Direct Observation of Fermi Surface in $YBa_2Cu_3O_{7-\delta}$

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We have performed a high-precision measurement (5×10^8 coincidence counts) of the basal-plane electron-positron momentum density in well oxygenated, twin-free, single crystals of $YBa_2Cu_3O_{7-\delta}$. The raw, processed, and **k**-space reduced spectra unambiguously show a clear image of a major Fermi surface sheet. The form and profile of that image are in substantial quantitative agreement with theoretical predictions of a Γ-X electron ridge section associated with states in the Cu-O chains.

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In the last two years a considerable amount of scientific effort has been directed at elucidation of the nature of the electronic ground state and of the elementary excitations in the ceramic high-temperature superconductors. Direct experimental observation of a Fermi surface (FS) in these materials would place a severe constraint on the choice of theoretical models of high-temperature superconductivity. Angle-resolved photoemission measurements have provided evidence for a variety of FS sections in $YBa_2Cu_3O_{7-\delta}$ (YBCO) [1] and $Bi_2Sr_2CaCu_2O_8$ [2] at specific sampling momenta. Novel de Haas–van Alphen studies in YBCO [3] have disclosed frequencies consistent with a small closed FS orbit predicted by band calculations and associated with states predominantly on the Cu-O planes. The positron ACAR (angular correlation of annihilation radiation) technique is much less sensitive to surface effects than is photoemission and can, in principle, expose an overall FS topology in a single spectrum measurement. However, the interpretation of the ACAR results has, thus far, been complicated by the limited statistical precision of much of the data and, in YBCO, by the use of twinned crystals. The results [4–8] have been in reasonable agreement in regard to the grosser features of the as-measured spectra but have differed markedly in respect to the identification of finer structures in the processed spectra and their interpretation. This paper reports an ultrahigh-precision ACAR study of essentially twin-free YBCO crystals with results that amount to one of the most clear-cut demonstrations of a major FS section in an electronically complex system as yet obtained with this technique.

The measurements were performed with the new University of Texas at Arlington 2D-ACAR spectrometer on a planar assembly of six small crystals of YBCO which had been rendered essentially twin free by annealing under compression along the a direction [9]. The crystals were of high quality and were free of twinning over more than 90% of their area as determined by observation of the residual twinned regions under polarized light and confirmed by x-ray diffraction. The dc magnetic susceptibility of all the samples was separately measured in a SQUID magnetometer to determine the onset and width of the superconducting transition. All showed a transition onset at a temperature greater than 92 K. For four, the transition width was narrow with the midpoint at 90 K or above. In the remaining two the width was broader with midpoints at 85 K and completed transitions at 80 and 70 K, respectively. These data indicate that the level of oxygenation was uniformly high, $\delta\sim6.9$, for all but isolated portions of two of the samples. To make the sample assembly, the six crystals, each approximately 1 mm×1 mm [(001) face] by 50 μm thick, were "tack" mounted, with small deposits ($\sim$50 μm dimension) of epoxy adhesive, onto a "substrate" comprising eight 25-μm-diam W wires strung, under tension, into the form of a 3 mm×3 mm net of very low areal density. Then by a process of repeated attachment and reattachment, guided by the results of Laue-back-reflection x-ray photography, the crystals were brought into a common crystal orientation to within 0.5° with respect to both the azimuthal and the polar angles (referred to the [001] direction). The completed assembly was finally mounted in the apparatus with the (mean) [001] direction parallel to the major (integration) sample-detector axis and with the resolved momentum components p_x along [100] and p_y along [010]. The subsequent spectrum accumulations (set 1) were made on a 256×256 matrix of 0.143×0.143 mrad cells, at an instrumental angular resolution of 0.72 mrad (to 0.76–0.80 mrad when the effect of positron motion is included) and with the specimens at 300 K under a vacuum of 10^{-6} torr. The developing spectra (coincidence counting rate $\sim$80 counts/s) were down-loaded approximately every 2×10^7 accumulated counts to monitor for electronic drift and to guard against data loss arising from any major equipment malfunction. At a total count of 2.5×10^8 the two position-sensitive γ-ray detectors were rotated through 90° and the same pattern of measurement repeated with p_x along [010] and p_y along [100] (set 2).

Throughout the experiment the symmetry and the cen-

ter of each partial spectrum were determined by a variety of different procedures including the visual examination of the anisotropic residuals obtained by subtracting from each spectrum its computed angular average. Within the statistics all the spectra were identical and, in light of that, were added together channel by channel to create two consolidated raw spectra each $\sim 2.5\times10^8$ total counts. The usual correction for the finite aperture and other characteristics of the detectors was then applied and the corrected spectra were examined again by way of their angular averages and anisotropies. They too were identical. Both showed a close to perfect reflection of the expected D_2 symmetry, the sole deviation amounting to a very slight skewness of an amplitude less than the statistical uncertainty at even the largest measurement angles. That established, each was folded about the two major axes; the set-2 spectrum was rotated by 90° and added to that for set 1 thereby creating a final data set with 5×10^8 counts in each of its four equivalent quadrants.

Throughout the processing two main types of structure, already apparent in all the anisotropies of the as-measured spectra, became more clear and regular in the progressively more precise intermediate and final (Fig. 1) images. The first, the large closed areas of significant positive and negative excursion, often appear in this type

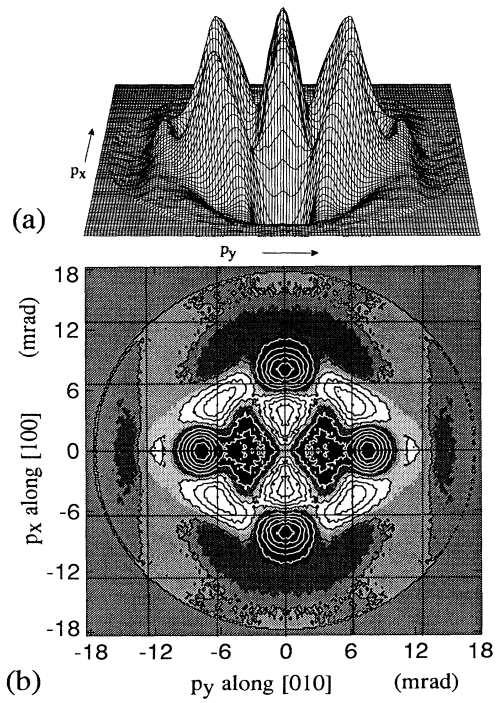

FIG. 1. (a) Isometric and (b) gray-scale-plus-contour plots of the anisotropy of the final spectrum. For clarity the data in (a) have been smoothed and rebinned into a 128×128 array. In (b), white is high, black is low, and the major symmetry lines in the Γ-X and Γ-Y directions are also illustrated. The circle defines the outer limit of the isotropic subtraction processes.

of anisotropy presentation. These structures are, as can be easily verified by simple addition of the two sets of originally orthogonal data, fully consistent with those observed in our earlier study of twinned specimens and bear an essentially similar interpretation in terms of wave-function effects [7]. More remarkable, and not seen in the earlier results, were two close-to-straight line discontinuities running parallel to the Γ-X direction and symmetrically disposed close to p_y (the [010] direction) $= \pm 4\pi/b$. These were apparent in all of the as-measured partial spectra and clear in the two intermediate, consolidated spectra. In the final symmetrized spectrum the equivalent discontinuity is clear and statistically significant ($>3\sigma$) in each and every single (0.14 mrad) section throughout the range -8 mrad $< p_x < +8$ mrad. Deduced parameters defining the discontinuity around selected points are listed in Table I. The sharpness of the break (at Γ consistent with the convolution of a vertical step with the estimated total 0.76–0.80 mrad resolution), its continuity over a range of ~ 24 mrad, and the symmetrical disposition in the original unsymmetrized spectra all point to a FS. All these features are quite remote from those expected of wave-function effects.

That these breaks mark a FS supports a considerable body of theoretical work. A number of recent calculations [10–12] are in substantial agreement in regard to the electronic band structure and the quasi-two-dimensional FS of YBCO. All predict (i) two large "barrel" hole sections centered on S [of area ~ 0.6 and ~ 0.7 of that of the projected Brillouin zone (BZ)], (ii) a further and smaller closed hole pocket at S (of area ~ 0.01 or less of that of the BZ), and (iii) an open electron "ridge" (width ~ 0.15 a.u.) centered on and running roughly parallel to the Γ-X direction. The barrel sections arise from bands whose states are predominantly on the Cu-O planes and the remaining pair from states on the Cu-O chains. Further calculations of the corresponding electron-positron momentum density [13,14] and of the related electron-positron **k**-space density [15,16] are also in substantial agreement in that both predict poor definition of the sections (i) (because the positron resides predominantly on the chains) and (ii) (because that section is small and may not even exist at small levels of oxygen depletion) and the potential for a clear manifestation of section (iii) in the measured spectra.

TABLE I. The mean height H, mean width (0.1–0.9 level) W, and reduced wave vector k_y for selected points on the line discontinuity of the spectrum of Fig. 1.

p_x (mrad)	p_y (mrad)	H (σ units)	W (mrad)	k_y (mrad)
−0.2–0.2	13.1 ± 0.1	12	0.8 ± 0.1	0.6 ± 0.1
6.1–6.5	12.8 ± 0.2	9	1.2 ± 0.1	0.3 ± 0.2
12.3–12.7	13.1 ± 0.1	4	0.8 ± 0.1	0.6 ± 0.1

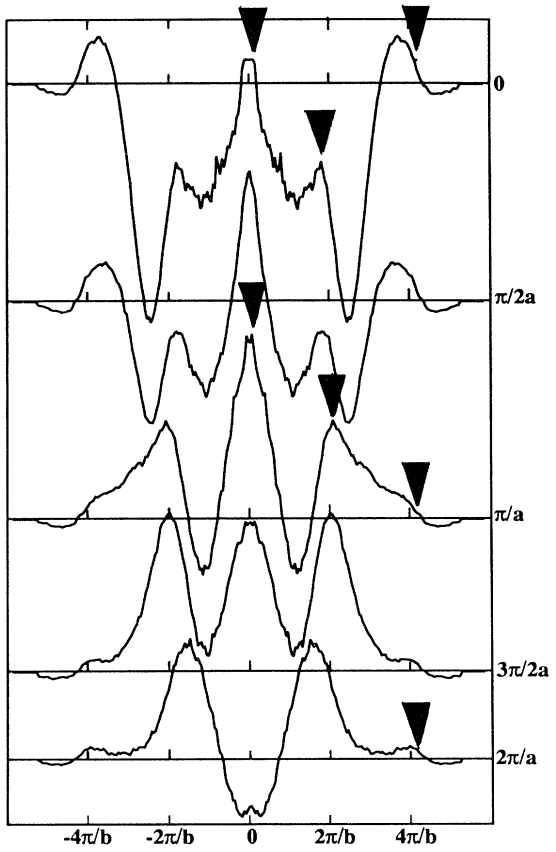

FIG. 2. Integrated (over 0.74 mrad) sections through the anisotropy spectrum of Fig. 1. The positions of the principal discontinuities predicted by Bansil, Mijnarends, and Smedskaer [14] for the FS ridge are marked by the vertical arrows. $2\pi/b = 6.25$ mrad.

The present results are consistent with this overall theoretical picture. The major, wave-function-derived anisotropies are in close agreement with those predicted by Bansil, Mijnarends, and Smedskjaer [14]. More importantly the line discontinuities in the anisotropy plots of Fig. 1 are of the same form and are at almost exactly the same positions as those arising from the FS ridge that appear near $p_y = \pm 4\pi/b$ in all the relevant sections of the theoretical momentum density. These discontinuities are particularly clear because they occur in outlying regions of the momentum density where the wave-function anisotropies are small. But other manifestations of the same FS section at lower momenta can also be found, close to the lines $p_y = 0$ and $p_y = \pm 2\pi/b$. Here, again (Fig. 2), there is good correspondence with the more clear-cut of the relevant discontinuities in the published theoretical sections [14]. The precise shape of the electron-ridge section varies from calculation to calculation but the image in Fig. 1 and the data of Table I both suggest a small dispersion in p_y ([010]) and a variation in

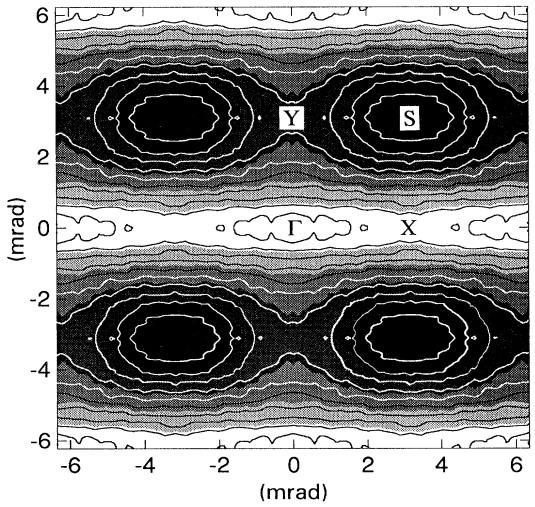

FIG. 3. Electron-positron **k**-space density. White is high, black is low. The high-symmetry points are indicated in the top right quarter.

the sharpness of the discontinuity (sharpest near Γ, least sharp near X) that could well be a manifestation of further dispersion in p_z ([001]), the integration direction. In both cases the suggested dispersion is consistent with that proposed by Pickett, Cohen, and Krakauer [17].

We have found a clear image of the electron ridge but we have found no evidence of the other predicted FS sections in any of our raw or symmetrized momentum spectra. So clear an image of FS is nevertheless rare in essentially unprocessed momentum data and is more usually found only after that data have been transformed into a **k**-space density by the LCW procedure [18]. We applied that transformation to the final corrected and symmetrized data set after first subtracting an isotropic substrate (wire plus epoxy) spectrum whose form and intensity (20%) had been determined in a subsidiary experiment [19]. The essential results (Fig. 3) are the expected remapping of the original **p**-space discontinuities into a central electron FS ridge and the emergence of a major depression centered on S. The image of the electron ridge is now clear, with a FWHM (marked by the extremities of the white region) ~ 1.3 mrad and a profile consistent with the resolution. The apparent p_y and p_z dispersions are as deduced above. An interpretation of the remaining depression around S as simply the smeared, composite image of the remaining three FS hole sections is tempting. But that interpretation cannot be entirely justified, for (i) there is no evidence of the sharper discontinuities already found for the ridge, and (ii) roughly similar depressions also result when the angular average of the same momentum density is identically transformed. On the other hand, the general form of the LCW distribution is compatible with theoretical predictions [15,16] when the effects of wave funtions, of oxy-

genation, and of resolution are taken into account.

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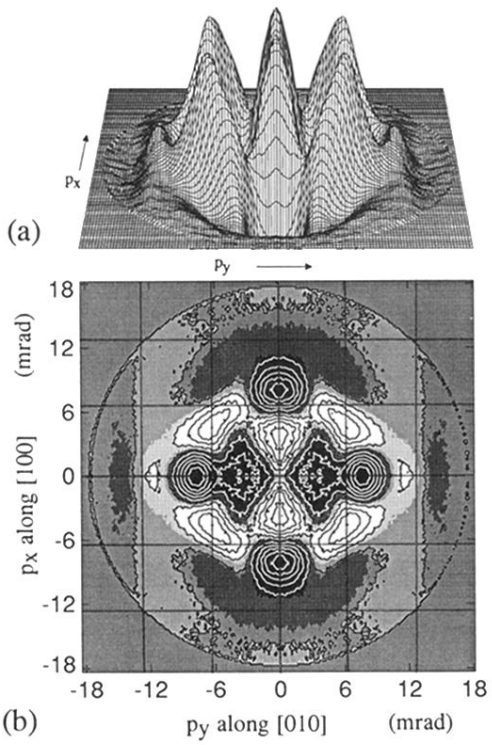
[19] The measured background was accurately isotropic and could not therefore have contributed to the anisotropy of the momentum data and was accordingly not considered or included in that analysis or the associated interpretation. It must, and indeed does, distort the LCW **k**-space data. Hence its inclusion. Any uncertainty in the estimation of its proper intensity ($\pm 5\%$) has only a marginal effect and does not change the conclusions.



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