Temperature and doping dependence of the Bi-Sr-Ca-Cu-O electronic structure and fluctuation effects

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Angle-resolved photoemission data from bulk $Bi_2Sr_2CaCu_2O_{8+\delta}$ show changes in the excitation gap and line shape versus both doping and temperature. We employ two different quantitative analyses; one to search for a gap closing temperature $T^\star$, and the other to further characterize its relation to the superconducting transition temperature T_c. We present observations of the sharp feature near crystal momentum $\mathbf{k}=(1,0)$ in superconducting spectra, and its temperature and doping dependence. This temperature dependence is analyzed together with the shift in the spectral weight's lowest binding energy (leading edge). Finally, we find evidence for persistence of this sharp peak at temperatures slightly above T_c. [S0163-1829(97)06442-4]

Recent angle-resolved photoemission experiments reveal evidence for an excitation gap in the normal state of underdoped high-temperature superconductors (HTS).[1,2] This gap is similar in magnitude and momentum dependence to the superconducting gap, implying that the two have a common origin. These results are in concert with observations of a pseudogap by other experimental techniques, and are consistent with theories of electronic pairing well above the superconducting transition temperature T_c.[3] In this picture, phase fluctuations play a critical role in the temperature interval between the mean-field pairing temperature $T^\star$, and the global superconducting transition temperature T_c. This suggests that strong fluctuation effects are an important hallmark that distinguish the HTS from the conventional superconductors.

In this paper, we report results of an angle-resolved photoemission (ARPES) study of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212), focusing on the doping and temperature dependence of the spectral line shape. In the superconducting state, we observe a resolution limited peak near $(\pi,0)$. This persists to a temperature very close to, but slightly above T_c, indicating that feature is related to the superconducting state. As the temperature approaches T_c from below, this peak moves slightly towards lower binding energies, consistent with a reduction in the superconducting gap. On the other hand, this reduction is far smaller than what one would expect from the complete gap closure indicated by the BCS theory; this is consistent with the earlier report of an excitation gap in the normal state. At temperatures above T_c, although the sharp peak is absent, the spectra's low energy (leading) edge near E_F show a significant energy shift, implying the presence of an energy gap. The difference in leading edge energy position between the $(1,0)\rightarrow(1,1)$ cut (near maximum d-wave gap) and the $(0,0)\rightarrow(1,1)$ one (no d-wave gap) is detectable up to a temperature roughly twice the superconducting T_c. (We write crystal momenta in units of π/a, where a is the lattice constant.) We will also discuss the doping dependence of the spectral line shape, and the persistence of the sharp peak slightly above T_c.

We measured bulk single crystals of Bi2212, annealed to achieve the desired carrier doping.[4] These samples were comparable in quality to typical Bi2212 crystals; all show strong dispersion along the $(0,0)\rightarrow(1,1)$ direction, and all have superconducting transition widths of 2 K or less as measured by superconducting quantum interference device magnetometry. The samples were optically flat, and showed clean Laue patterns. The data were collected with a Scienta SES-200 analyzer, mounted on a UHV system with a helium lamp light source. The total energy resolution was 20 meV, with an angular acceptance of $\pm 1°$, and base pressure 4×10^{-11} torr. Fermi levels and resolution were determined by measuring a gold reference in electrical contact with the sample.

Figure 1 shows the temperature dependent ARPES data at (1,0) from an underdoped Bi2212 sample ($T_c=79$ K).[5] This crystal momentum does not correspond to a Fermi surface crossing, but the dispersive feature is close to the Fermi level, and it is the point at which a $d_{x^2-y^2}$ gap has its maximum. At low temperature, the feature has two distinct components: a broad, incoherent structure at around 150 meV binding energy, and a sharp, resolution limited peak at 40 meV, which disappears in the normal state. Unlike the other ARPES features seen in Bi2212, this sharp peak's damping is small enough that it can be identified as a quasiparticle. As such, one might expect its energy to shift as the gap magnitude changes. Strikingly, however, the sharp peak's movement with temperature is very small. This lack of movement implies that the excitation gap does not close at T_c.

In an effort to explore the relation between T_c and $T^\star$ quantitatively, we use the following technique. As shown in Fig. 2, we decompose the (1,0) line shape into the two components discussed above, and fit the sharp peak. The low binding energy slope of the broad piece is visible between the sharp and broad peaks, and again near the background level; these two regions are fit to a second order polynomial, interpolating the broad feature's leading edge and thus allowing isolation of the sharp feature. The sharp peak ex-

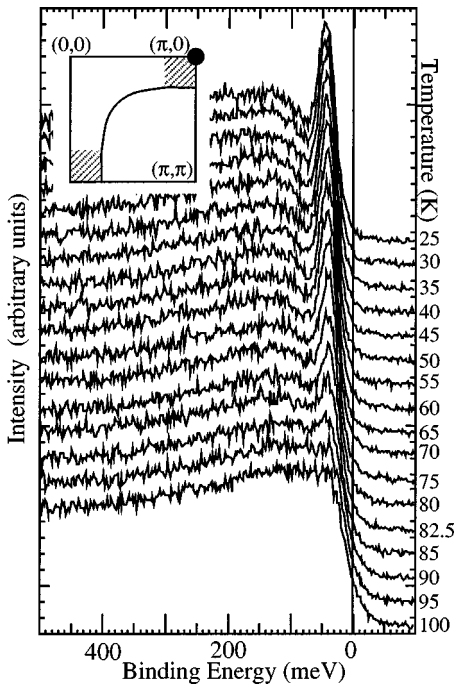

FIG. 1. Temperature dependence of raw data, taken at $(\pi,0)$. The inset shows the position of the measurement in the Brillouin zone.

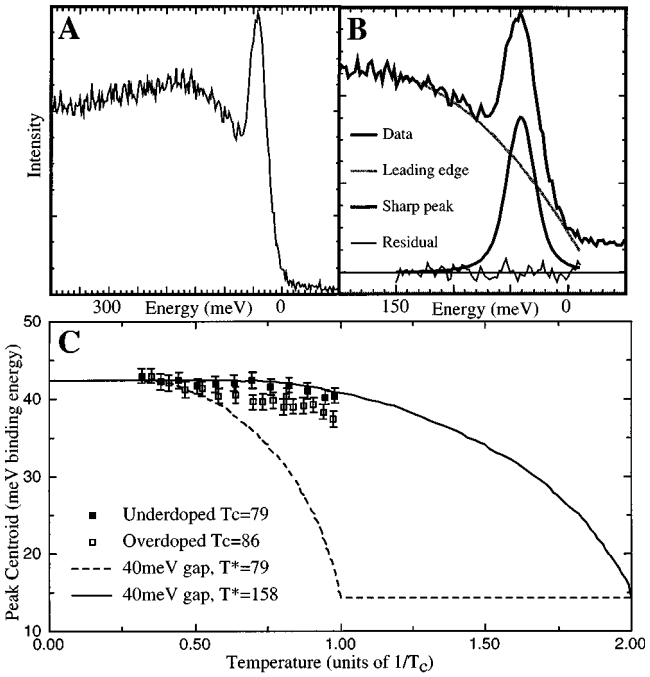

FIG. 2. Analysis of the sharp feature as a Bogoliubov excitation. (A) The superconducting line shape at $(\pi,0)$. (B) The fitting procedure—a broad, sloping leading edge is subtracted from the data, and the difference is fit with a resolution-broadened Lorentzian peak. (C) Comparison of measured peak position, and the position expected for the underdoped sample, for two gap closing temperatures.

tracted in this way fits very closely to a resolution broadened Lorentzian feature, shown in Fig. 2(B). We use this method to provide a systematic, quantitative measure of the sharp feature's position and width.

We measured two samples with $T_c = 79$ K (underdoped) and 86 K (overdoped) over a broad range of (superconducting) temperatures, and characterized the sharp feature. Interestingly, the full width at half maximum was fairly constant in both cases, with 16 ± 1 meV for the underdoped sample, and 18 ± 1 meV for the overdoped one. Both widths increased roughly 4 meV in the last 10 K below T_c. Details of these and other measurements will be available elsewhere.[6] As mentioned above, the peak position in Fig. 2(C) moves only slightly over the range of superconducting temperatures.

Under the assumption that this peak is a Bogoliubov excitation,[7] one can infer the gap magnitude Δ_k from the peak position $\sqrt{\epsilon_k^2 + \Delta_k^2}$ where ϵ_k is the kinetic energy of the quasiparticle. The low-temperature value of Δ_k can be determined by examining the $(1,0) \rightarrow (1,1)$ Fermi surface crossing. While the $d_{x^2-y^2}$ gap changes by less than 4% between (1,0) and the crossing, ϵ_k reaches zero when the quasiparticle crosses the Fermi energy. The minimum binding energy of the sharp peak along this cut is roughly equal to Δ_k; in the case of the underdoped sample at low temperature, this is just over 40 meV. Plugging this number into the equation for the peak position implies an ϵ_k between 10 and 15 meV for momentum (1,0).

Figure 2(C) shows the experimental peak position from the two samples. At first glance, this peak energy appears to be temperature independent. A more detailed inspection shows that its energy decreases slightly as one approaches T_c from below. A possible interpretation is that the gap will be closed at a temperature far above T_c, and the superconducting transition temperature itself is determined by other factors. With the reasoning from the previous paragraph, we can obtain information about $T^\star$ from the temperature dependence of the Bogoliubov excitation energy. The temperature dependence of the gap in weak coupling theory[8] is superimposed on Fig. 2(C), after adjustment for a nonzero ϵ_k, with an assumed zero temperature gap of 40 meV, and a critical temperature T_c. The model energy curves are drawn to match ϵ_k of the underdoped sample, indicated by the dark squares. Clearly, the curve is not close to the experimental data. In this model, the trouble with the curve is that it corresponds to a gap closing at T_c; by analogy to weak coupling theory, this is further evidence that the gap actually closes at a $T^\star$ which is significantly higher.

Of course, the measurement of a superconducting state feature will be insufficient to determine the value of $T^\star$, but we show a curve with $T^\star = 2.0 T_c$ just for comparison—it has a much better fit with the underdoped data. The point of this method is not to identify a pairing mechanism, but rather to show what parameters are consistent given a common assumption. Interestingly, the overdoped data points also imply a gap function which persists well above T_c; we will return to this issue later.

We should point out that this method of gap determination is different from that which we have used in the past. Here, we rely on the assumption that the sharp peak is indeed due to a Bogoliubov excitation, with behavior analogous to the weak coupling case. This technique has the disadvantage that it cannot measure the gap above T_c, making direct measurement of $T^\star$ impossible. Another method of gap determi-

nation is to find the shift in the leading edge between spectra taken for crystal momenta where the gap is expected to be large, and small.[1] Here, large qualitative changes in the line shape will confuse the technique, since the leading edge energy is simply a heuristic measurement. Fortunately, there is no such change above T_c, especially in the underdoped samples, so we can use the leading edge measurement to obtain information about $T^\star$.

To find the temperature dependence of the normal state gap's magnitude we measured samples in the temperature range from 60 to 300 K, from the superconducting state to the highest temperature at which the dispersive features are still clearly evident. At each temperature, a pair of crystal momentum cuts were taken which cross the apparent local-density approximation Fermi surface along the $(0,0)\rightarrow(1,1)$ direction, where a $d_{x^2-y^2}$ gap has its node, and along the $(1,0)\rightarrow(1,1)$ direction near (1,0), where the gap is large. For each cut, we measured the minimum binding energy of the ARPES spectra's leading edge (LE). This allows us to characterize the gap through the shift in LEs at the two **k** values, a heuristic described in an earlier paper,[1] where we argue that a leading edge shift is evidence for the existence of an anisotropic excitation gap.

Figure 3 shows LE data from two underdoped samples. Panel 3(A) shows, for a single sample ($T_c = 84$ K), the LE binding energies from the two momentum cuts, as a function of temperature. Some of the movement in the LE points is due simply to the changing line shape. As the temperature increases, the Fermi function broadens, changing the binding energy of the LE regardless of the gap value. Also, the broadening Fermi cutoff and decrease in intensity leads to higher uncertainty in the determination of the edge midpoint, a trend which is reflected in the error bars. At higher temperatures, the LE movement towards negative binding energy may be due to the changing ratio between the widths of the dispersive feature and the Fermi function. We measured the gold reference at each temperature, to catch any drift in the Fermi level. It is possible that the dispersive features' linewidths start to differ at higher temperatures, leading to the slight separation of LE values. Due to the low intensity as the temperature increases above 250 K, it is unclear to what extent this effect exists. Still, at lower temperatures, the difference in LEs will be a robust indicator of the anisotropic gap.

In measuring this gap, it is important to note that we are measuring the difference in the LE of spectra from two momenta. Here, the LE is indicative of the lowest energy excitation available at that momentum, but not equal. Its actual position is also determined by the ratio of net linewidth (including dispersion) to Fermi function width, and by the instrument resolution. Every good Bi2212 sample will yield spectra from the $(0,0)\rightarrow(1,1)$ direction in which the LE binding energy is negative by several meV. Clearly this is different from the (strictly non-negative) excitation gap, so the LE binding energy is at least offset from that value. Here, we take the shift between spectra from two **k** values where the dispersive feature has a similar linewidth. In this case, the offset from the gap value should cancel to within one or two meV, and yield a measurement of the anisotropic component in the gap function. In the context (largely accepted in the community) of a gap with mostly $d_{x^2-y^2}$ symmetry, the LE shift is indeed indicative of the maximum excitation gap magnitude.

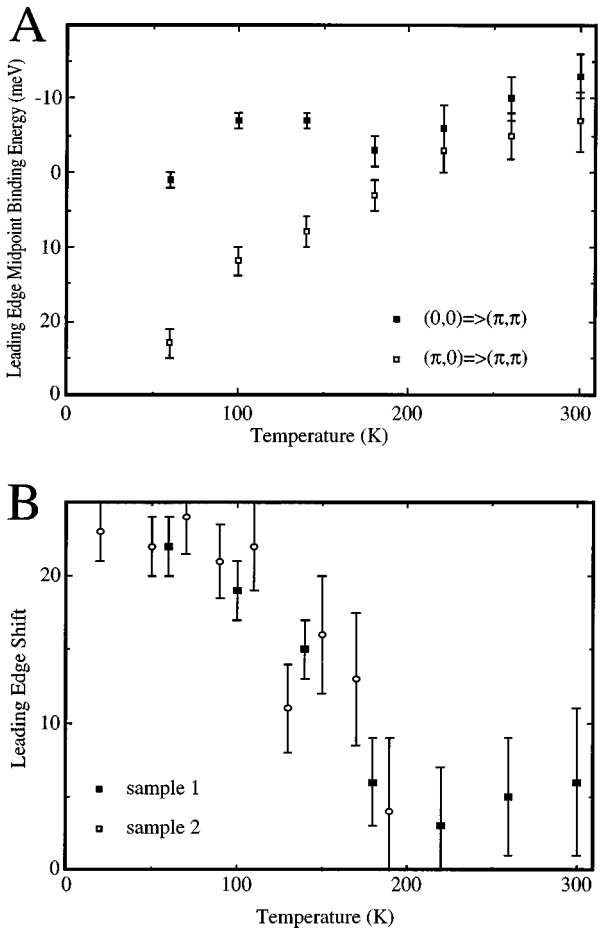

FIG. 3. Leading edge shift vs temperature, for two samples. The top panel shows the leading edge midpoint energies along two indicated momentum cuts, for a single sample. The bottom panel the leading edge shift from the same sample (dark squares) and noisier data from the second sample (open circles).

Figure panel 3(B) presents the LE shift from the data in panel 3(A), along with (noisier) data from another sample with the same T_c. Consistent with more detailed comparisons between the normal and superconducting state,[1] there is no great change in the LE shift at the superconducting transition. Rather, there is a smooth, decreasing curve which reaches zero at roughly 200 K, which we will take as an ARPES data point for $T^\star$. Another recent study also derived values for $T^\star$,[2] with numbers that interpolate to roughly 70 K lower. In this study, however, it is stated that the leading edge position was in fact measured relative to the Fermi energy, instead of another leading edge at a different momentum point; one would expect that this lowers the inferred gap values, and thereby also lowers $T^\star$. Our second sample is also consistent with $T^\star = 200$ K. This $T^\star$ should be regarded as the temperature below which the gap is detectable by ARPES. The relatively poor energy resolution of ARPES makes it difficult to observe small (compared to the maximum magnitude) gaps; the temperature reported here is a lower bound.

Here, there are two technical issues which we must address. First, the apparent discrepancy in the gap value determined by the two methods deserves an explanation; it is rooted in the different criteria used in the two cases. If a Bogoliubov quasiparticle is clearly identifiable, then that technique may give a result closer to the real Δ value, provided ϵ is known. If a Bogoliubov quasiparticle is not well defined in the spectrum, i.e., one only sees a broad feature, then one must use the LE shift criteria for gap measurement. Although the magnitude of the LE shift is not exactly equal to the gap, it is a sufficient measurement for identifying the value of $T^{\star}$. In the past, the LE shift has been used successfully to identify the superconducting gap anisotropy.[9] This result was corroborated by a study in which even the broad features were fit using the Bogoliubov quasiparticle formalism.[7]

Second, the implication (from Bogoliubov quasiparticle analysis) that the overdoped ($T_c = 86$ K) sample has a gap well above T_c deserves further attention [see Fig. 2(C)]. Even though the gap is smaller in magnitude at this doping (the normal state ϵ_k is higher), one cannot draw a suitable curve for a gap closing at T_c. In fact, recent tunneling data suggests that the gap may not close (in the conventional sense) regardless of doping, at all.[10] In our analysis, the pairing onset does not coincide with superconductivity even at higher carrier dopings; of course, one may also want to compare the data with predictions of strong coupling theory, rather than weak coupling theory. Future studies of higher doping levels are important.

This result is not consistent with that of LE shift analysis. From the LE criteria, the 86 K sample has a very small gap because of the small LE shift from the $(0,0)\rightarrow(\pi,\pi)$ cut to the $(\pi,0)\rightarrow(\pi,\pi)$ cut. There are also results[11] from a further overdoped sample ($T_c = 78$ K), in which the LE gap is zero, but there is a (smaller) Bogoliubov quasiparticle gap. This discrepancy stems from the different methods used, and further studies in this area are needed. Although our methods may not be able to pinpoint $T^{\star}$, they do provide a qualitative picture in which the gap in underdoped samples does not close until a temperature much higher than T_c, with signs of similar behavior in overdoped samples.

If the gap does not close until temperatures well above T_c, but the sharp feature at $(\pi,0)$ disappears near T_c, then this feature must not be indicative of a gapped state, but rather of superconductivity. The exact origin of this peak is not known; to support the model used above, it is important to make a distinction between the sharp and broad parts of the spectrum; past studies have not provided explicit evidence for their separate origin. After the original report of the line shape,[12] papers have argued that the features are not due to separate dispersive modes in the excitation spectrum.[13,14] While we cannot address the origin of the broad part of the spectrum here, Fig. 4(A) provides an important clue, the doping dependence. The figure shows ARPES spectra from $\mathbf{k}=(1,0)$ for an underdoped ($T_c = 79$ K) and an overdoped ($T_c = 86$ K) sample. Since the T_c vs doping curve is flat at the top, this represents a large change in carrier concentration. While the sharp feature shifts very little from one spectrum to the other, the broad peak moves roughly 90 meV. As discussed in our earlier work,[15] the broad dispersive feature's centroid moves to lower binding energy as the doping is increased. This can be accomplished simply by adding spectral weight near the Fermi energy; most likely it is drawn from higher binding energy. It has recently been suggested that the cause of the weight shift is collective excitations coupled to a distinct quasiparticle excitation.[16]

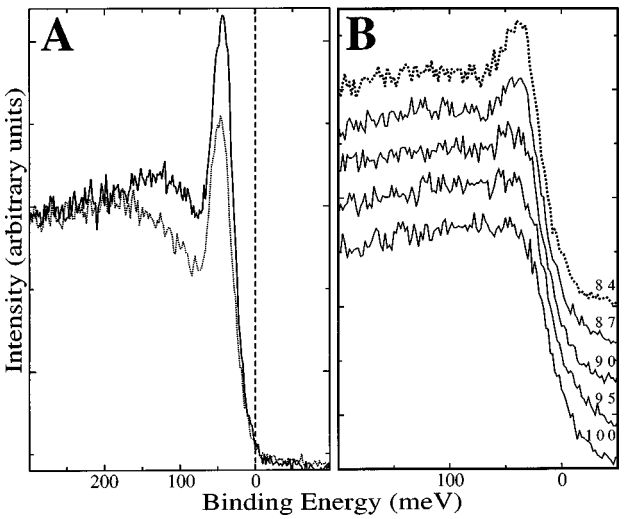

FIG. 4. Behavior of the sharp peak. (A) Doping dependence of superconducting spectra at $(\pi,0)$. The underdoped (dotted) spectrum and the overdoped (bold) one have the sharp feature at almost the same energy; meanwhile, there is a significant shift in the broad spectral weight. (B) Temperature dependence at $(\pi,0)$ in an overdoped sample ($T_c = 86$ K). The peak persists at temperatures slightly above T_c.

While our previous work[1] demonstrated the absence of a sharp feature in an underdoped normal gapped state, Fig. 4(B) shows that the feature persists in overdoped samples, at temperatures slightly above T_c. The sample in this figure has $T_c = 86$ K, so only the 84 K spectrum was taken in the superconducting state. Using the criteria established in our earlier work,[1] we know that the sample has a small normal state gap even at 100 K, as there is a few millivolts leading edge shift between the $(0,0)\rightarrow(1,1)$ crossing and the $(1,0)\rightarrow(1,1)$ one. The 100 K spectrum does not have the sharp feature, as in the previous figure, but the 90 K spectrum probably does. At 87 K, although the curve is noisy, there definitely is a peak and a dip greater than the noise level. Care was taken to ensure that any error in the temperature measurement was towards the high side; the 84 K spectrum may actually be the normal state, but the 87 K spectrum is definitely not superconducting.

Superconducting fluctuations above T_c in BCS superconductivity are, of course, well known.[8] It is becoming clear, however, that the transition at T_c in HTS has a different nature from the BCS superconducting one. If our observation is of superconducting fluctuations, then the explanation may lie in the local phase coherence of electron pairs. To go further requires a better understanding of the sharp feature to begin with. Since the transition at T_c is not determined by mean-field calculations, the nature of the fluctuations must be different as well.

In conclusion, we have presented observations of the

superconducting and gap formation transitions at various dopings, in Bi2212. We show that the energy of the superconducting state Bogoliubov excitation does not shift appreciably with temperature, indicating by analogy with weak coupling theory that the gap formation temperature ($T^\star$) is significantly higher. This is corroborated by the temperature dependence of the LE shift, determining $T^\star$ for a doping below optimal. In addition, we examine the sharp feature in the superconducting spectrum at well separated doping levels and temperatures. On a finer temperature scale, we find fluctuations in the sharp feature existence, above T_c in an overdoped sample.

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[5] The surface of an underdoped sample is reactive in vacuum. We notice that, with time, the spectra evolve slightly towards those obtained from more overdoped samples. Since the temperature dependent study requires time to accumulate, the spectra in Fig. 1 are probably from a less underdoped sample. However, this does not affect the purpose of our discussion here, namely there is a gap transition higher than the superconducting one. For a fresh spectrum (underdoped, $T_c = 79$ K), see Fig. 4(A).
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