Doping Dependence of Bilayer Resonant Spin Excitations in (Y,Ca)Ba$_2$Cu$_3$O$_{6+x}$

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Resonant magnetic modes with odd and even symmetries were studied by inelastic neutron scattering (INS) experiments in the bilayer high-$T_c$ superconductor $Y_{1-x}Ca_xBa_2Cu_3O_{6+y}$ over a wide doping range. The threshold of the spin excitation continuum in the superconducting state, deduced from the energies and spectral weights of both modes, is compared with the superconducting $d$-wave gap, measured on the same samples by electronic Raman scattering in the $B_{1g}$ symmetry. Above a critical doping level of $\delta \simeq 0.19$, both mode energies and the continuum threshold coincide. We find a simple scaling relationship between the characteristic energies and spectral weights of both modes, which indicates that the resonant modes are bound states in the superconducting energy gap, as predicted by the spin-exciton model of the resonant mode.

In high-$T_c$ copper oxides superconductors, inelastic neutron scattering (INS) experiments have shown that the superconducting (SC) phase exhibits an unusual spin triplet excitation, the so-called magnetic resonance mode $\mathbf{q}$. This excitation has been observed in several families of copper oxides with SC critical temperatures $T_c \sim$90 K: $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$, and $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. Two kinds of theoretical models attempt to describe this mode. Within an itinerant-electron picture, it is described as a spin-exciton, that is, it corresponds to a spin-triplet bound states stabilized by the electron-electron interaction in the plane below the electron-hole ($e$-$h$) spin-flip continuum, which is gapped in the SC state $\mathbf{q}$. As the mode is an intrinsic feature of the SC state, it disappears above $T_c$. In other approaches $\mathbf{q}$, the collective spin excitations pre-exist in the normal state, where they are damped by scattering from charge excitations. In the SC state, the damping is suppressed, and the mode sharpens if its energy is below the threshold of the gapped $e$-$h$ continuum. The charge carriers interacting with the spin excitations can be either uniformly distributed $\mathbf{q}$ or segregated into quasi-one-dimensional stripes that separate nearly insulating antiferromagnetic domains $\mathbf{q}$. Although the quantum numbers characterizing the mode are identical in both of these competing approaches, the underlying physical pictures are quite different. It is hence important to establish criteria that allow quantitative experimental tests of these models.

The bilayer structure of YBCO offers an interesting opportunity in this regard. While most calculations have been carried out for a single CuO$_2$ plane per unit cell $\mathbf{q}$, both theoretical approaches predict two distinct magnetic resonant modes in a bilayer system. They are characterized by even ($c$) or odd ($o$) symmetries, respectively, with respect to the exchange interaction between the two layers in a bilayer unit. Recent INS measurements have shown that the bilayer system YBCO indeed exhibits two magnetic resonant modes, both of which are apparent as a strong enhancement of the magnetic intensity at the planar antiferromagnetic wave vector $\mathbf{q}_{\text{AF}} = (\pi, \pi)$ in the SC state $\mathbf{q}$. Their intensities display an order-parameter-like temperature dependence and vanish at $T_c$, without any significant shift of their characteristic energies. A crucial difference between both theoretical approaches concerns the energy-integrated spectral weight (SW) of the magnetic modes. In spin exciton models, this quantity is a function of the binding energy of the modes $\mathbf{q}$ that, in the SC state $\mathbf{q}$, is related to the location of the mode with respect to the $e$-$h$ continuum.

Here we report an INS study of the odd and even resonant spin excitations at $\mathbf{q}_{\text{AF}}$ spanning a wide doping range from the underdoped to the overdoped regimes in YBCO. The threshold for $e$-$h$ excitations, $2\Delta_{\text{max}}$, was measured by electronic Raman scattering (ERS) in the $B_{1g}$ channel $\mathbf{q}$ on the same samples used in the neutron experiments. This eliminates systematic errors invariably associated with comparisons of measurements on samples from different origins. We found that the resonant spin excitations are always located below $2\Delta_{\text{max}}$. Assuming that the spin excitations are bound states in the energy gap of the $e$-$h$ continuum, its threshold at $\mathbf{q}_{\text{AF}}$, $\omega_c$, is estimated using the measured energies, $E_{\text{p}}$, and spectral weights, $W_{\text{p}}$, of the odd and even resonance peaks. The threshold energy $\omega_c$, estimated in this way is found to be below the independently determined gap $2\Delta_{\text{max}}$, so that the analysis is self-consistent. We also observed a systematic scaling relation between $W_{\psi}$ and the reduced binding energy $(\omega_c E_{\text{p}}) / \omega_c$. As such a relation is only expected for collective bound states of the $e$-$h$ continuum, our results support the spin-exciton description of the resonant mode. Further, our study reveals that $2\Delta_{\text{max}}$ and the total SW of the resonant spin excitations at $\mathbf{q}_{\text{AF}}$ drop precipitously in the overdoped regime close to the hole doping level $\delta \simeq 0.19$. Previous work identified...
of this doping level as the end point of the pseudo-gap phase [11, 12].

In addition to previous INS experiments on a slightly overdoped sample (OD85, Y$_{0.9}$Ca$_{0.1}$Ba$_2$Cu$_3$O$_y$, Ref. [7]) and a slightly underdoped sample (UD89), YBa$_2$Cu$_3$O$_{6.85}$, Ref. [8]), INS studies of bilayer excitations were performed on two YBCO samples. The first sample is overdoped (OD75), Y$_{0.85}$Ca$_{0.15}$Ba$_2$Cu$_3$O$_7$ with $T_c = 75$ K [13, 14]. Following our previous work with sample OD85, about 50 single crystals, obtained by a top-seeded solution growth method [15], were co-aligned on aluminum plates and fixed with glue or Al screws. The second sample (UD63), YBa$_2$Cu$_3$O$_{6.6}$ with $T_{\text{onset}}$ = 63 K, is underdoped and consists of 180 detwinned square-shaped single crystals [16]. The mosaic spread of these crystals arrays exceeds 1.4°. INS measurements have been performed on the thermal triple axis spectrometer IN8 at the Institute Laue Langevin (Grenoble). The experimental setup and scattering plane are the same as Ref. [8] except that we utilized a PG(002) monochromator. A fixed final neutron energy of 35 meV has been used yielding a typical energy resolution of 7-8 meV.

The spin susceptibility of a bilayer system reads:

$$\chi(Q, \omega) = \sin^2(\pi z L)\chi_o(q, \omega) + \cos^2(\pi z L)\chi_e(q, \omega)$$

where $Q = (H, K, L)$ is the full wave vector and $q = (H, K)$ is the planar wave vector in CuO$_2$ plane, $z = 0.28$ stands for the reduced distance between the planes of the bilayer. The L-component of the wave vector can thus be used to select either the odd or the even channel [8]: the magnetic structure factor is maximum for $L \sim 5.2$ in the odd channel and $L \sim 3.4$ or 7 in the even. Following the standard terminology [8, 17, 18], we define the magnetic resonant mode as the enhancement of the imaginary part of the magnetic susceptibility, $\Delta m\chi(q_{AF}, \omega)$ in the SC state, which is derived from the difference between measurements in the SC state at 10 K and the normal state above $T_c$. The imaginary part of the dynamical magnetic susceptibility is also calibrated in absolute units ($\mu_0^2$ eV$^{-1}$/f.u.) following a standard procedure with a reference phonon at 42.5 meV [17, 18].

Fig. 1a provides evidence of the existence of odd and even resonant magnetic peaks in the INS spectra of the highly overdoped sample OD75. These results differ significantly from previous reports at lower doping levels [8, 18], because the energies of both mode are very similar: $E_{\text{e}}^o = 34$ meV and $E_{\text{e}}^e = 35$ meV. Further, both modes also display similar amplitudes. In order to determine the amplitudes $I_{\text{o}}^o$ of both modes, the $L$-dependence of the magnetic intensity at 34 meV (Fig. 1b) was fitted with Eq. 1. This yields a ratio $I_{\text{o}}^o/I_{\text{o}}^e \approx 0.4$, compared to 1/3 for sample OD85 [7] and 1/6 for YBCO$_{6.85}$ [8]. However, as for lower doping levels [7, 8], the temperature dependence of the intensities of both modes exhibits a marked change at the SC temperature (Fig. 1c), indicating that the phenomena have a similar origin. In the strongly underdoped sample UD63 (Fig. 1d), the two mode energies and their intensities are very different: $E_{\text{e}}^o = 37$ meV, $E_{\text{e}}^e = 55$ meV, and $I_{\text{o}}^o/I_{\text{o}}^e \approx 1/20$. As already observed in the OD85 sample [7], the odd mode exhibits a broader energy line shape in OD75, $\Delta E \approx 12$ meV, than the resolution-limited energy profile observed in underdoped samples [10]. The energy width of the even mode, $\Delta E \approx 15$ meV, is always broader than the resolution, for all doping level.

In order to assess whether or not these resonant modes should be regarded as bound states in the electronic continuum, one has to determine the amplitude of the SC gap. The good surface quality of the single crystals removed from the crystals array OD85 and OD75 enables ERS experiments. The ERS technique probes electronic excitations in selected areas of the Brillouin zone. In the $\text{B}_{1g}$ channel, the antinodal areas around $(\pm \pi, 0)$ and $(0, \pm \pi)$ are probed, and a peak appears in the electronic spectrum in the SC state at twice the energy of the maximum $d$-wave SC gap, $2\Delta_{\text{max}}$ [8]. These measurements were carried out with a triple-grating spectrometer in quasi-back-scattering geometry. The crystals were mounted on the cold finger of an He circulation cryostat.
The 514 nm excitation line of a Ar⁺/Kr⁺ mixed-gas ion laser was used.

Figure 2 shows the B₁g peaks in the ERS spectra of single crystals extracted from samples OD85 and OD75. The B₁g peak is observed at 69.3 meV in the first sample and shifts to 35.5 meV for the second, whereas T_c is only reduced from 85.5 K to 75 K. The same results were obtained on other crystals selected randomly from the OD85 and OD75 arrays [21]. Further experiments on samples with different T_c also confirm that the characteristic energy of the B₁g peak drops steeply by almost a factor two with a minor reduction of T_c [22]. As the information about the mode energy obtained from tunneling is quite indirect, the interpretation of these data has remained controversial. The total SW of the resonant modes, \( \Delta \), in \( \delta \), in the present study (\( \gamma \)), is nearly constant in the SC state and at 90 K in the normal state is displayed. In addition to sharp features due to phonon renormalization, the difference indicates a broad peak due to the enhancement of the electronic response in the SC state. The electronic peaks were fitted with Gaussian profiles. The inset shows the peak energy of the 5 samples we have studied (see Fig. 2 caption for the determination of the doping levels).

The coincidence of the resonant mode energy and the SC energy gap in the overdoped range was suggested by prior tunneling experiments [22]. As the information about the mode energy obtained from tunneling is quite indirect, the interpretation of these data has remained controversial. The total SW of the resonant modes, \( W_\omega + W_\omega \), declines precipitously around a similar doping level \( \delta \) ≃ 0.19.

In Fig. 2a, we report the hole doping dependence (i) of the of 2\( \Delta_{\text{max}} \), determined by ERS from the present study and Refs. [12, 20], and (ii) the odd and even resonant energies obtained from INS data. For all samples, the even resonance appears at higher energy and with less intensity than the odd resonance. While \( E^e_\omega \) evolves as a function of hole doping as \( \sim 5k_B T_c \) (Fig. 2a), \( E^e_\omega \) does not scale with \( T_c \); it remains almost constant in the underdoped regime and begins to decrease upon entering the overdoped regime. With increasing hole doping, the distance in energy between the magnetic modes decreases from 17 meV in UD63 to 1 meV in OD75. Owing to the differences between the energy lineshapes of the odd and even modes, the most meaningful comparison is based on their energy-integrated spectral weights: \( W^{\alpha,e}_{\omega} = \int_0^\infty d\omega \Delta f_{\alpha,e}(\mathbf{q}, \omega) \). The ratio and the sum of the energy-integrated SWs of both modes are shown in Fig. 3b. Obviously, \( W^{\alpha}_{\omega} + W^{e}_{\omega} \) increases with increasing doping.

The dramatic change of the SC gap measured by ERS (Fig. 3a) separates two distinct regimes on both sides of \( \delta_c \): in regime I, encompassing the underdoped samples up to OD85, one obtains the following hierarchy: 5\( k_B T_c \sim E^e_\omega < E^e_\omega < 2\Delta_{\text{max}} \). In regime II (OD75), all energies collapse to 5\( k_B T_c \sim E^e_\omega \simeq E^e_\omega \simeq 2\Delta_{\text{max}} \). The coincidence of the resonant mode energy and the SC energy gap in the overdoped range was suggested by prior tunneling experiments [22]. As the information about the mode energy obtained from tunneling is quite indirect, the interpretation of these data has remained controversial. The total SW of the resonant modes, \( W^e_\omega + W^e_\omega \), declines precipitously around a similar doping level (Fig. 3b). It is interesting to note that the doping level \( \delta \) ≃ 0.19 separating these two regimes coincides with the hole concentration where previous work had uncovered rapid doping-induced modifications of the physical properties such as resistivity, specific heat, Knight shift, and muon spin relaxation rate [10, 11, 12]. According to [22], this point in the phase diagram is a quantum critical point associated with the doping-induced disappearance of a putative order parameter characterizing the pseudo-gap phase. Therefore, it is tempting to relate the anomalous hole doping dependencies shown in Fig. 3a to...
the pseudo-gap phenomenon.

In Fig. 3a, one also notices that the resonance energies are systematically lower than twice the SC gap. This indicates that the resonant modes are below the continuum threshold at $q_{AF}$, $\omega_c$, which can be estimated as $1.5\Delta_{\text{max}}$ using the typical Fermi surface topology observed in cuprates [24]. As discussed in Refs. [25], another estimate of $\omega_c$ can be made within the spin-exciton model where $W_{r,e} \propto (\omega_c - E_{r,e})/\omega_c$. $\omega_c$ is simply deduced by the ratio of the SWs of the odd and even modes [2, 3]. For the four samples, $\omega_c$ extracted in this way from INS (Fig. 3a) agrees with the value deduced from ERS data.

We now discuss these observations in the light of the various models attempting to describe the resonant modes [2, 3, 4, 5, 6]. As sketched in Fig. 4, one can envisage two situations: either the modes are bound states in the energy gap of the electronic continuum (Fig. 4b), as assumed in the spin-exciton model [2, 3, 4, 5], or they already exist above $T_c$ [2, 3, 4, 5, 6, 7] and their SWs are only enhanced in the SC state (Fig. 4c). In the former case, the SWs of the modes are proportional to each other with a constant scaling factor over a wide doping range. This is the expected behavior for collective bound states in the SC energy gap where the scaling factor is mostly controlled by doping independent microscopic parameters. In the pre-existing mode picture, on the other hand, the SW ratio $W_r/W_c$ would be also determined by the spin dynamics in the normal state. To be sure, well-defined magnetic modes are observed in the underdoped samples above $T_c$ [17], but we recently found that there are a number of qualitative differences between the excitation spectra in the SC and normal states at energies comparable to the SC gap [27]. In overdoped samples, normal-state excitations appear to be heavily overdamped and have thus far not been clearly identified. It is therefore reasonable to consider the excitations in the SC state separately, as we have done.

In summary, odd and even symmetry resonant magnetic excitations have been observed in bilayer YBCO over a wide doping range. A detailed analysis of the modes energies and spectral weights show that the resonant mode in the high-$T_c$ cuprates arises from bound states in the superconducting energy gap. Our results have no natural explanation in models where the resonant excitations are associated with excitations pre-existing in the normal state.

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