Cu_3O_{6.2}$. An explanation of these features presents a challenge to theories of the spin dynamics in the cuprates.[24]

The reports on $La_{1.85}Sr_{0.15}CuO_4$ (Refs. 1 and 2) did not contain information about the temperature evolution of $\chi''(\omega)$, and the measurements did not have sufficient resolution to establish the presence or absence of a dip feature in the spectrum. However, the general shape of 2D **q**-integrated $\chi''(\omega)$ in deeply underdoped $YBa_2Cu_3O_{6+x}$ and optimally doped $La_{2-x}Sr_xCuO_4$ are substantially similar. Despite the different **q** dependences of $\chi''(\mathbf{q}, \omega)$ at low energies (four sharp incommensurate peaks in $La_{1.85}Sr_{0.15}CuO_4$, one broad commensurate peak in $YBa_2Cu_3O_{6.5}$), both spectra are peaked near 25 meV. However, the maximum of χ'' per CuO_2 layer is almost twice as large in absolute units in $YBa_2Cu_3O_{6.5}$ (If given per formula unit, as in Fig. 2, the susceptibility of the bilayer compound exceeds the one of the single-layer compound by about a factor of 4). By contrast, fully oxygenated $YBa_2Cu_3O_7$ has a much smaller normal-state susceptibility and a sharp resonance peak in the superconducting state.[8-12]

In summary, we have reported the observation of the even part of the dynamical spin susceptibility of the bilayer superconductor $YBa_2Cu_3O_{6.5}$, an important part of the experimental description of spin fluctuations in the cuprates. Both the odd and the even components are characterized by strong and unusual temperature dependences and by a dispersive behavior at high energies. Our observation of two different gaplike features in the dynamical spin susceptibility demonstrates that the spin-gap phenomenon, a hallmark of the underdoped cuprates, is more complex than previously thought. Our measurements of the spin correlations in absolute units over a wide range of energies, wave vectors, and temperatures provide an excellent basis for theories of this phenomenon.

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