Momentum, Temperature, and Doping Dependence of Photoemission Lineshape and Implications for the Nature of the Pairing Potential in High-T_c Superconducting Materials

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The anomalous momentum and temperature dependence of the spectral lineshape in data from underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) indicates that the quasiparticles are strongly coupled to collective excitations centered near $\mathbf{Q} = (\pi, \pi)$. The doping dependence of the spectral lineshape and its correlation with the size of the superconducting gap indicate these collective excitations are related to the pairing interaction in high-T_c superconductors, in analogy with phonon induced structures in tunneling spectra of low T_c materials. [S0031-9007(97)02505-2]

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Over the last decade, angle-resolved photoemission spectroscopy (ARPES) has contributed significantly to the understanding of the high-T_c superconductors [1]. The most recent example of this is the observation of an anisotropic excitation pseudogap in the normal state of underdoped Bi2212 [2–4]. Despite the progress, the line shape of photoemission spectra remains poorly understood. This is particularly true for the underdoped materials. In this paper, we analyze the momentum, temperature, and doping dependence of the spectral line shape, with emphasis on a broad feature at 100–200 meV that is always present in spectra taken near $(\pi, 0)$ in underdoped samples. This feature is absent in spectra taken at the Fermi surface along the $(0, 0)$ to (π, π) direction in underdoped samples and along both of these directions in overdoped materials. We argue that these results are due to a strong dressing of the photohole for $\mathbf{k} \cong (\pi, 0)$ in the underdoped material, while the dressing is modest in the other cases. This result is consistent with the hole coupling strongly to collective modes of momentum $\mathbf{q}$ whose spectral function $\chi''(\mathbf{q}, \omega)$ peaks near $\mathbf{Q} = (\pi, \pi)$ for the underdoped case, while the coupling is weak because $\chi''(\mathbf{q}, \omega)$ is weak and broad in momentum for the overdoped case. The doping dependence of the ARPES spectrum and the superconducting gap suggests that these collective excitations are related to the pairing potential with momentum dependence consistent with d pairing symmetry [5].

Angle-resolved photoemission data were obtained using a Vacuum Science Workshop chamber attached to the beam line V-3 of the Stanford Synchrotron Radiation Laboratory (SSRL) and a Scienta-200 analyzer using a He discharge lamp. Details of the experimental setup and the sample preparation are the same as those reported earlier [3]. The energy resolution of data discussed here is either 35 or 20 meV as specified later, and the momentum resolution is ± 1 degree. Doping of the sample is accomplished through oxygen content adjustment by annealing under various conditions. The notation of underdoping (overdoping) represents smaller (larger) hole concentration than those needed to achieve maximum T_c.

Figure 1 shows typical normal state photoemission spectra recorded at 100 K from an underdoped and an overdoped Bi2212 sample with T_c of 84 and 80 K, respectively. (α) and (β) represent spectra taken at the Fermi surface (FS) crossing along the $(0, 0)$ to (π, π) direction and the $(\pi, 0)$ to (π, π) directions, respectively. The spectra from the overdoped sample show Lorentzian-like peaks at the Fermi level. The spectrum from the underdoped sample recorded at (α) is very similar to that from the overdoped sample. In contrast, the spectrum at (β) is dramatically different. It is an edgelike structure with a very broad maximum near 100–200 meV. Furthermore, the leading edge is pulled back from the Fermi level by about 20 meV, reflecting the reported $d_{x^2-y^2}$ like normal state excitation pseudogap [2–4].

The difference between the spectra at (β) from the underdoped and overdoped samples is generic. In the

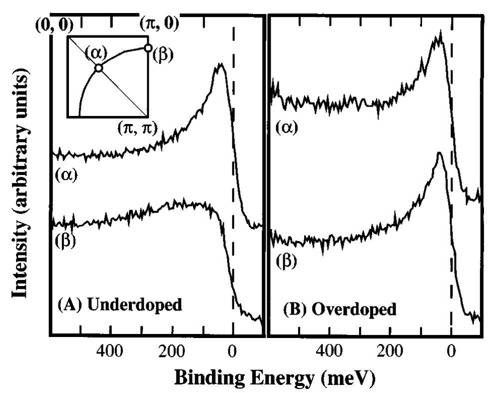

FIG. 1. ARPES data from an underdoped Bi2212 (A) and an overdoped Bi2212 (B) sample with an energy resolution of 35 meV. The inset shows k-space locations where the spectra were recorded. The curve depicts the experimental Fermi surface. (α) and (β) denote the Fermi surface points along the $(0, 0)$ to (π, π) and $(\pi, 0)$ to (π, π) directions, respectively. (β) is very close to $(\pi, 0)$.

underdoped regime, the spectra are very insensitive to doping, although the broad maximum may shift with doping level and the details of the line shape may show small variations. Once in the overdoped regime, the spectra change rapidly from a broad feature with a normal state pseudogap to a much sharper peak at the Fermi level without the normal state gap. Our recent experimental investigations find that the superconducting gap size decreases rapidly with the increase of carrier density in the overdoped regime [6]. This empirical correlation between the disappearance of the broad feature at 100–200 meV and the decrease of superconducting pairing strength suggests that these features are closely related to the pairing interaction.

Although analysis of ARPES line shapes in high-T_c cuprate is controversial [7–11], the doping, momentum, and temperature dependence provide new information to gain added insight. First, a few words about the experimental background due to inelastic scattering of photoelectrons. The background is usually a featureless signal that monotonically decreases towards lower binding energy with a steplike intensity drop near, but slightly below the Fermi level [3,8]. This background exists in all spectra, and our discussion in this paper is based on features observed above this background. Spectra taken from the overdoped and underdoped samples at (α) can be modeled with a broadened Lorentzian at the Fermi level, together with an experimental background. The spectrum from the underdoped sample at (β) cannot be modeled in the same way unless one is willing to use a very large width and an asymmetric shape. On the other hand, since the spectrum is taken at the Fermi surface as determined by the relative intensity change as a function of angle, only a single Lorentzian at the gap edge is expected if the data solely reflect the excitation of a quasiparticle. Therefore, the broad feature at (β) in Fig. 1(A) signals other excitation processes that are coupled to the process of creating a photohole.

The fact that the ARPES data establish the existence of other excitations in addition to the quasiparticle is most evident in spectra from the superconducting state near $(\pi, 0)$ where two components are directly resolved. The top two sets of curves of Fig. 2 are from an underdoped sample recorded with 35 meV resolution, while the bottom set of curves is from a less underdoped sample with 20 meV resolution. In the superconducting state, the experimental data consist of a resolution limited peak, a broad feature near 100–200 meV that is separated from the peak by a dip in intensity [12,13] and the experimental background. This peak represents the excitation of a quasiparticle in the superconducting state, with its position at (β) being determined by the superconducting gap. On the other hand, this peak only represents a fraction of the experimental spectral weight. A significant portion of the spectral weight remains as a broad feature which can only be the signal of additional excitations. It should be noted here that dispersion and polarization analysis suggest that the broad feature is not caused by one-electron excitation from a second band [14,15].

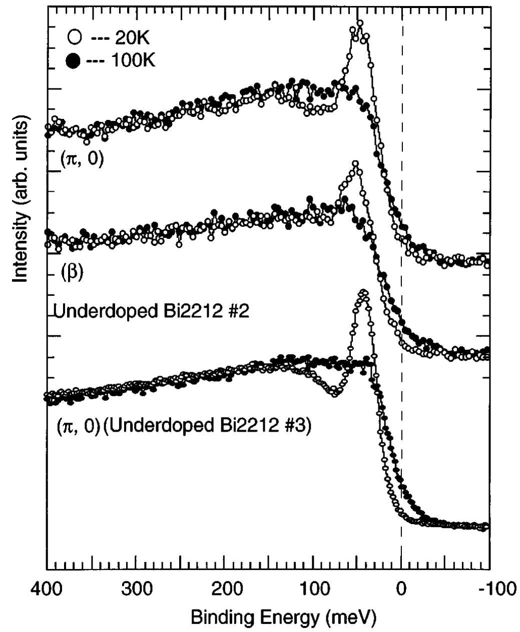

FIG. 2. ARPES data from normal and superconducting states of underdoped Bi2212 near $(\pi, 0)$. As illustrated in the inset of Fig. 1(β) is the Fermi surface crossing point along the $(\pi, 0)$ to (π, π) line and it is very close to $(\pi, 0)$. The upper two sets of curves were recorded with 35 meV energy resolution while the low set of curves was recorded with 20 meV energy resolution.

In Fig. 3 we propose an interpretation of the data discussed in Figs. 1 and 2. Figure 3(α) depicts the hole spectral function in the weak coupling case in which the quasiparticle peak is approximately a Lorentzian centered at ε_k, plus a weak incoherent background. Although the present system is far from a normal Fermi liquid, the spectra of the overdoped and of the underdoped samples at (α) resemble this picture, albeit the quasiparticle width is too large and does not vary as $|\varepsilon_k|^2$ which the Landau theory predicts. Figure 3(β) depicts the spectrum of a hole in a system where it is strongly coupled to collective excitations. In addition to the quasiparticle peak at ε_k, one has excitations at higher binding energy due to the excitation of collective modes. In this case the probability of creating a quasiparticle without creating collective excitations is small, resulting in a weaker spectral weight at ε_k. Thus, the creation of a photohole is more likely to produce collective excitations plus a hole in the quasiparticle band in this case. This loss feature, caused by collective excitations, will have a broad energy distribution because of the momentum dependence of the collective mode spectrum as well as the recoil energy of the hole when a collective excitation is emitted. In a simple boson model [16] of a localized hole coupled a collective excitation of frequency ω_0, this crossover from weak to strong coupling is evident in the spectrum $A(\omega)$, where ω is the negative of the binding energy. Thus,

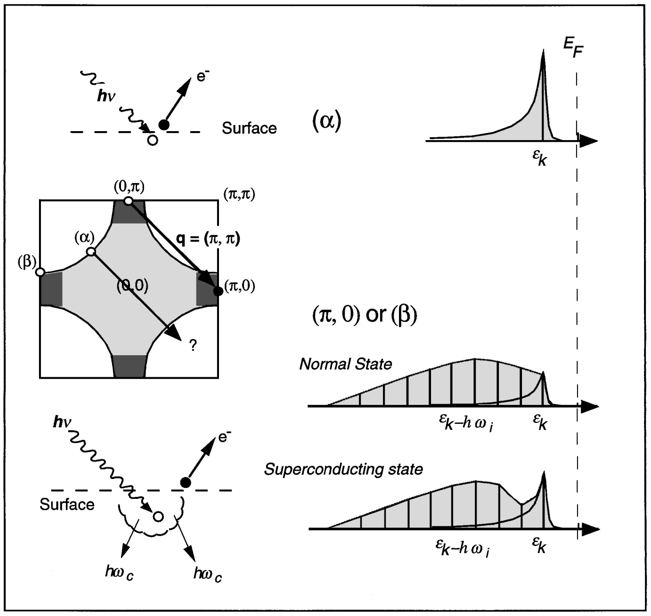

FIG. 3. Illustration of photoemission process and spectral shape in systems with weak (α) and strong couplings [(β) and $(\pi, 0)$]. The Fermi surface picture depicts the phase space considerations for the coupling between the quasiparticle and collective excitations near (π, π). The light shaded area indicates the filled states, and the dark shaded area indicates the flat band region near the Fermi level.

$A(\omega) = e^{-g^2} \sum_{n=0}^{\infty} (g^2/n!) \delta(\omega - \varepsilon + n\omega_0)$; where ε is the quasiparticle energy, equal to the bare hole energy $\varepsilon_0 < 0$ plus the self-energy $g^2\omega_0$. For $g > 1$, this model yields a spectrum in which the quasiparticle peak is comparable to or weaker than that of the collective excitations, and corresponds roughly to the normal state data. In $C_{60}{}^-$ and related compounds, broadening of photoemission features has been attributed to this effect [17,18]. Under favorable conditions, the individual phonon sidebands have been resolved, facilitating a realistic estimate of the electron-phonon coupling constant that is consistent with the T_c of doped C_{60} compounds [17].

From the above discussion, we believe the broad feature in spectra from near $(\pi, 0)$ in the underdoped sample is a manifestation of a strong coupling between the quasiparticle and the collective excitations. From a recent study of underdoped and overdoped samples with similar doping to those reported in Fig. 1 [6], it is found that the superconducting gap of the overdoped sample is about 35% smaller than the underdoped sample. This means that the pairing strength is significantly reduced in the overdoped sample. Hence, the presence and absence of the broad loss feature is empirically correlated to a stronger and weaker pairing. This fact points directly to the collective excitations being the glue that pairs the electrons. In conventional superconductors, the same role is played by phonons which were detected in tunneling experiments [19,20].

While the doping dependence of the loss feature in the photoemission spectra suggests that the collective excitations are a key feature of the pairing interaction, the **k** dependence of the loss spectra gives added weight to this conclusion. There are two possible routes to explain why the above loss feature is highly **k** dependent in the underdoped samples. The first is that the coupling constant g_{kq} may be strongly dependent on the photohole momentum **k**, while the second is a phase space argument. We believe the second route is sufficient to explain the data. Based on neutron scattering data from other cuprates [21–23], we assume the collective modes have a wide energy distribution, with a characteristic energy ω_0, and their spectral function $\chi''(\mathbf{q}, \omega)$ is peaked near $\mathbf{Q} = (\pi, \pi)$. With this assumption, it follows that a photohole created at the Fermi surface near $(0, \pi)$ can emit a collective excitation $\mathbf{q} \cong \mathbf{Q}$, with the hole recoiling to a quasiparticle state in the vicinity of $(\pi, 0)$ near the Fermi energy. Thus, the energy loss in this case is dominated by the collective modes; i.e., it occurs at an energy scale of order ω_0. On the other hand, for photohole near the Fermi surface at (α), emission of a collective mode **Q** will lead to the hole recoiling to a state **k-Q** far from the Fermi energy, and the loss spectrum will be shifted to considerably larger binding energy due to the hole's recoil energy and be intrinsically weak being off resonance (see below) or is forbidden if the hole is scattered to an unfilled state. A further effect enhancing the strong loss spectrum near (β) or $(\pi, 0)$ is the flatness of the quasiparticle bands in this vicinity [24]. Thus, even if the peak of $\chi''(\mathbf{q}, \omega)$ is spread over a sizable region around **Q**, the recoil energy of the quasiparticle is modest and the above conclusions continue to hold.

To quantify these ideas consider a coupled fermion-boson model with band structure corresponding to the Fermi surface in Fig. 1. The photohole spectral function is given by

$$A(k, \omega) = \Sigma_2(k, \omega)/\{[\omega - \varepsilon_k - \Sigma_1(k, \omega)]^2 + \Sigma_2^2(k, \omega)\}, \tag{1}$$

where the self-energy is given within the rainbow approximation by

$$\Sigma_2(k, \omega) = \int g_{kq}^2 \chi''(q, v) A(k - q, \omega') \times \delta(\omega - \omega' - v) dv\, d\omega'\, d^3q/(2\pi)^5, \tag{2}$$

$$\Sigma_1(k, \omega) = P \int [\Sigma_2(k, \omega)/(\omega - \omega')] d\omega'/\pi. \tag{3}$$

The quasiparticle peak occurs when the resonance condition $\omega - \varepsilon_k - \Sigma_1(k, \omega) = 0$ is satisfied and the level width $\Sigma_2(k, \omega)$ is small. The loss peaks occur when $\Sigma_2(k, \omega)$ is large. This leads to a strong loss spectrum if the hole energy $\varepsilon_k + \Sigma_1(k, \omega)$ is small compared to ω_0. However, the magnitude of these peaks are reduced by the large energy denominator in (1) when the recoil hole is in a high energy state or is in an unfilled state above the Fermi surface. This is the case in the underdoped samples at (α). However, near (π, 0) or (β) point, the recoil quasiparticle is near the Fermi energy and the admixture of collective modes is large for coupling constant $g \geq 1$. It is the geometry of phase space imposed by the peak in $\chi''(\mathbf{q}, \omega)$ coupled with the shape of the Fermi surface which enhances the loss spectrum at $(\pi, 0)$ or β while suppressing it at (α). In overdoped specimens, $\chi''(\mathbf{q}, \omega)$ is weak and broad in $\mathbf{q}$, and such enhancement effects vanish. We note that in the limit of small hole band width one can sum the diagram series to retrieve the result of the localized hole model discussed above.

While the quasiparticle excitation cannot be resolved from the higher energy loss features in the normal state, it becomes a resolution limited sharp peak below T_c with a dip separating it from the loss features. As stressed before, this fact is particularly striking in the underdoped samples where the gap already opens in the normal state [3]. One way to understand the data is through a redistribution of the collective excitation energy. If the low energy collective excitations are suppressed for some reason, the low energy portion of the loss feature will reduce its intensity to form the dip. At the same time, the intensity of the quasiparticle will increase because of higher probability to excite it without the low energy portion of the loss features. Without detailed neutron data from Bi2212 and given the significant difference seen in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-\delta}$, it is difficult to be more specific about the energy distribution of the collective excitations. Data from $La_{2-x}Sr_xCuO_4$ with $\mathbf{q} \cong (\pi, \pi)$ do show a depressed scattering intensity at low frequency in the superconducting state [22]. The scattering intensity appears to be moved to an edgelike feature at a somewhat higher energy. This is qualitatively consistent with our picture. On the other hand, data from $YBa_2Cu_3O_{7-\delta}$ is somewhat different [21]. The scattering intensity shows a sharp peak in the superconducting state with weight from both the lower and higher energy regimes. To make a more quantitative analysis, we need to know $\chi''(\omega, \mathbf{q})$ of Bi2212 and calculate $A(k, \omega)$ using (1)–(3). Aside from the change of the collective excitations, the observation of the resolution limited quasiparticle seen in the superconducting state is also due to the longer lifetime in the superconducting state [25,26]. The sharpening of the peak will also make the dip look stronger.

In summary, momentum, temperature, and doping dependence of photoemission line shape yields critical information about the collective excitations which appear to be the glue to pair the electrons. Unlike the phonon anomalies in conventional superconductors that have their distinct characteristics in the energy axis, the collective excitations here have their important characteristics in both momentum and energy space. The collective excitations, and thus the pairing interactions, are peaked at $\mathbf{Q} = (\pi, \pi)$. Although collective excitations of similar properties are observed in neutron experiments, it is important to detect them in the single particle spectral function because it measures the combination $g^2_{\mathbf{kq}}\chi''(\omega, \mathbf{q})$ rather than $\chi''(\mathbf{q}, \omega)$ alone.

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