Origin of the Peak-Dip-Hump Line Shape in the Superconducting-State $(\pi, 0)$ Photoemission Spectra of $Bi_2Sr_2CaCu_2O_8$

A. A. Kordyuk,[1,2] S. V. Borisenko,[1] T. K. Kim,[1] K. A. Nenkov,[1] M. Knupfer,[1]
J. Fink,[1] M. S. Golden,[3] H. Berger,[4] and R. Follath[5]

[1]*Institute for Solid State Research, IFW Dresden, P.O. Box 270016, D-01171 Dresden, Germany*
[2]*Institute of Metal Physics of National Academy of Sciences of Ukraine, 03142 Kyiv, Ukraine*
[3]*Van der Waals-Zeeman Institute, University of Amsterdam, NL-1018 XE Amsterdam, The Netherlands*
[4]*Institut de Physique Appliquée, Ecole Politechnique Féderale de Lausanne, CH-1015 Lausanne, Switzerland*
[5]*BESSY GmbH, Albert-Einstein-Strasse 15, 12489 Berlin, Germany*

(Received 18 October 2001; published 30 July 2002)

From detailed high-resolution measurements of the photon energy dependence of the $(\pi, 0)$ superconducting-state photoemission spectrum of the bilayer Bi high-temperature superconductors, we show that the famous peak-dip-hump line shape is dominated by a superposition of spectral features originating from different electronic states which reside at different binding energies, but are each describable by essentially identical single-particle spectral functions. The previously identified bilayer-split CuO_2 bands are the culprit: with the ''superconducting'' peak being due to the antibonding band, while the hump is mainly formed by its bonding bilayer-split counterpart.

DOI: 10.1103/PhysRevLett.89.077003 PACS numbers: 74.25.Jb, 71.18.+y, 74.72.Hs, 79.60.–i

The nature of the low-energy electronic excitations near the $(\pi, 0)$ region of **k** space is of vital relevance to a number of key physical properties of the cuprate high-temperature superconductors (HTSC), as it is here that the superconducting order parameter has its maximal magnitude and where many believe the coupling that causes superconductivity makes itself most strongly felt. Unfortunately, this region of **k** space does not willingly reveal its secrets and the true situation is veiled by the effects of, for example, the coupling between the different CuO_2 planes in multilayer HTSC. As angle-resolved photoemission spectroscopy (ARPES) is a very direct, **k**-resolved probe of the electronic system and its coupling to other degrees of freedom, the low-energy photoemission spectra from the $(\pi, 0)$ point of the Brillouin zone (BZ) of the HTSC have been the focus of unceasing experimental and theoretical interest over the past 15 years. Of particular interest has been the now-famous peak-dip-hump (PDH) line shape seen at $(\pi, 0)$ in the superconducting state [1]. This line shape was, up to the present, widely believed to be the result of a single spectral function (see, for instance, Refs. [2–4]), caused by, e.g., strong coupling to bosons [4–6], the details of which are expected to reveal the identity of the interactions involved in the generation and perpetuation of the superconducting state in these systems [5–8]. It is safe to say that the line shape of the photoemission spectrum at this point in **k** space and its interpretation is undoubtedly one of the outstanding features in the high T_c canon.

In this Letter we report the results of a detailed, high resolution ARPES study of this important BZ region in overdoped, modulation-free Pb-Bi2212 crystals using a wide range of excitation energies. The central result is that the strongly differing photon energy dependence of the intensity of the ''peak'' and ''hump'' in the $(\pi, 0)$ spectrum, as well as the variation in the binding energy of the dip as a function of excitation energy, essentially exclude a scenario in which the peak, dip, and hump features in the superconducting state are dominated by a single-band spectral function. The data support a paradigm change in our interpretation of the PDH line shape, and argue for a straightforward picture in which—at least for the overdoped compounds—the ''superconducting'' peak is related to the antibonding CuO-bilayer band and the hump is mainly formed by the bonding band.

The ARPES experiments were carried out using angle-multiplexing photoemission spectrometers (SCIENTA SES200 and SES100). The momentum distribution maps and series of energy distribution curves (EDCs) were measured at 300 or 39 K using $h\nu = 21.218$ eV photons from a He source [9–11]. The $(\pi, 0)$ EDCs were recorded using radiation from the U125/1-PGM beam line at BESSY. The total energy resolution ranged from 8 meV (FWHM) at $h\nu = 17$–25 eV to 22.5 meV at $h\nu = 65$ eV, as determined for each excitation energy from the Fermi edge of polycrystalline gold (which also gives the energetic calibration in each case), and the angular resolution was less than 0.1°. All data were collected on two similar overdoped crystals of Pb-Bi2212 ($T_c = 69$ K) with the exception of the left panel in Fig. 1, which is from pure Bi2212 ($T_c = 89$ K). All $(\pi, 0)$ EDCs were measured at a temperature of 27 K—deep in the superconducting state.

We start by presenting Fermi surface maps of pristine and Pb-doped Bi2212 in Fig. 1. It is well known that in the case of pure Bi2212 such maps contain an additional set of extrinsic features originating from the scattering of the outgoing photoelectrons on the incommensurably modulated BiO layers [9,12] called diffraction replicas (see

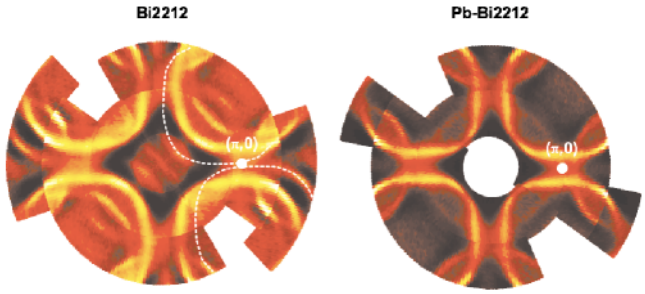

FIG. 1 (color). Fermi surface maps of pure Bi2212 (left panel) and Pb-Bi2212 (right panel) measured at room temperature.

white dashed lines in Fig. 1). With the aid of such maps, it is also easy to estimate to what extent the line shape of a given EDC may be contaminated by diffraction replicas. Obviously, the most perilous region for examination would be one in which a high density of different features overlap or are closely separated, such as the $(\pi, 0)$ point of Bi2212.

The use of modulation-free samples allowed us to clarify the Fermi surface (FS) topology [9] and show that contradictions in this point could be explained by the presence of the diffraction replicas and by strong influence of excitation energy dependent matrix elements on photoemission spectra from the $(\pi, 0)$ region [10,13]. On the other hand, by analyzing changes in the line shape upon varying experimental parameters linked to the matrix elements, one gains insight into the nature of a given feature. For instance, the observation that the $(\pi, 0)$ superconducting-state PDH line shape turned out to be insensitive to an alteration of the polarization conditions at a fixed photon energy [2] has been one of the experimental cornerstones of many of the single-band theories developed to explain the PDH line shape in the superconducting state. In this Letter, we utilize a possibility to significantly alter the photoemission matrix elements by variation of the excitation energy. In this context, it is interesting to note that the PDH spectra reported in all intensively referenced publications over the last ten years have been recorded from pristine Bi2212 using only a very narrow range of photon energies (19–22.4 eV) [1–4,7,14].

Figure 2 shows a collection of superconducting-state $(\pi, 0)$ EDCs for Pb-Bi2212 recorded using different excitation energies $h\nu$ (18–65 eV). The black lines show the raw experimental data. Upon a visual inspection of the ARPES data in Fig. 2, it is evident that the PDH line shape can no longer be considered to be dominated by a single-band spectral function with a sophisticated self-energy, as, although at some excitation energies (e.g., 20 eV) all three components (peak, dip, and hump) are present, there are also photon energies at which there is virtually no dip (25 or 42 eV), no hump (50 eV), or even no peak (39 eV). A further, firm conclusion which can be made from inspection of the raw data alone is that the hump and the dip in the EDC cannot be considered as features appearing with fixed binding energies for all $h\nu$ (e.g., compare the EDCs for $h\nu$ 20 and 37 eV). In the many-body scenario for the superconducting state $(\pi, 0)$ PDH line shape, both the peak and

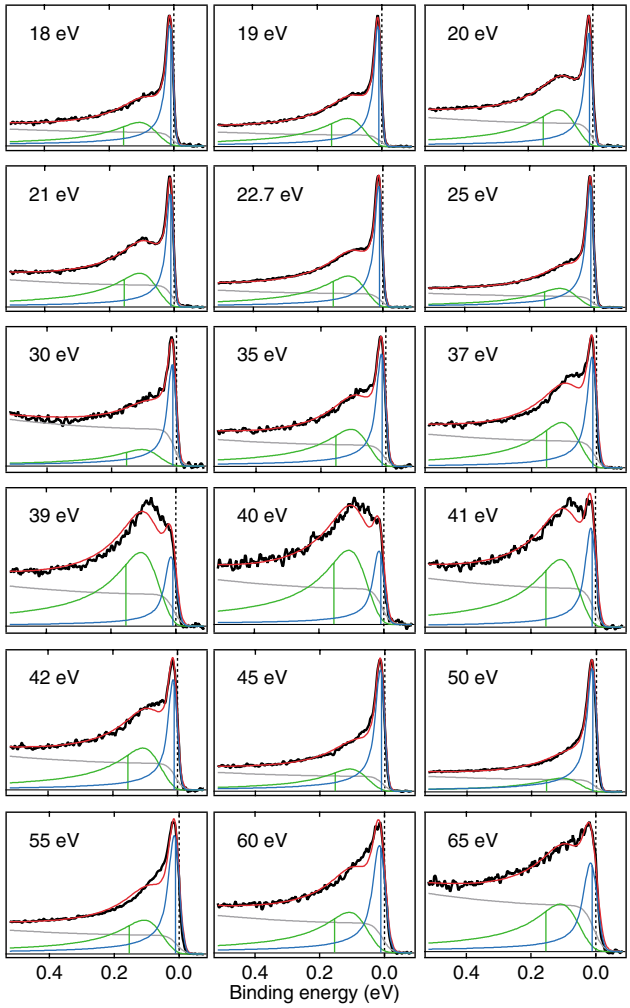

FIG. 2 (color). The $(\pi, 0)$ photoemission spectra from the superconducting state of an overdoped (69 K) sample for different excitation energies: the black lines show the experimental data and the colored lines represent the results of a fitting procedure described in the text.

dip originate from only a single spectral function [4,5] and are connected to significant changes in the imaginary part of the self-energy on going into the superconducting state. Consequently, within this picture, they cannot behave differently to each other upon varying an external parameter to which the spectral function is insensitive, such as the excitation energy. Thus, these experimental observations on their own—without any further interpretation—argue strongly against self-energy effects in a single spectral function as the dominant process determining the PDH line shape. We note that recent intrinsic tunneling studies also present strong evidence in favor of a separate origin for the peak and hump features seen in the conductance curves [15] (although there are conflicting opinions in this regard [16]).

In order to take the next step of trying to identify the dominant causes of the PDH line shape, we have applied a simple fitting procedure to the raw data. If a single spectral function—regardless of the form of the self-energy—

cannot explain the $h\nu$-dependence of the line shape, then it is natural to start by discussing a two-peak fitting procedure. This is also intuitive, bearing in mind the recent experimental observation of the bilayer splitting of the Cu-O-related bands in low temperature ARPES data of Bi2212 [17–19]. The results are shown in Fig. 2. In the fit (shown in red), the spectrum $I(\omega)$ is composed of two spectral features residing at different binding energies (ε_b and ε_a for the bonding and antibonding bands, respectively), whereby both possess identical single-particle spectral functions $A(\omega)$:

$$I(\omega, T, h\nu) \propto \{[M_a(h\nu)A(\omega, \varepsilon_a, T) + M_b(h\nu) \times A(\omega, \varepsilon_b, T)]f(\omega, T)\} \otimes R_\omega + B(\omega, T), \quad (1)$$

$$A(\omega, \varepsilon, T) \propto \frac{|\Sigma''(\omega, T)|}{(\omega - \varepsilon)^2 + \Sigma''(\omega, T)^2}, \quad (2)$$

where $\Sigma''(\omega, T) = \sqrt{(\alpha\omega)^2 + (\beta T)^2}$ is the renormalized [20] imaginary part of the self-energy with $\alpha = 1.1(1)$, $\beta = 2$ [21] and the temperature T in energy units, $f(\omega, T)$ is the Fermi function, $B(\omega)$ is the background which we approximate (assuming it to be $\mathbf{k}$ independent) by taking an EDC from the $(\pi/2, \pi/2)$ point in the empirical form $B(\omega, T) \propto (1 + b\omega^2)f[\omega - \Delta_b(T), T + T_b]$ with $b = 1\ \mathrm{eV}^{-2}$, $\Delta_b(30\ \mathrm{K}) = 5$ meV, and $T_b = 90$ K. The background could be $h\nu$-dependent, but unless this dependence was very dramatic, the influence on the peak positions and relative intensities is marginal. The components of each spectrum are shown in Fig. 2 as thin solid lines: gray lines represent the background, green and blue lines represent the photocurrent from the ''bonding'' and ''antibonding'' bilayer bands, respectively.

Before going further, we wish to stress three points:

(i) The position of the renormalized band, ε (indicated in each fit as a solid vertical line), does *not* coincide with a maximum in the EDCs, ω_m. This is most clearly seen for the bonding peak where $\omega \gg T$, and one can evaluate

$$\varepsilon = \omega_m\sqrt{1 + \alpha^2} \quad (3)$$

(alternatively, one can write $\varepsilon_{\mathrm{bare}} = (1 + \lambda)\varepsilon$ for the bare band position [20]).

(ii) Although the superconducting gap Δ can be included in the dispersion, with ε then becoming $\sqrt{\varepsilon^2 + \Delta^2}$, Δ cannot exceed the ε_a value for the antibonding band. As described below, ε_a is 11 meV, which is close to Δ derived in the weak-coupling BCS scheme for the T_c of the sample.

(iii) The model upon which the fit is based and, in particular, the self-energy function are simple and represent only one choice from myriad possibilities. In the low-energy region the superconductivity has been taken as having no effect on Σ'', and no coupling to any low-energy bosons was taken into account (this simplification will be returned to later). Given that the spectral form of feature ''a'' depends upon the low-energy properties of the self-energy function and that the width of this narrow feature is partially determined by the resolution, it is not possible to extract reliable information on the low-energy behavior of Σ'' from the fit. On the other hand, from the spectral form of feature ''b,'' one can reliably obtain that Σ'' at higher binding energies is linear in ω, not only along the diagonal of the BZ (nodal line) [10] but also at the $(\pi, 0)$ point. Having said that, the exact functional form of the self-energy is not the issue at stake here: the point is that emission from *two* separate electronic states is required to even get close to accounting for the strong $h\nu$-dependence of the PDH line shape.

As a first step, the parameters $\alpha = 1.1(1)$, $\varepsilon_a = 11(1)$ meV, $\varepsilon_b = 154(4)$ meV, M_a, and M_b were fixed in the fit using the spectra with $h\nu = 19$, 20, and 21 eV, and then all other spectra were fitted using only two free parameters: M_a and M_b. These intensity prefactors—which we refer to in the following as ''matrix elements''—are given in the upper panel of Fig. 3 as functions of $h\nu$ and clearly show that the global assignment of the peak and hump in the PDH spectra to two different electronic states is justified. We wish to make it clear that as the spectral functions used in the ''toy model'' are quite possibly oversimplified and as there was no $h\nu$ for which either M_a or M_b were exactly zero, we cannot exclude that each individual spectral function has in reality a small, residual PDH-like line shape due, for example, to many-body effects in the self-energy [22]. However, the crucial point is that the easily visible, ''large scale'' PDH $(\pi, 0)$ line shape and its excitation energy dependence at higher doping levels are not dominated by many-body effects, but rather by the presence of two bilayer-split Cu-O related bands.

Turning back to Fig. 2, one can easily notice that while at excitation energies spanning the whole range (18–30, 50, and 65 eV) the two-band fit is very good; at other energies there are small deviations within the binding energy range from 50 to 100 meV. As regards these deviations, one could consider the following two extremes: (i) the toy model is too simple and the introduction of many-body effects in the model could lead to a change in line shape for the antibonding and/or the bonding component which may then be able to fit the data, and (ii) the $h\nu$-dependence of the deviation from the two-band fit forces us to conclude there must be a third structure which has a *different* photoionization cross section than either that of the bonding or antibonding bands. It turns out that a three component fit perfectly coincides with the experimental data for all photon energies [$\varepsilon_a = 11(1)$ meV, $\varepsilon_b = 150(4)$ meV, $\varepsilon_c = 75(5)$ meV, with matrix elements M_a, M_b, and M_c shown in the lower panel of Fig. 3]. The fact that the $M_b(h\nu)$ and $M_a(h\nu)$ functions exhibit much the same form for both the two- or three-component fits shows that this result is robust with respect to the introduction of the 3rd peak. Consequently, Fig. 3 provides two pieces of evidence that exactly these two main features originate from the c-axis

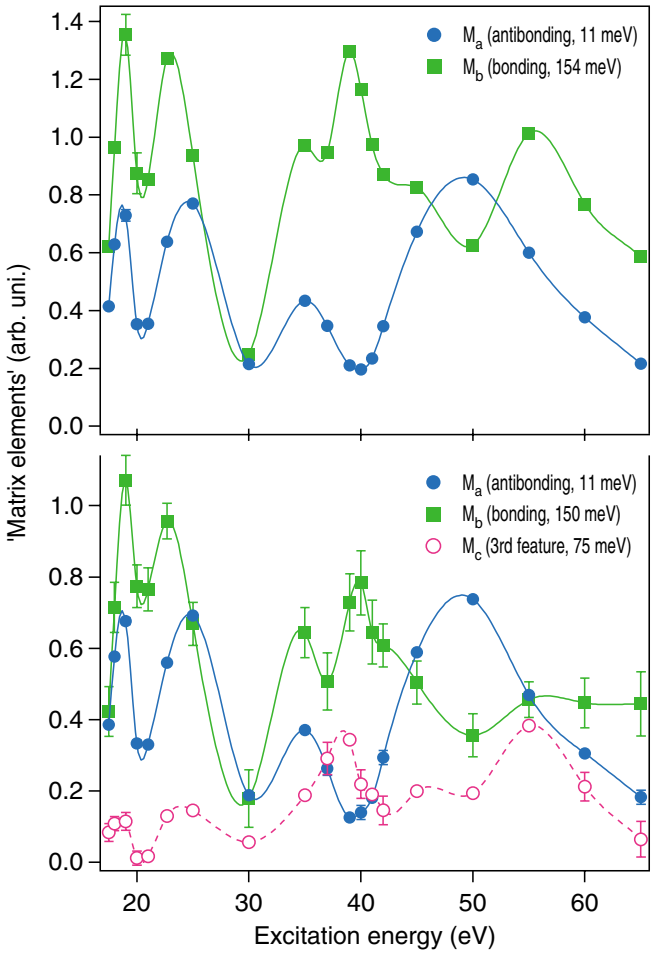

FIG. 3 (color). The intensity prefactors M_a, M_b, and M_c as functions of excitation energy for the two-feature (upper panel) and three-feature (lower panel) fitting procedure.

bilayer-split CuO bands: (1) The $h\nu$-average values of M_b and M_a are comparable ($\langle M_b\rangle/\langle M_a\rangle = 1.2$), as could well be expected from a pair of bands of the same atomic character split by the c-axis bilayer coupling (the average value for M_c is much smaller: $\langle M_b\rangle/\langle M_c\rangle = 2.9$). (2) There are remarkable coincidences between the measured data (Fig. 3) and recent calculations (see Fig. 3 in Ref. [23]) as regards both the minima at 20 and 28–30 eV, the maxima at 18 and 23 eV, the a band minimum at 40 eV, as well as the relative intensities of these bands in the 30–40 eV range. Further support for the antibonding/bonding assignment comes from the fact that the energetic separation between the renormalized bands' positions is about 140 meV, which leads to a difference between the peak maxima in the EDCs of 65–85 meV, in keeping with the normal state bilayer splitting observed recently in [24].

Finally, we note that we have also observed a similar strong photon energy dependence of the $(\pi, 0)$-photoemission line shape for underdoped samples, but that case requires more sophisticated theoretical analysis.

To sum up, we have presented a detailed investigation of the superconducting-state $(\pi, 0)$-photoemission spectra of overdoped, modulation-free Pb-Bi2212 single crystals. We demonstrate that the PDH line shape is strongly dependent on the excitation energy, which is practically irreconcilable with models in which the peak, dip, and hump are considered to stem from a single-band spectral function. The line shape of the spectra can be quantitatively reproduced by the superposition of spectral features described by essentially identical single-particle spectral functions residing at different binding energies: the hump is mainly formed by the bonding and the peak originates from the antibonding component of the bilayer-split Cu-O-derived bands.

We acknowledge stimulating discussions with O. K. Andersen, S.-L. Drechsler, H. Eschrig, A. N. Yaresko, and D. S. Dessau and thank B. Keimer for the pure Bi2212 crystals, S. Cramm for technical assistance, and the DFG and Fonds National Suisse de la Recherche Scientifique for support.

[1] D. S. Dessau *et al.*, Phys. Rev. Lett. **66**, 2160 (1991).
[2] H. Ding *et al.*, Phys. Rev. Lett. **76**, 1533 (1996).
[3] M. R. Norman *et al.*, Phys. Rev. Lett. **79**, 3506 (1997).
[4] J. C. Campuzano *et al.*, Phys. Rev. Lett. **83**, 3709 (1999).
[5] A. V. Chubukov and D. K. Morr, Phys. Rev. Lett. **81**, 4716 (1998); A. Abanov and A. V. Chubukov, *ibid.* **83**, 1652 (1999).
[6] A. Lanzara *et al.*, Nature (London) **412**, 510 (2001).
[7] D. L. Feng *et al.*, Science **289**, 277 (2000).
[8] J. Orenstein and A. J. Millis, Science **288**, 468 (2000).
[9] S. V. Borisenko *et al.*, Phys. Rev. Lett. **84**, 4453 (2000).
[10] S. V. Borisenko *et al.*, Phys. Rev. B **64**, 094513 (2001).
[11] A. A. Kordyuk *et al.*, Phys. Rev. B **66**, 014502 (2002).
[12] H. Fretwell *et al.*, Phys. Rev. Lett. **84**, 4449 (2000).
[13] S. Legner *et al.*, Phys. Rev. B **62**, 154 (2000).
[14] See, e.g., H. Ding *et al.*, Phys. Rev. Lett. **78**, 2628 (1997); A. G. Loeser *et al.*, Phys. Rev. B **56**, 14 185 (1997); A. V. Fedorov *et al.*, Phys. Rev. Lett. **82**, 2179 (1999).
[15] V. M. Krasnov *et al.*, Phys. Rev. Lett. **84**, 5860 (2000); **86**, 2657 (2001).
[16] See, e.g., J. F. Zasadzinski *et al.*, Phys. Rev. Lett. **87**, 067005 (2001).
[17] D. L. Feng *et al.*, Phys. Rev. Lett. **86**, 5550 (2001).
[18] Y.-D. Chuang *et al.*, Phys. Rev. Lett. **87**, 117002 (2001).
[19] P. V. Bogdanov *et al.*, Phys. Rev. B **64**, 180505 (2001).
[20] Σ' is included in both Σ'' and the band position ε as a result of renormalization. If, for simplicity, one assumes $\Sigma'(\omega) = -\lambda\omega + o(\omega^3)$ $(\lambda > 0, \omega > 0)$, then such a renormalization can be done using a simple scaling procedure: $\varepsilon/(1+\lambda) \to \varepsilon$, $\Sigma''/(1+\lambda) \to \Sigma''$, $(1+\lambda)A \to A$.
[21] α and β are similar to values derived from a similar analysis of EDCs recorded along Γ-(π, π) [10].
[22] A kink in the dispersion (A. D. Gromko *et al.*, cond-mat/0202329) and a weak PDH line shape of the bilayer-split bonding band (A. D. Gromko *et al.*, cond-mat/0205385) have been observed very recently near $(\pi, 0)$ in overdoped Bi2212.
[23] M. Lindroos *et al.*, Phys. Rev. B **65**, 054514 (2002).
[24] Y.-D. Chuang *et al.*, cond-mat/0107002.