Magnetic Resonance, Electronic Spectra and Bilayer Splitting in Underdoped Bi2212

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We report the first inelastic neutron scattering (INS) experiments of the resonance in the underdoped regime of Bi2Sr2CaCu2O8+δ. The energy of the resonance is found to be 34 meV and the temperature dependence shows a smooth evolution through Tc, with a remnant persisting in the pseudogap state. Besides the INS data, we present also angle resolved photoemission (ARPES) spectra taken on the same crystals. As a function of temperature, the neutron intensity scales with the square of the ARPES gap estimated by the leading-edge midpoint at (π, 0). We also show that the energy of the collective mode inferred from the peak-dip-hump structure compares well with the energy of the resonance. Based on kinematics, we argue that the correlation of these two energies is consistent with the absence of bilayer splitting in the electronic dispersion.

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It is now widely accepted that strong electronic correlations are responsible for many unusual properties of the high temperature cuprate superconductors (HTSC) 1,2. Evidence is provided by momentum resolved spectroscopies such as inelastic neutron scattering (INS) and angle resolved photoemission spectroscopy (ARPES), which probe the fundamental excitations connected with the spin and single particle response functions.

In the normal state of underdoped HTSC, no quasiparticles can be observed 3 suggesting unusually strong electronic correlations. The situation changes drastically below Tc, since well defined quasiparticles can be observed at each point of the Fermi surface 4. In particular at the (π, 0) point, the low-temperature spectral function is characterized by a peak-dip-hump structure which is believed to be due to the interaction of the electrons with an electronic collective mode 5.

Strong correlations are also inferred from INS since, unlike in weakly correlated metals, spin excitations could be measured in La2−xSrxCuO4 6,7. YBa2Cu3Ox 8 (YBCO), and recently in optimally and overdoped Bi2Sr2CaCu2O8+δ 9 (Bi2212). While for the optimally and overdoped YBCO and Bi2212 samples, a sharp resonance can be observed only below the critical temperature Tc, data obtained in the underdoped regime of YBCO suggest that a broadened remnant persists above Tc.

The question of the origin of the resonance has been widely addressed and several models involving a spin exciton in the particle-hole channel, a collective mode in the particle-particle channel, or antiferromagnetic domains due to formation of stripes 10,11 have been proposed. In order to establish whether or not the magnetic resonance is relevant to the origin of high temperature superconductivity, quantitative comparisons of experimental results obtained by different techniques are very useful. An example is given by the attempt to compare the weight of the resonance mode and the superconducting condensation energy in YBCO 12. In a related vein, we have recently shown that the doping dependence of the collective mode energy inferred from ARPES in Bi2212 compares favorably with the resonance’s energy measured by INS in YBCO 13. So far, a direct comparison between ARPES and INS measurements in the underdoped regime of Bi2212 could not be realized since all INS data in that doping range have been obtained only on YBCO.

We report in this paper the first INS measurement of the resonance in underdoped (Tc = 70 K) Bi2212. In parallel, we have performed temperature-dependent ARPES measurements of the spectral function at the (π, 0) point on the same sample.

The INS measurements were performed on the triple-axes spectrometer IN8 at the Institut Laue-Langevin in Grenoble, France. We used a vertically focused Cu(111)-monochromator and a vertically and horizontally focused PG(002)-analyser with 35 meV fixed final energy. The wave vector Q is given in units of (1/d∥, 1/d⊥, 1/d⊥), where d∥ is the Cu-Cu in-plane distances and d⊥ is the distance between two adjacent Cu-O planes.

The sample was built of an alignment of five crystals with an overall volume of about 80 mm3 and overall mo-
sisity < 3°. All crystals were grown from the same batch and showed a critical temperature \( T_c = 70 \text{K} \) and a transition width of 2-4K (see insert of Fig. 1).

Fig. 1a shows energy scans at \( \mathbf{Q} = (\pi, \pi, -3\pi) \) taken at \( T = 10 \text{K} \) and \( T = 250 \text{K} \). As it is known from phonon density-of-state measurements [12], the spectra, for energies \( 20 < E < 40 \text{meV} \), are contaminated by phonon branches whose intensities, following Bose population factors \( B(T, E) = (1 - \exp ( -E/k_B T ))^{-1} \), will increase with increasing temperature \( T \). In order to separate out the lattice contribution from the magnetic one, and knowing that phonons are strongest around 20 meV, we normalize the data at 24 meV, after subtraction of a flat background [13]. We observe that at low temperature, a clear signal emerges between 30 and 40 meV. A fit to the difference of the low- and high-temperature data shows that the peak is centered at 34 \( \pm \) 2 meV (Fig. 1b) [14]. This value agrees with the energy of the resonance measured in underdoped YBCO of similar doping level [8].

The magnetic origin of the peak is further confirmed by the observation of the characteristic modulations of the resonance’s intensity along the in- (Fig. 2b) and out-of-plane directions (Fig. 2a), confirming its \((\pi, \pi, \pi)\) (odd) symmetry [14]. The odd symmetry along the \( z \)-direction will turn out to be crucial for an understanding of the ARPES spectra. The in-plane \( Q \)-width of the resonance is 0.59 \( \pm \) 0.07 \( \AA^{-1} \) at low temperature, a value comparable to that measured for optimally doped Bi2212 [11] but about twice as large as the low-temperature value measured in underdoped YBCO [17].

As the temperature is increased, the energy-integrated weight of the neutron signal (see Fig. 3) is found to weaken regularly, giving rise to a remnant far into the pseudogap state, similar to what has been observed in underdoped YBCO [18]. Notice that no break at \( T_c \) can be resolved.

We now turn to the ARPES data obtained on three portions of the crystals used for the INS experiments. The experiments were performed at the Synchrotron Radiation Center, Wisconsin, using a high-resolution 4-meter normal incidence monochromator, with a resolving power of \( 10^4 \) at \( 10^{11} \) photons/sec. We used 22 eV photons, with a 20 meV (FWHM) energy resolution, and a momentum window of radius 0.045\( \pi \).

Fig. 4a shows ARPES data taken at \((\pi, 0)\) as a function of temperature. The low temperature spectra consist of a sharp spectral peak, followed at higher binding energy by a pronounced spectral dip, then by a broad maximum (the hump). This structure evolves with temperature into a single broad peak with a leading edge gap (the pseudogap). For binding energies above the hump energy, the spectra are temperature independent. Below, we will comment further on the temperature evolution, but for now, we will try to make a correlation of the energy scales apparent in the low temperature ARPES spectra with the INS resonance energy. Fig. 4b shows an expanded view of the near-\( E_F \) region and we find that the peak- (=\( \Delta_0 \), dip- and hump-energies are 48\( \pm \) 2, 80\( \pm \) 2 and 193\( \pm \) 5 meV, respectively, characteristic of underdoped Bi2212 samples [2]. \( \Delta_0 \) represents the maximum of the \( d \)-wave superconducting gap.

It has been proposed that the peak-dip-hump structure is caused by the interaction of the electrons with a collective mode whose energy \( \Omega < 2\Delta_0 \). This can be understood in the following way: for quasi-two dimensional systems and within the impulse approximation, ARPES measures the \( \mathbf{k} \) and \( \omega \) dependence of the single particle spectral function [18]. For binding energies \( \omega > \Delta_0 + \Omega \), the inelastic scattering process by the collective mode is forbidden since the final state of the electrons would lie in the gap. As a consequence the lifetime of the quasiparticles is long which gives rise to the sharp peak in the ARPES spectra. For binding energies \( \omega > \Delta_0 + \Omega \), a relaxation channel opens and the lifetime drops, creating a spectral dip. The dip is further enhanced by a maximum of \( \Sigma'^{\prime} \) at \( \omega = \Delta_0 + \Omega \) which is required by the Kramers-Kronig relations for the self-energy [3]. It is then easy to realize that the energy of the mode can be directly inferred from the ARPES data by measuring the energy difference between the dip and the peak. For this underdoped sample we obtain 32 \( \pm \) 4 meV for the mode energy, which is in excellent agreement with the value of 34 \( \pm \) 2 meV obtained for the resonance from INS (see Fig. 1b). In order to illustrate this correspondence between the ARPES collective mode and the INS resonance, we have also drawn, in Fig. 4b, the magnetic resonance (bold-broken line), with its energy-axis shifted by 48 meV, the peak energy.

We now return to the evolution of the ARPES line-shape as a function of temperature (Fig. 4a). As the temperature is raised, the quasiparticle peak smears out, and the spectral dip fills in, leading eventually to a single broad peak with a leading edge gap [19]. Fig. 3b shows (open symbols) the temperature evolution of the square of the gap estimated by the leading-edge midpoint (normalized to the \( T = 50 \text{K} \) value). Below \( T_c \), it remains constant and then decreases substantially above \( T_c \). This temperature evolution is very similar to that of the neutron intensity, which we also plot (closed symbols). The neutron data points were obtained from Fig. 3a by subtracting the \( T = 250 \text{K} \) value.

We remark that the correlation we see in Fig. 3b was first suggested by Demler and Zhang based on a particle-particle interpretation for the neutron resonance [1]. In such a picture, the neutron resonance energy should not depend on temperature (which we also observe), but its intensity should scale with the square of superconducting energy gap. This scaling has been verified for the optimally- to over-doped region of the phase diagram of YBCO [1] and Bi2212 [4]. For our underdoped sample, it is interesting to note that, within the error bars, we also find that this scaling persists in the pseudogap phase.
This is strong evidence that the leading edge pseudogap is a consequence of pairing correlations [23]. A similar conclusion has been reached recently by a neutron scattering study of the field dependence of the resonance [2].

We finally discuss the implication of our results for the question of bilayer splitting below $T_c$ [22]. On general grounds we expect that the two CuO$_2$ layers hybridize to form non-degenerate bonding (B) and antibonding (A) states. We now argue that, for the observed [23] hole-like Fermi surface, only a small and most likely vanishing bilayer splitting between the A and B states is consistent with the kinematics shown in Fig. 5. If the two states were split the quasiparticle peaks for the two would be at the corresponding gaps $\Delta_A$ and $\Delta_B$. Because of the ($\pi, \pi, \pi$) symmetry of the resonance, scattering by the resonance implies both in- and out-of-plane wavevector transfers. The in-plane component connects the ($\pi, 0$) and ($0, \pi$) points while the $Q_x = \pi$ out-of-plane component connects bands A and B as shown in Fig. 5. Thus the threshold for decay, i.e., the dip energy, at $k = (\pi, 0)$ via the mode of energy $\Omega$ is at $\Delta_B + \Omega$ for the A state, and $\Delta_A + \Omega$ for the B state. It follows that the ARPES spectra consist of a weighted superposition of bands A and B, and that the measured peak and dip positions are averaged values of the positions in both channels. If there were significant bilayer splitting, the peak would be broadened (or even duplicated) and the dip less sharply defined. Furthermore, the averaged peak-dip difference is equal to $\Omega$+bilayer splitting. Since we obtained 32 ± 4 meV and 34 ± 2 meV for the peak-dip and INS resonance energies, respectively, we can put an upper bound of 8 meV for the bilayer splitting of the underdoped sample studied here. This is in agreement with all known ARPES experiments on Bi2212 since, even with resolutions as high as 10 meV, no bilayer splitting could be detected [24].

In conclusion we have shown, performing both INS and ARPES experiments on the same crystals, that the energy of the mode inferred from the ARPES spectra agrees well with the energy of the INS magnetic resonance. It is also found that a magnetic remnant persists well into the pseudogap state, whose intensity as a function of temperature scales with the leading-edge gap measured at (\pi, 0) from ARPES. We have also argued that the correspondence of the ARPES peak-dip separation with the INS resonance energy implies that the bilayer splitting in the electronic dispersion must be very small.

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[1] P. W. Anderson, Science 256, 1526 (1992).
[2] J. C. Campuzano et al., Phys. Rev. Lett. 83, 3709 (1999).
[3] A. Kaminski et al., Phys. Rev. Lett. 84, 1788 (2000).
[4] M. R. Norman et al., Phys. Rev. Lett. 79, 3506 (1997); M. R. Norman and H. Ding, Phys. Rev. B 57, R11089 (1998).
[5] For a review see, M. A. Kastner et al., Rev. Mod. Phys. 70, 897 (1998).
[6] For a review see, P. Bourges et al. in the Gap Symmetry and Fluctuations in High Temperature Superconductors Edited by J. Bok, G. Deutscher, D. Pavuna and S. A. Wolf (Plenum Press, 1998) p. 349 [cond-mat/9901333].
[7] H. A. Mook et al., Phys. Rev. Lett. 70, 3490 (1993); H. F. Fong et al., Phys. Rev. Lett. 75, 316 (1995).
[8] H. F. Fong et al., Phys. Rev. Lett. 78, 713 (1997); P. Dai et al., Phys. Rev. Lett. 80, 1758 (1998).
[9] H. F. Fong et al., Nature 398, 588 (1999); H. He et al., Phys. Rev. Lett. 86, 1610 (2001).
[10] D. L. Liu, Y. Zha and K. Levin, Phys. Rev. Lett. 75, 4130 (1995); I. I. Mazin and V. M. Yakovenko, Phys. Rev. Lett. 75, 4134 (1995).
[11] E. Demler and S. C. Zhang, Phys. Rev. Lett. 75, 4126 (1995).
[12] V. J. Emery and S. A. Kivelson, Physica C 209, 597 (1993).
[13] P. Dai et al., Science 284, 1344 (1999).
[14] L. Pintschovius and W. Reichhardt in Neutron Scattering in Layered Copper-Oxide Superconductors Edited by A. Furrer (Kluwer Academic Publishers, 1998) p. 165.
[15] Although the Bose population factor $B(T, E)$ strongly depends on the energy $E$, $B(T_1, E)/B(T_0, E)$, is only weakly E-dependent for $20 < E < 50$meV and the temperature range considered.
[16] Exploiting the Q-dependence of the Cu-magnetic form factor, we have obtained similar plots by comparing scans taken at equivalent points in various Brillouin zones.
[17] Ph. Bourges et al., Science 288, 1234 (2000).
[18] M. Randeria et al., Phys. Rev. Lett. 74, 4951 (1995).
[19] For technical reasons we were limited to temperatures below 110 K. However, it is known from previous measurements [2] that for underdoped $T_c=70$ K, the pseudogap remains up to a temperature $T^*$ about 300K.
[20] M. Randeria, in Proceedings of the International School of Physics “Enrico Fermi” Course CXXXVI edited by G. Iadonisi, J. R. Schrieffer, and M. L. Chiafalo, (IOS Press, 1998), p. 53; cond-mat/9710223.
[21] P. Dai et al., Nature 406, 965 (2000).
[22] H. Ding et al., Phys. Rev. Lett. 76, 1533 (1996).
[23] H. M. Fretwell et al., Phys. Rev. Lett. 84, 4449 (2000).
[24] A. Kaminski et al., Phys. Rev. Lett. 86, 1070 (2001).
FIG. 1. a) Energy spectra of the neutron intensities at wave vector $\mathbf{Q} = (\pi, \pi, -3\pi)$ at temperatures $T = 10\, \text{K}$ (open squares) and $T = 250\, \text{K}$ (filled diamonds). The spectra have been normalized according to the procedure described in the text. Insert: magnetization curve of one of the Bi2212-crystals used in both the INS and ARPES experiments. The dashed line indicates $T_c$. b) Difference spectrum of the normalised data shown in Fig. 1a. The solid line represents a Gaussian fit to the data.

FIG. 2. a) Difference of neutron intensities ($I(\pi, \pi, Q_z) - I(0.4\pi, 0.4\pi, Q_z)$) of Q-Scans perpendicular to the CuO$_2$ planes at 34 meV and $T = 10\, \text{K}$. The solid line is a $\sin^2(Q_z/2)$ function describing the odd character of the resonance along the z-direction. b) Neutron intensities of Q-scans $I(Q, Q_z = -3\pi)$ parallel to the $(\pi, \pi)$ direction at 34 meV, $T = 10\, \text{K}$. The background measured at $Q_z = -2.2\pi$ has been subtracted.

FIG. 3. a) Temperature dependence of the energy-integrated intensity of the neutron signal at $(\pi, \pi, -3\pi)$. b) Temperature dependence of the square of the ARPES gap estimated by the leading-edge midpoint (open triangles) and of the neutron intensities (filled triangles). All data have been normalized to the $T = 50\, \text{K}$ values. For the neutron data, the high temperature ($T = 250\, \text{K}$) value has been subtracted. The dashed lines indicate $T_c$.

FIG. 4. a) Temperature dependence of the ARPES spectra taken at the $(\pi, 0)$ point. b) Expansion of the near $E_F$ region at $T = 40\, \text{K}$ together with the peak, dip, hump and mode energies. Also shown (bold-broken line) is the magnetic resonance as measured by INS. An offset of 48 meV ($=\Delta_0$), has been added to the energy axis of the INS data.
FIG. 5. Dispersions of the bonding (solid line B) and anti-bonding (broken line A) bands assuming a non-vanishing bilayer splitting. Also shown are the \((\pi, \pi, \pi)\) wavevector connecting portions of the Fermi surface around the \((\pi, 0)\) and \((0, \pi)\) points in both bands. The interaction diagram of the electron with the magnetic resonance is shown on the top.