Electronic Spectrum of the High-Temperature Superconducting State

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Improved experimental conditions enabled us to increase the signal-to-noise ratio of the photoemission spectra for the superconducting state of $Bi_2Ca_2SrCu_2O_8$, taken with high angular and energy resolution. This also enabled us to reveal a pronounced minimum that separates the two basic features of the spectrum, the narrow quasiparticle excitation peak and the still controversial broad band at lower kinetic energies. The minimum is approximately 3Δ below the Fermi level.

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High-resolution photoemission spectroscopy has recently emerged as a fundamental probe of the superconducting state for high-temperature superconductivity [1]. Early experiments clearly established the presence of a gap in the electronic spectrum [2–5]. The spectrum in the superconducting state is dominated, of course, by the narrow peak corresponding to quasiparticle excitations [2,6]. This peak, however, is accompanied by a broad excitation band at lower kinetic energies, whose nature has not been clarified and whose relevance to the mechanism of high-temperature superconductivity has been emphasized by several authors [7–9].

We carefully analyzed the spectrum with angular and energy resolution similar to or better than those of previous high-resolution studies—but with enhanced signal-to-noise ratio. This enabled us to clearly reveal an additional feature that appears important for the general interpretation of the spectra and of the corresponding superconducting state: a pronounced minimum that separates the broad band from the narrow peak. The minimum could not be easily seen in previous studies because of the relatively large noise, and therefore its position could not be determined. We found that the position is close to 3Δ below the Fermi level, where Δ is the gap parameter (i.e., 2Δ is the gap). A broad-band threshold at 3Δ is predicted, for example, by Littlewood and Varma [7,8] based on the "marginal-Fermi-liquid" model, by Müller, Arnold, and Swithart [10], and by other theorists [11].

Our experimental procedure was a standard synchrotron-radiation photoemission approach, enhanced in order to achieve high angular and energy resolution with a high signal-to-noise level; the results are, to the best of our knowledge, state of the art in solid-state photoemission spectroscopy. The main factors for simultaneously reaching high resolution and high signal-to-noise levels were as follows: the use of very high-quality single crystals of $Bi_2Ca_2SrCu_2O_8$ cleaved *in situ* under ultrahigh vacuum, whose structural and transport properties were carefully characterized; the use of a 4-m normal-incidence photon monochromator with resolving power up to 4.3×10^3; the use of a carefully magnetically shielded high-resolution VSW electron analyzer; and the use of synchrotron radiation.

In order to test reproducibility, the experiments were performed on twenty different samples from different batches. In each case, many different points of the Brillouin zone were explored by varying the photoelectron collection geometry. The results presented here, although a subset of all our data, lend support to conclusions consistent with the entire set of data for different samples and different collection directions.

The results, for example those shown in Fig. 1, demonstrate that we achieved a maximum overall energy resolution of approximately 15 meV [Gaussian full width at half maximum (FWHM)] with an angular resolution of $\pm1°$. The signal-to-noise level can be appreciated from the data of Fig. 1 as well as from those of the other figures.

The data of Fig. 1 exhibit the narrow peak approximately 40 meV below the Fermi level E_F, already reported, e.g., in Ref. [2]. In this case, the peak's FWHM is 25 meV, due to an intrinsic width of 20 meV and to an instrumental broadening of 15 meV. The two curves in Fig. 1 were normalized to each other in the figure's spectral region far from the Fermi energy, where the effects of the superconducting transition are negligible. The corresponding normalization factor coincided within 3% with the ratio between the synchrotron-source electron-beam currents for the two spectra.

Note that the opening of the superconducting gap is revealed in Fig. 1 by the shift in energy of the leading spectral edge. The dot in the inset of Fig. 1 shows, in the two-dimensional Brillouin zone, the k vector corresponding to the peak of the normal-state spectrum.

We can clearly observe in Fig. 1 the minimum that follows the narrow peak and marks the onset of the broad excitation band. The position of the minimum is approximately 75–85 meV below E_F. Previous estimates of the gap parameter Δ from the photoemission spectra ranged

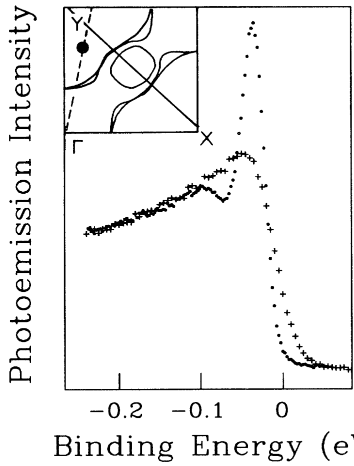

FIG. 1. High-resolution angle-resolved photoemission spectra of $Bi_2Ca_2SrCu_2O_8$, taken in the normal state (crosses) and in the superconducting state (dots), with a photon energy $h\nu=22$ eV. The energy scale is referred to the Fermi level. The overall energy resolution (Gaussian full width at half maximum) was 15 meV for these particular spectra. The angular resolution was $\pm 1°$. Inset: The two-dimensional Brillouin zone corresponding to the cleavage planes, with the Fermi surface [14]. The dashed line shows the direction of the two-dimensional k vectors of the collected photoelectrons. The dot shows the two-dimensional k vector corresponding to the peak of the normal-state spectrum.

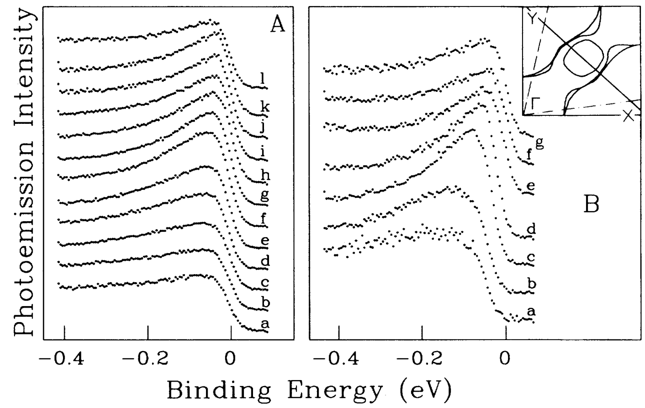

FIG. 2. Photoemission spectra taken in the normal state at $h\nu=22$ eV, along two different directions of the two-dimensional Brillouin zone. The directions are shown in the inset: for (A), dashed line; and for (B), dash-dotted line. For (A), the collection angle from the normal direction was 6°, 8°, 10°, 12°, 14°, 16°, 18°, 20°, 22°, 24°, 26°, and 30° for spectra a–l. For (B), it was 1°, 3°, 5°, 7°, 11°, 13°, and 15° for spectra a–g.

from 24 to 30 meV [2–5]; from the data of Fig. 1, using the method of Müller, Arnold, and Swithart [10], one obtains a value of 25 meV. Thus, if we express the minimum as $\eta\Delta$ with η being an integer, the best estimate is $\eta=3$.

In order to correlate our data with those of other authors [2,6,12,13], we extensively explored the normal-state spectra along different directions in k space. We confirmed the results of other studies, specifically finding the highly dispersive feature whose width increases with the distance from the Fermi level, reported in Refs. [2] and [13] along the Γ-X direction [14]. In other directions of the Brillouin zone, we found a less dispersive behavior, again similar to the results of other studies [13]. Examples of normal-state spectra with different degrees of dispersion are shown in Fig. 2 for two directions in the Brillouin zone.

During repeated temperature cycles above and below the critical temperature, we always found that the normal-state peak was replaced by the much narrower excitation peak shown in Fig. 1. This peak was observed in large portions of the Brillouin zone. For example, Fig. 3 shows superconducting-state spectra taken along the direction of the two-dimensional Brillouin zone shown in the inset [which is the same as for Fig. 2(A)]. The narrow peak is visible for most of the k-space line, except for the portion close to the Γ point—indicating a relation with the Fermi surface [15].

Experiments conducted along different directions in k space and at different distances in k space from the Γ point are systematically consistent with an onset of the broad excitation band at approximately 3Δ. The broad band extends in all spectra over hundreds of meV. No evidence was found for k dependence of the position of

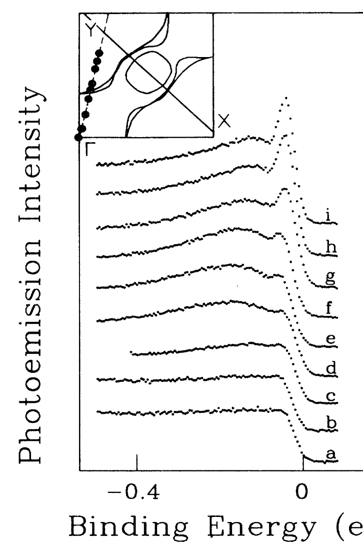

FIG. 3. Photoemission spectra taken in the superconducting state at $h\nu=22$ eV, along the direction of the two-dimensional Brillouin zone shown by the dashed line in the inset. The dots in the inset show the two-dimensional k vectors corresponding to the sharp peak for each of the spectra. The collection angle from normal was 0°, 2°, 6°, 10°, 12°, 14°, 18°, 20°, and 22° for spectra a–i. Note that the apparent differences between this figure and Fig. 1 are due to a difference in the instrumental broadening.

the minimum, although the minimum is less pronounced for some regions of the Brillouin zone.

The interpretation of the nature of the broad band and of its onset depends on the adoption of a specific theoretical framework for high-temperature superconductivity, which is beyond the experimental scope of this article. Without risking theoretical speculations, we limit our analysis to stating the following facts. First, the direct comparison of normal-state and superconducting-state spectra shows that the peak visible in the superconducting state is much narrower than any feature observed in the normal state. Second, the onset of the broad band at 3Δ is consistent, for example, with the predictions of Littlewood and Varma [7,8]. Specifically, their model calculation of the quasiparticle excitation spectrum produces an incoherent "tail" above this threshold.

We conclude then that the detailed quasiparticle spectra produced by our study lend support to the hypothesis that the broad band in the photoemission spectra is *not* a mere "background," but an intrinsic and important feature of the excitation spectrum [7–9]. The identification of the nature of such a feature is therefore as important as that of the narrow quasiparticle peak.

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Note added.— After submission of the present article, Dessau *et al.* [16] published a work previously unknown to us, with results and conclusions similar to ours. In particular, they found the minimum below the main peak; leading-edge and peak differences between spectra in different directions indicate gap anisotropy magnitudes similar to those of Ref. [16].

There are, however, some significant differences between the two studies. Dessau *et al.* [16] observe the absence of the spectral minimum along the Γ-*X* direction, and present data on its presence along the direction perpendicular to it. In our case, we also clearly observe the minimum in the direction of Figs. 1 and 3, and, as already mentioned, in many other parts of the Brillouin zone, with no evident *k* dependence; Fig. 4 shows, for example, data similar to those of Fig. 1, but along the direction near Γ-*M* shown in the inset: The minimum is clearly visible. This point is extremely important for the interpretation of the spectra— see, in general, our discussion, the discussion of Ref. [16], and, in particular, Anderson's work mentioned there.

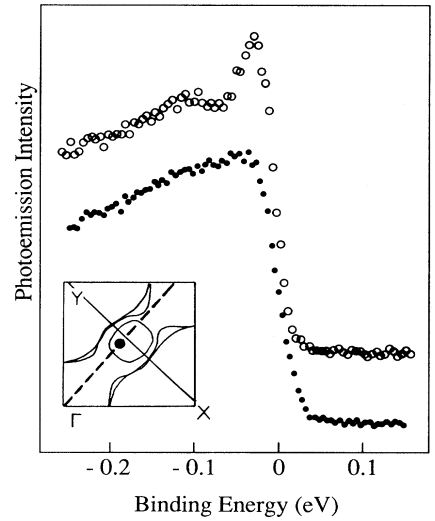

FIG. 4. Spectra similar to those of Fig. 1, but along a direction near Γ-*M*.

We emphasize that we did see the minimum in a direction that is quite close to Γ-*X*, suggesting that dispersion arguments can be ruled out as an explanation of its absence along Γ-*X*. Having extended the observation of the minimum to most of the Brillouin zone, we can confirm the striking difference in *k* dependence between the normal and the superconducting state.

Also note that, whereas Ref. [16] puts most emphasis on the minimum and other spectral features, our data clearly show the band below the minimum as a separate peak; this point is important since the band plays an important role in several theoretical models. We note, for example, that it has been recently proposed as a fingerprint of the resonating valence state [17]. In our experiments we found that the band and the minimum are not affected in the same way by contamination; the latter, in some cases, is removed while the former is still present.

Finally, we found that the conclusions of Ref. [16] concerning the nonconservation of the area under the spectrum during the superconducting transition cannot be generalized to all spectra, since, for example, the area in Fig. 1 is conserved within 2%.

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