SHADOW BANDS IN OVERDOPED Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$

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We performed angle-resolved photoemission experiments on overdoped single crystal Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ samples with Tc values as low as 55K and very narrow transitions as measured by AC susceptibility. The photoemission data indicate the presence of a “shadow” peak in the quasiparticle spectral function $A(\omega)$ shifted by $(\pi, \pi)$ from the conventional quasiparticle peak. The ratio of the shadow/quasiparticle peak amplitudes strongly increases with the binding energy. This is consistent with the idea that the shadow peak is not a structural replica of the quasiparticle peak, but rather a maximum in a spectral function which can exist due to interaction with spin fluctuations peaked at $(\pi, \pi)$. © 1997 Published by Elsevier Science Ltd

The question of what boson mediates the superconducting pairing interaction is of great interest in high-temperature superconductors. Alternatives to phonons include spin fluctuations [1]. The exchange by spin fluctuations peaked at antiferromagnetic momentum $Q = (\pi, \pi)$ yields a $d$–wave pairing state which is consistent with a number of experiments on high-$T_c$ materials. However, there are restrictions which have to be met in order to have strong attraction in a $d$–wave channel: (i) spin fluctuations should be peaked at or very near $Q$; (ii) the Fermi surface should be present near $(0, \pi)$ and $(\pi, 0)$ points in the $k$–space where the $d$-wave gap has a maximum; (iii) in the density of states, there should be no precursors of the upper and lower bands of the magnetically ordered state — otherwise vertex corrections would strongly decrease pairing interaction [2]. Near optimal doping, neutron scattering [3], and photoemission experiments [4], and numerical studies of the density of states [5] indicate that all three conditions are satisfied.

An open issue is whether the weak peak at $k = Q$ in the dynamical susceptibility observed at optimal doping means that spin fluctuations are strong enough to yield $T_c$ values consistent with experiment. In photoemission studies, a way to check whether or not spin fluctuations are relevant at optimal doping is to study the form of $A(k, \omega)$ for $k \approx k_F + Q$ where $k_F$ is the (angle dependent) Fermi momentum. Except for a near vicinity of $(0, \pi)$ and $(\pi, 0)$ points, this $k$–region is located far from the Fermi surface in the hole-occupied portion of the Brillouin zone. Accordingly, the quasiparticle peak at $k_F + Q$ is located at high (positive) energies not accessible in conventional photoemission measurements. Nevertheless, if spin fluctuations at optimal doping are still strong, they should modify the low-frequency behavior of the spectral function. Namely, $A(k)$ measured at a given small frequency should have a second maximum at $k = k_F + Q$ due to a peak in the imaginary part of the self-energy (see [6] and the discussion below).

The momentum dependence of the spectral function in Bi$_2$2212 at the smallest possible frequency has been measured by Aebi et al. [8]. They reported observing a second, shadow peak, but have been careful to note that it is an open question whether this peak to due to antiferromagnetic fluctuations or it merely reflects the presence of the structural replica of the Fermi surface and the quasiparticle band.

In this communication we report on measurements of the relative intensity of the shadow peak and the main quasiparticle peak as a function of the binding energy. These measurements distinguish between magnetic and structural origins of the shadow peak. The reason is as follows. If the shadow peak is the struc-
tural replica of the quasiparticle peak, it should have the same Fermi-liquid frequency dependence at the lowest frequencies, $A(\omega) \propto \omega^{-2}$, as the main peak, and the ratio of the two should be virtually independent on frequency. If, on the contrary, the second peak is just an enhancement of the low frequency part of $A(\omega)$ at $k_F + Q$ due to spin fluctuations, then the relative intensity of the shadow peak should vanish when binding energy tends to zero.

We have studied overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ single crystals, with transition temperatures, $T_c=55–62\,\text{K}$ and report observing shadow peaks using conventional angle-resolved photoemission spectroscopy in which one measures spectral function as a function of frequency at a given momentum. We found that the relative intensity of the shadow peak strongly increases with increasing binding energy which agrees with the idea that the shadow peak is not a second quasiparticle peak.

Single crystal samples of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ were grown using a self-flux technique in a strong horizontal thermal gradient to stabilize the direction of solidification.[10,11] The individual crystals were manually separated from the bulk charge utilizing their propensity to cleave micaciously perpendicular to the crystallographic $c$-axis. Typical dimensions for samples in this study were $4\,\text{mm} \times 4\,\text{mm} \times 30\,\text{mm}$, with the shortest length along the $c$-axis.

Our AC susceptibility measurements have shown that the crystals exhibit a $10–90\%$ transition in a $T$ range of $0.5\,\text{K}$ for both optimally doped and overdoped samples. Previous reports [9] indicated a best transition width of $1.3\,\text{K}$ for optimally doped material and $3\,\text{K}$ for somewhat overdoped samples ($T_c=72–75\,\text{K}$). The narrow transition width indicates a more homogeneous oxygen and cation doping than in earlier studies. Figure 1 illustrates AC susceptibility measurements of overdoped and optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ single crystals. There is no indication of either $\text{Bi}_2\text{Sr}_2\text{CuO}_{6+x}$ or $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{10+x}$ phases. This is confirmed by x-ray diffraction and resistivity measurements, as reported in detail elsewhere [10].

Angle-resolved photoemission measurements were performed using a 50 mm radius hemispherical electron energy analyzer, mounted on a two-axis gonioimeter. The samples were transferred from a load lock chamber to the ultrahigh vacuum photoemission chamber (base pressure $6–7\times 10^{-11}\,\text{torr}$). The samples were cooled to just above their superconducting transition temperature of $55–62\,\text{K}$ and cleaved while cold. The sample orientation with respect to the sample surface normal was adjusted in situ and confirmed

![Fig. 1. AC susceptibility measurements of optimally doped, as grown, and overdoped samples. The $10–90\%$ transition width is $0.5\,\text{K}$ for all samples studied.](image)


Figure 2 illustrates angle-resolved photoemission spectra taken along the $\Gamma–X(k_x = k_y)$ and $\Gamma–Y(k_x = -k_y)$ directions in the Brillouin zone, for both cases we plotted the data for an overdoped sample with $T_c = 55\,\text{K}$. Several noteworthy points emerge from the data. First, the main Fermi surface crossing, at $(0.38 \pm 0.01)Q$, occurs at a slightly smaller wavevector than that for optimally doped or slightly overdoped samples [12], because our samples have a higher carrier concentration. Second, we found another peak in the photoemission intensity, that appears at our smallest frequency at $(0.64 \pm 0.02)Q$. Within our experimental error, this second peak is located exactly at a distance $Q = (\pi, \pi)$ from the Fermi surface, i.e., it is a shadow peak. As the wavevector is increased further from $0.64Q$, the shadow peak disperses towards higher binding energies. By a wavevector of $0.75Q$, the peak, now very weak, is at a binding energy of $180–200\,\text{meV}$.

We reproducibly observed these new features of the photoemission spectra on three different samples and in both the $\Gamma–X$ and $\Gamma–Y$ directions. Including looking at more than one angular cut on some samples, we looked for, and found shadow peaks in a total of eight angular cuts. Our results obtained on different samples are presented in Fig. 3. The main Fermi surface measured by P. Aebi et al. is shown by a solid line, the location of the shadow peak by a dotted line, and our experimental results by open circles. The Fermi surface wavevectors obtained for all three samples are, within our experimental error, the same, so the same
Fig. 2. (a) Series of photoemission energy distribution curves (EDCs) versus location in the Brillouin zone along the \((\pi, \pi)\) direction. Inset illustrates location in Brillouin zone at which each spectrum was taken. (b) Corresponding EDCs for the \((\pi, -\pi)\) direction. Note the appearance of a shadow peak for both directions.

Fig. 3. Location of main Fermi surface crossing and a shadow peak nearest to the Fermi energy (open circles), obtained from a series of data like as in Fig. 2. Solid line is Fermi surface for optimally doped samples. Dotted line is shadow peak observed in Ref. [8] for optimally doped samples. Samples are more overdoped than Ref. [8], so the main crossing is closer to the center of the Brillouin zone. The shadow peaks are shifted from the main peaks by \((\pi, \pi)\).

open circle symbol is used for all samples. The strength of the intensity at the shadow peaks is polarization-dependent; the details of this will be reported elsewhere [13].

Figure 4 illustrates the ratio, \(R\), of the shadow peak/main quasiparticle peak amplitudes as a function of binding energy. Data were obtained from six angular cuts on four samples. The ratio increases as the binding energy increases; the increase is as much as by a factor of 4.2. It is very unlikely that such increase can be explained if one assumes that the shadow peak is just a structural replica of the quasiparticle peak. A more realistic conclusion from the data is that there is just one Fermi surface, located at \(k_F\), one band of propagating quasiparticle excitations near \(k_F\), while the shadow peak is just an anomaly in the spectral function. This conclusion also agrees with our experimental observation that the increase of the shadow peak/main peak amplitude ratio is primarily due to the decrease of the main peak amplitude as binding energy increases; the amplitude of the shadow peak demonstrates only weak dependence on the binding energy.

An essential issue in our analysis is background subtraction. The cuprates are well-known to exhibit a sizeable incoherent background in photoemission spectra. For that reason, we took particular care in
Fig. 4. Ratio $R$ of shadow peak/main quasiparticle peak amplitudes as a function of binding energy $E_b$, taken from a series of data like as in Fig. 2. The data and the error bars represent $R$ measured as the ratio of the spectral areas of the main and shadow peaks. The error bars for other three approaches which we used to extract $R$ (see text) are about the same. The ratio $R$ has been normalized to 1.0 at lowest binding energy (45 meV). Open circles are data taken on three different samples. Dotted line is the result expected if the shadow peak is structural in origin. Solid line is the result expected if the shadow peak is due to short-ranged spin fluctuations.

Considering the background in obtaining the analysis illustrated in Fig. 4. We measured the intensity of the shadow and quasiparticle peaks in four different ways: (i) the ratio of the peak height to background at immediately lower binding energy; (ii) the ratio of the peak height to background at immediately higher binding energy; (iii) the total spectral area of the peak; (iv) the spectral area for a fixed, limited kinetic energy range around the peak (to determine the effects of any change in lineshape with wavevector). Further, we normalized the background to have the same value at all wavevectors, and compared shadow and main peaks. Finally, we checked as to whether both the shadow and main peaks had the same change, relative to the background, as a function of binding energy. For all our data analysis methods, our results were similar to Fig. 4: the relative intensity of the shadow peak increases as the binding energy increases.

As we already mentioned, the existence of the second peak in $A(k, \omega \to 0)$ located at $k_F + Q$, can be explained as due to the interaction with spin fluctuations. The exchange of spin fluctuation yields fermionic self-energy $\Sigma(k, \omega) = g^2 \int dq G_0(k + q, \omega + \Omega) \chi(q, \Omega)$, where $g$ is the coupling constant, $G_0(k, \omega) = \omega - (\epsilon_k - \mu) + i\delta \chi g \omega$ is the Green function for free fermions ($\epsilon_k$ is an excitation energy and $\mu$ is the chemical potential). The magnetic susceptibility $\chi(q, \Omega)$ describes overdamped spin excitations and is assumed to be peaked at $Q$. The low-energy part of $\chi(q, \Omega)$ is well approximated by $\chi(q, \Omega) \approx \Delta^2 + \nu^2 (q - Q)^2 + 2i\gamma \Omega$ [14], where $\gamma$ (the damping term) and $\nu$ are of the order of the electronic bandwidth, $\nu$, and $\Delta$ is the gap which vanishes at the antiferromagnetic transition point. The near antiferromagnetism implies that $\Delta \ll \nu$.

Consider now a region of $k$ values which are close to the shadow point $k_F + Q$, but are relatively far from $k_F$ such that $\epsilon_k - \mu = O(\nu)$. In this situation, $A(k, \omega) \propto \text{Im} \Sigma(k, \omega)$. The key result is that the self-energy is nearly singular for $k = k_F + Q$ and $\Delta \ll \nu$, and this near singularity gives rise to a maximum in $A(k)$ at $k = k_F + Q$. The full expression for the low-frequency part of $\Sigma(k, \omega)$ was obtained in [6,15]. Thus, for $\Delta = 0$, we have

$$A(k, \omega) \approx \frac{\omega^2}{(\epsilon_k + Q - \mu)^2}$$

for $\omega < (\epsilon_k + Q - \mu)/\omega_0$ ($\omega_0 = O(\nu)$), and

$$A(k, \omega) \approx \omega^{1/2}$$

for $\omega > (\epsilon_k + Q - \mu)/\omega_0$. Clearly, $A(k)$, measured at a given, small $\omega$ has a maximum when $\epsilon_k + Q = \mu$, i.e., $k = k_F + Q$. For $\Delta \neq 0$, the behavior is more complex, but the peak in $A(k)$ at $k = k_F + Q$ survives as long as $\Delta \ll \nu$.

The above arguments can be applied to the experiments of Aebi et al., who measured $A(k)$ at a given frequency. The present experiments were performed in a different way: the photoemission spectra were obtained as functions of the binding energy (i.e., frequency) for several values of $k$. The location of the shadow peak was obtained as the position of the maximum of $A(\omega)$ at a given $k$. To account for these data, the low-energy spin-fluctuation approach has to be modified because in the present form, it allows one to predict only the form of $A(\omega)$ when it increases as a function of frequency. At larger frequencies, $A(\omega)$ indeed passes through a maximum and then decreases. However, to find the self-energy in this frequency range, one has to utilize the sum rule for the susceptibility which in turn requires one either to add higher powers of frequency and momentum into the expression for susceptibility or introduce a cutoff.
in the momentum/frequency integration over bosonic variables. In both cases, the maximum of $A(\omega)$ is expected to be located at some frequency compared to $\omega_0$ which apparently does not depend crucially on how close $k$ is to $k_F + Q$. The amplitude of $A(\omega)$ near the maximum is also expected not to depend strongly on $\epsilon_{k+Q} - \mu$. In other words, within weak coupling spin-fluctuation approach (when coupling is small enough to yield a pseudogap), only the low-frequency part of $A(\omega)$ changes drastically around $k = k_F + Q$, while the rest of $A(\omega)$ doesn't change much. The near independence of the shadow peak amplitude on the binding energy is consistent with experiment — the shadow peak amplitude does demonstrate some frequency dependence with the maximum at $\omega = 70 - 90 meV$, but the maximum observed change in the amplitude is much less than that of the main band, by a factor of $(x\times3.3)$ or greater. In Fig. 4 we fitted the data for the amplitude ratio by an $\omega^2$ dependence which would be the case if the shadow peak was completely $\omega$ independent, while the main peak follow the Fermi-liquid prediction $A(\omega = \epsilon_k - \mu) \propto \omega^{-2}$. We see that at small frequencies, the fit works reasonably well, but there are deviations at higher frequencies as expected due to higher-order terms in the frequency expansion.

At the same time, the observed strong variation of the shadow peak location with the deviation from $k_F - Q$ cannot presently be explained within the weak-coupling spin-fluctuation approach. One possibility is that some features of the strong-coupling, almost spin-density-wave solution [16], specifically the existence of a second, propagating quasiparticle pseudoband, can still be observed at not very small frequencies even in the overdoped case. Another possibility is that already in a weak-coupling spin-fluctuation theory, the position of the peak in $A(\omega)$ in fact does have some moderate $k$ dependence as a result of the interplay between the parameters in spin susceptibility and electronic dispersion. Recently, Langer et al. performed numerical studies of the shadow peak at large enough doping [17]. They obtained the results consistent with ours in the sense that the shadow peak is not a second quasiparticle peak. Further, consistent with our experimental results, they found some dispersion of the peak position with changing binding energy.

To conclude, in this paper we reported the observation of the shadow peaks in photoemission studies of overdoped 2212Bi samples. We studied the relative intensity of the shadow and main peaks and found that it strongly increases with frequency. This indicates that the shadow peak is not a structural replica of the main peak, but rather an enhancement of the spectral function at $k = k_F + Q$. This enhancement may be due to the interaction with spin fluctuations, and our data thus provide some support to the idea [1] that spin-fluctuations remain peaked at $(\pi, \pi)$ even in the overdoped regime. That is, even highly overdoped samples appear to exhibit paramagnons as low-energy collective excitations. On the other hand, the shadow peak exhibits a strong $k$-dependence of the peak position. Within the spin-fluctuation model, we cannot presently provide a definitive explanation for this observation.

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13. LaRosa, S., et al., unpublished. We had previously
performed such measurements without conclusively observing shadow peaks. The present samples are more homogeneously oxygen doped; the susceptibility width is a factor of \((\times 3 - \times 5)\) more narrow, making it easier to observe weak signal from shadow peaks.


