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BILAYER SPIN DYNAMICS IN UNDERDOPED $YBa_2Cu_3O_{6+x}$

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Abstract—The bilayer structure of $YBa_2Cu_3O_{6+x}$ supports spin excitations that are either even or odd under a symmetry operation that exchanges two directly adjacent CuO_2 layers. Both types of excitation have been observed in underdoped $YBa_2Cu_3O_{6+x}$ by inelastic neutron scattering. Even and odd response functions are split in energy. The temperature evolution in the normal state is parallel in both channels, but different behavior is observed in the superconducting state where a resonant enhancement is observed only in the odd channel. The measurements shed new light on the nature of interlayer interactions in the cuprates. © 1998 Elsevier Science Ltd. All rights reserved

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1. INTRODUCTION

The interaction of adjacent CuO_2 planes in quasi-two dimensional superconducting cuprates is a subject of intense current interest. The theoretical debate centers on the question of whether superconductivity is essentially a two dimensional phenomenon in which interlayer interactions play a marginal role, or whether superconductivity is actually driven by interlayer interactions. In bilayer materials such as $YBa_2Cu_3O_{6+x}$, the two layers interact much more strongly among themselves than with their environment, and the interlayer interactions manifest themselves in a particularly simple form. Specifically, the electronic states centered on the two layers form bonding and antibonding combinations. As discussed in detail elsewhere [1–5], the cross-section for magnetic neutron scattering contains a structure factor that depends on whether the initial and final electronic states are of the same type (both bonding, or both antibonding) or of different types. The former excitations are even and the latter are odd under permutation of the layers.

In the antiferromagnetic parent compound, the nature of the magnetic interaction is well understood. The magnetic excitation spectra are well described by a spin wave theory based on superexchange interactions between localized spins [6–8]. We [9] and others [10] have recently extracted the interlayer superexchange constant $J_\perp$ from the experimentally measured energy gap for optical magnons (i.e. even excitations that involve antiphase precession of nearest-neighbor spins in directly adjacent layers). The result agrees remarkably well with quantitative predictions derived from band structure calculations [11]. Combined with previous results on the low energy, odd sector of the spin excitation spectrum, we have thus arrived at a complete, quantitative description of the magnetic dynamics in the antiferromagnetic regime (at least as far as nearest-neighbor terms in the spin Hamiltonian are concerned; because of the large zone-boundary energies, small next-nearest neighbor interactions are difficult to measure with neutrons).

By contrast, our understanding of the spin dynamics in the metallic and superconducting regimes is still very incomplete. The 'magnetic resonance peak', a remarkably sharp excitation that is present only in the superconducting state, has received much recent attention [12–28]. Its development with temperature and doping has been described in detail in previous reports [2–5, 14–16]. The fact that it carries the structure factor for odd excitations is testimony to the presence of strong bilayer magnetic interactions even in the optimally doped material, although its theoretical description is still a matter of considerable controversy [18–28]. Here we take a broader view and report measurements of both odd and even excitations over a wide range of energies and temperatures. Our experiments are confined to the underdoped regime; only an upper bound can be established on the much weaker normal-state magnetic cross section in

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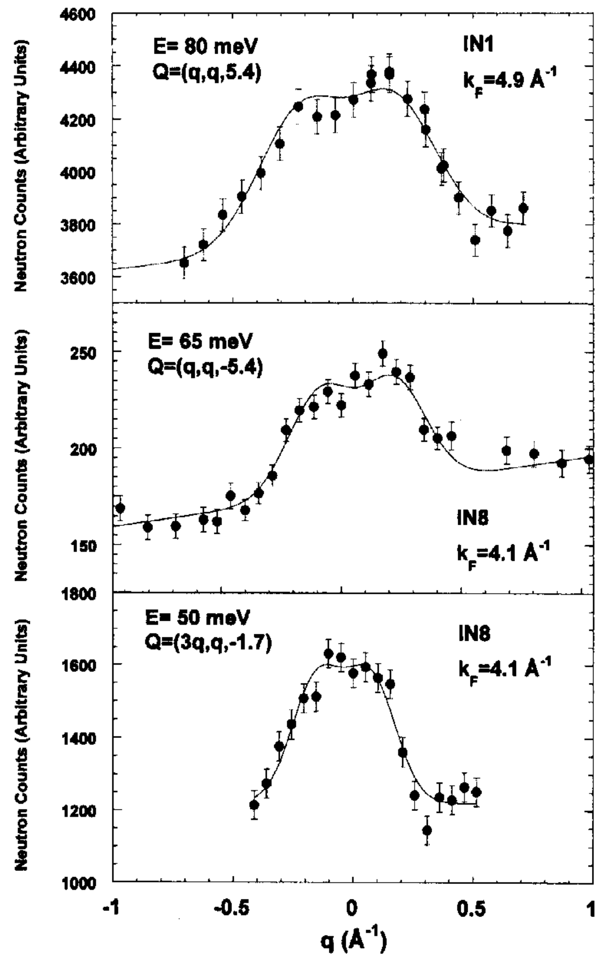

Fig. 1. Constant-energy scans in the odd channel for $YBa_2Cu_3O_{6.5}$, in arbitrary units. The spectrometers and experimental conditions are indicated. The lines are results of fits to two displaced Gaussians, as explained in the text. The reduced wave vector **q** is measured from the $(\pi/a,\pi/a)$ point.

the optimally doped concentration range [2–5]. Previously reported [29] measurements for one hole concentration, $YBa_2Cu_3O_{6.5}$ ($T_c = 52$ K), are combined with new data for a second concentration, $YBa_2Cu_3O_{6.7}$ ($T_c = 67$ K), in order to establish general trends for the doping dependence of the even and odd magnetic cross sections.

The sample preparation and characterization, including evidence for excellent oxygen homogeneity, are described elsewhere [15]. Most of the experiments were performed on 2T and IN8, double focusing triple axis spectrometers at the Laboratoire Léon Brillouin (LLB) and at the Institut Laue-Langevin (ILL), respectively. Earlier experiments were also conducted at the High Flux Beam Reactor at the Brookhaven National Laboratory. Various scattering geometries and spectrometer configurations were used to maximize the magnetic cross section and minimize the contamination from phonon scattering, as described previously [2–5]. The large size of our crystals and the efficiency of the double-focusing spectrometers allow us to measure excitations up to energies of about 80 meV with good counting statistics. Excitations of still higher energies have been studied on IN1 at the ILL.

2. RESULTS AND DISCUSSION

Fig. 1 shows some raw data, taken at fixed excitation energies as a function of wave vector. [The reduced two-dimensional wave vector, **q**, in the figure, is measured from the $(\pi/a,\pi/a)$ point in reciprocal space.] The cross-section is peaked at an incommensurate wave vector, $\mathbf{q}_{max}$, slightly displaced from $(\pi/a,\pi/a)$, although the splitting is always comparable to or smaller than the **q**-width, $\Delta\mathbf{q}$. (Evidence for an incommensurate response was first reported by Sternlieb *et al.* [30] at low energies.) Fig. 1 shows that the splitting increases dramatically with increasing energy but remains comparable to $\Delta\mathbf{q}$ which also increases. The energy dependence of $\mathbf{q}_{max}$ is reminiscent of the spin wave dispersion in the antiferromagnetic regime [29]. Since $\mathbf{q}_{max}$ and $\Delta\mathbf{q}$ are energy dependent, we henceforth quote the imaginary part of the dynamical susceptibility, $\chi''(\mathbf{q},\omega)$, in a **q**-averaged

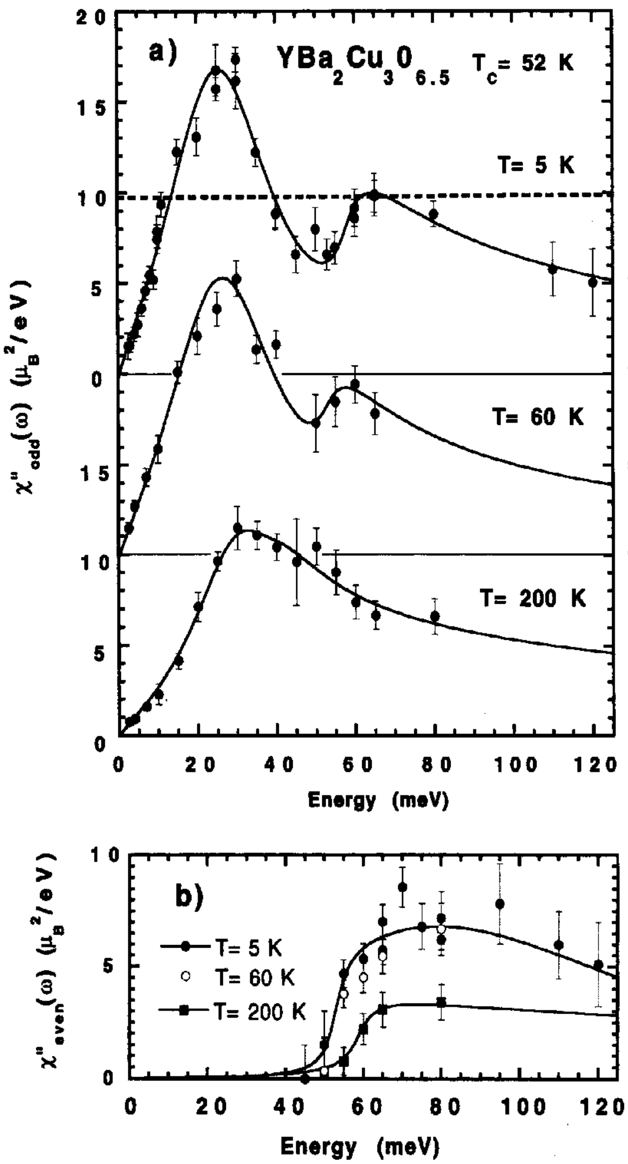

Fig. 2. Energy dependence of the **q**-averaged odd (upper panel) and even (lower panel) dynamical susceptibilities of $YBa_2Cu_3O_{6.5}$, in absolute units [29]. The lines are guides-to-the-eye. In the upper panel, the dashed line is the energy-independent response in the antiferromagnetic regime.

form:

$$\chi''_{2D}(\omega) = \frac{\int d\mathbf{q}\chi''(\mathbf{q},\omega)}{\int d\mathbf{q}} \qquad (1)$$

χ''_{2D} was obtained from the raw data by fitting the constant-energy scans to Gaussian profiles convoluted with the instrumental resolution function, integrating over **q**, and correcting for the Cu magnetic form factor [6, 7] and the thermal population factor. The data were further converted from arbitrary to absolute intensity units by calibrating against phonons whose cross-sections are accurately known [2–5].

Fig. 2 shows the energy dependence of χ''_{2D} for the $YBa_2Cu_3O_{6.5}$ crystal. For $YBa_2Cu_3O_{6.7}$, the basic structure of the spectrum is very similar [31], although the peak susceptibility at high temperatures is about a factor of two smaller. The reduction in maximum intensity with increasing hole concentration agrees qualitatively with trends established previously on the basis of neutron [32, 33] and NMR [24] experiments. For both doping levels, the odd excitations are gapless in the normal state (though the marked decrease in spectral weight for $\hbar\omega \to 0$ has been termed 'pseudo-gap' [8]), but the even excitations have a true energy gap. The gap for even excitations decreases with increasing doping, from about 55 meV in $YBa_2Cu_3O_{6.5}$ [29] to about 40 meV in

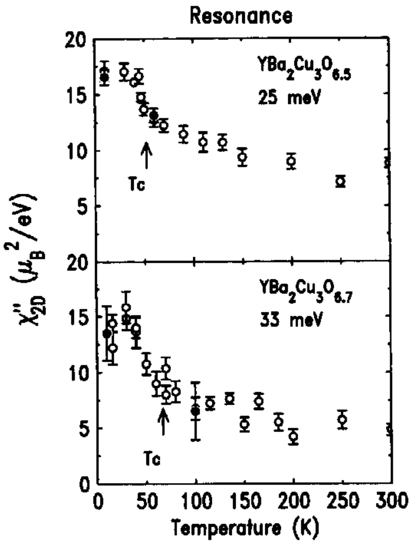

Fig. 3. Temperature dependence of the **q**-averaged odd susceptibilities in $YBa_2Cu_3O_{6.5}$ and $YBa_2Cu_3O_{6.7}$, in absolute units. An ~50% calibration error is not included in the error bars. For each sample, the energy corresponds to the energy of the magnetic resonance peak in the superconducting state. The open circles represent data taken with unpolarized neutrons, the closed circles are data taken with polarized neutrons.

$YBa_2Cu_3O_{6.7}$ at low temperatures [31]. This was anticipated in a phenomenological model of the bilayer coupling [24], but had not been experimentally observed.

Pronounced peaks develop in both even and odd channels as the temperature is lowered. For $YBa_2Cu_3O_{6.5}$, the peak energies are ~30 meV in the odd channel, and ~70 meV in the even channel (Fig. 2). There are two ways to look at these peaks and their doping dependences, depending on whether one chooses to start in the antiferromagnetic phase (x~0) or the optimally superconducting phase (x~1). First, for magnons in the antiferromagnetic phase, $\chi''_{2D}(\omega)$ is actually energy-independent at the energies under study (dashed line in Fig. 2). The pseudo-gap observed in the odd channel of underdoped metallic samples pushes some spectral weight to higher energies, resulting in a peak in $\chi''(\omega)$ whose energy is proportional to the pseudo-gap energy [1, 8, 32, 33]. However, for energies larger than 30 meV, the odd part of $\chi''_{2D}(\omega)$ dips significantly below the antiferromagnetic magnon response; this is not a simple consequence of the opening of a pseudo-gap. For $YBa_2Cu_3O_{6.7}$, $\chi''_{2D}(\omega)$ is even more concentrated in energy [31].

Being concentrated around a single point in both wave vector ($\pi/a,\pi/a$) and energy is the characteristic signature of the magnetic resonance peak which is most pronounced in the optimally doped concentration range. The (broad) normal-state peak in the odd channel may thus be interpreted as a normal-state precursor of the (sharp) magnetic resonance peak in the superconducting state. If this interpretation is correct, its relation to the

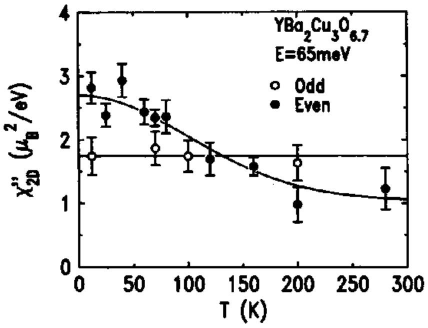

Fig. 4. Temperature dependence of the **q**-averaged even (closed circles) and odd (open circles) susceptibilities in $YBa_2Cu_3O_{6.7}$ at energy 65 meV, in absolute units. (An ~50% calibration error is not included in the error bars.)

normal-state pseudo-gap would be the same as the relation between the superconducting gap and the resonance peak. The temperature dependence of the odd-channel response, shown in Fig. 3, underscores this point. In these plots the magnetic resonance peak manifests itself in a sharp rise of χ''_{2D} at the superconducting transition temperature. As previously noted [15], this enhancement is centered at 25 meV for $YBa_2Cu_3O_{6.5}$, and around 33 meV for $YBa_2Cu_3O_{6.7}$. In both samples, χ''_{2D} already increases substantially when the temperature is lowered from room temperature to T_c. Note, however, that the energy of the normal-state peak [1, 8, 29, 32, 33] appears to be more weakly doping dependent than the energy of the magnetic resonance peak [15–17]. Whichever way one chooses to look at the doping dependence of $\chi''_{2D}(\omega)$, it is clear that there is a smooth crossover from the flat, weakly temperature-dependent response in the antiferromagnetic phase to the highly peaked, markedly temperature-dependent response in the optimally doped superconductor.

Parallel to this behavior in the odd channel, there is a similarly pronounced increase in spectral weight on cooling from room temperature to T_c in the even channel, as shown in Fig. 4 for $YBa_2Cu_3O_{6.7}$. For even excitations, the increase is centered around 65 meV, as compared with ~30 meV for odd excitations. (Note that the odd intensity around 65 meV is only weakly temperature dependent). Further, the even response saturates below T_c, in contrast with the dramatic sharpening below T_c observed in the odd channel (Figs 2 and 3). These data demonstrate explicitly that the magnetic resonance peak occurs exclusively in the odd channel. (Previous experiments on metallic and superconducting $YBa_2Cu_3O_{6+x}$ were confined to lower excitation energies and could thus only probe the odd excitations.) If the resonance energy is indeed directly proportional to $2\Delta_{max}$ (where Δ_{max} is the maximum of the d-wave superconducting energy gap function), as proposed by many models [18–24], the absence of the resonance in the even channel may simply be a consequence of the fact that $2\Delta_{max}$ happens to fall into an energy range in which the even excitations are gapped. However, there may also be more fundamental selection rules at work [25–28].

3. CONCLUSION

In summary, we have investigated the magnetic excitation spectra of $YBa_2Cu_3O_{6+x}$ over a wide range of energies, temperatures and hole concentrations, and we are now beginning to obtain a comprehensive microscopic picture of the spin dynamics in this system. Our data indicate a smooth, seamless crossover from the spin wave spectra of the antiferromagnetic insulator to the sharp resonance in the superconducting state of the optimally doped compound. In the underdoped regime, the energy splitting between the even and odd channel response functions on the one hand, and the different behavior of both response functions below the superconducting T_c on the other hand, must be viewed as consequences of the bilayer coupling. While attempts to explain the latter observation have been made within the framework of most models of the resonance peak, the former observation is new and is only beginning to be addressed by theory. Both observations indicate that interlayer interactions should be an important part of any theoretical description of the spin dynamics and its interplay with superconductivity.

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