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A positron study of the electronic structure of $YBa_2Cu_3O_{7-\delta}$

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Abstract. We have measured the two-dimensional electron–positron momentum density (resolved momentum components along (100) and (010)) for single crystals of $YBa_2Cu_3O_{7-\delta}$ at a significantly higher level of statistical precision for this orientation than that embraced in earlier published works. We have found in our data structures suggesting that the positron annihilate predominantly on oxygen sites in the copper–oxygen chains. These structures are also present in the results of a companion ligand field calculation and arise from spatial variations in the electron and positron wavefunction overlaps. Even at the present level of statistical precision, we are unable to argue for or against the existence of a Fermi surface.

1. Introduction

The discovery of high-temperature metal oxide superconductors has prompted much debate as to the nature of the electronic structure of these and related compounds. The current theories of the two mechanisms underlying the superconductivity in these materials fall into two main classes. One class assumes there is a conventional but quasi-two-dimensional Fermi liquid whilst the other group relies on a Mott–Hubbard-type split-band picture. The theories are complex but the essence of the debate can be most succinctly expressed in the single question 'Is there or is there not a Fermi surface?'

Among the contemporary electronic structure techniques the two-dimensional angular correlation of positron annihilation radiation (ACAR) measurement (for recent reviews see Berko 1979, Mijnarends 1983) can provide the most direct view of the Fermi surface in a wide variety of conditions. The measured two-photon angular correlation curves reflect the momentum-space density, $n_{ep}(\boldsymbol{p})$, of the electrons sampled by injected positrons and contain information about the occupied regions of k-space (and thus the Fermi surface), the nature of the participating electron and positron states and some aspects of the many-body interactions between the charged positron and the polarisable electron medium. In those situations where the Fermi surface is the predominant interest, the so-called LCW folding procedure (Lock *et al* 1973), which remaps the measured momentum density into an equivalent Bloch-wave (k-space) density, $n_{ep}(\boldsymbol{k})$, has proved very useful. In the k-space density the singularities and other structures that

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reflect the Fermi surface are brought into sharper relief and the single- and many-particle 'wavefunction' effects are relatively less in evidence. In those favourable cases where these residual wavefunction effects are small, and $n_{ep}(\boldsymbol{k})$ closely approximates to the equivalent electron density, $n_e(\boldsymbol{k})$, the Fermi surface parameters can be established (see for example Kaiser *et al* 1987) with a precision approaching that of the longer established magneto-oscillatory methods. In general, however, some wavefunction effects are always present and must be acknowledged in any Fermiological interpretation of an experimental $n_{ep}(\boldsymbol{k})$.

Several LCW analyses of positron spectra for high temperature superconductors have already been made and reported. Tanigawa *et al* (1988) have reported measurements on both undoped and doped superconducting La_2CuO_4 and have identified structures in their transformed k-space spectra as manifestations of Fermi surface. Peter *et al* (1988), Smedskjaer *et al* (1988) and Bansil *et al* (1988) have reported similar studies and conclusions for $YBa_2Cu_3O_{7-\delta}$. The common assumption in these latter works has been that the structures in $n_{ep}(\boldsymbol{k})$ are Fermi surface in origin: the possible contribution of the earlier noted wavefunction residuals thus far would seem to have been largely discounted. This is surprising. It is true that the structures observed in $n_{ep}(\boldsymbol{k})$ are roughly those to be expected from the predictions of Fermi surface offered by existing band structure calculations but there is also a considerable weight of evidence from other positron results that, in compounds, the wavefunction residuals in $n_{ep}(\boldsymbol{k})$ are usually appreciable and, in metallic compounds, can sometimes be so large as to completely obscure the Fermi surface signatures (see, for example, Shiotani *et al* 1986).

In this paper we report and discuss our own results for $YBa_2Cu_3O_{7-\delta}$. In the section that follows we give details of the positron and other complimentary macroscopic measurements on the single crystal specimens. In § 3 we discuss the positron results and their analyses in the context of complimentary LCAO–MO calculations. Finally, in § 4, we present our conclusions.

2. Experimental details

The $YBa_2Cu_3O_{7-\delta}$ single crystals were grown by a flux technique (Ong *et al* 1988) that results in high-quality single crystals which are superconducting without the need for subsequent oxygen annealing. The crystals, in common with all those prepared in this manner, showed a T_c between 90 and 93 K with a transition width ΔT less than 0.5 K and a strong and abrupt field cooled Meissner effect. Measurements of these parameters before and after the positron study, during which the specimens were held under vacuum for five weeks, showed no more than minor differences suggesting that there had been little loss of oxygen from the samples while they were under vacuum.

The positron measurements were performed on the UETA two-dimensional angular correlation spectrometer a full description of which has been given elsewhere (West *et al* 1981). Four approximately 1 mm × 1 mm (001 face) and 60 μm thick crystals were tiled onto a 3 mm wide by 25 μm thick mylar support strip using vacuum grease as adhesive and brought into a common orientation by aligning (we estimate to within 2°) their twin bands (⟨110⟩ directions) under polarised light. The final assembly was then mounted in the apparatus with the crystal faces perpendicular to the major (integration) sample–detector axis and with the resolved momentum components lying along [100] and [010]. The subsequent spectrum accumulation was made, under a vacuum of 10^{-4} Torr and at a specimen temperature of 310 K, on a 128 × 128 matrix of $(0.34 \text{ mrad})^2$

cells and at an angular resolution of $(0.80\ \text{mrad})^2$. The final spectrum contained ~65 K counts at peak and $\sim 5 \times 10^7$ counts overall. Immediately following this measurement a further background spectrum measurement was made with the specimens removed from the mylar strip but all other parameters as before. This provided a form for the background spectrum. The results of a final series of count rate measurements involving totally (positron) absorbing aluminium samples, of different sizes and geometries mounted on the foil were then used to arrive at an estimate, for the overall intensity of the background, of $12 \pm 2\%$.

3. Data analyses

A preliminary and necessary step in the analysis of any 2D angular correlation spectrum is the removal of the distortions produced by the spatial variation of the single detector efficiencies and the finite aperture of those detectors. This is achieved by dividing the as measured spectrum by a momentum sampling function (or 'correction matrix') which is generated from single detector responses obtained in subsidiary experiments. Once this is done, the corrected spectrum can be examined in terms of its symmetries and other structures. Where those symmetries are clear and compatible with the point symmetry of the crystal, they can be and often are invoked by folding the spectrum about the appropriate rotation and/or reflection axes thereby achieving an effective increased statistical precision.

Even during the intermediate stages of the present measurement it was clear that the spectrum was almost isotropic and structureless. In such a case, it is paramount that the correction, symmetry folding, and any other spectrum refining procedures be carried out with extreme care. All of them involve parameters (such as correction centres, rotation centres, reflection axes, etc.) which are uncertain to some small degree. If these parameters are even slightly in error, extraneous structures and symmetries can be introduced.

In the present work the centres of both measured (data and background) spectra was already defined, by prior alignment and calibration procedures, to within ± 0.1 channels (in both x and y). The correction matrix (for a description see West *et al* 1981) has a statistical precision much greater than that of the specimen spectrum but a centre which is only defined to within ± 0.5 channels. In arriving at the various total and anisotropy spectra presented below we have used an iterative spectrum refining procedure in which these centre uncertainties were used to define a fitting parameter space in which we searched for an optimal solution defined by the following criteria:

(i) Optimal reproduction of the effective C_{4h} symmetry of the twinned crystal as manifested in the major anisotropies (see figure 2 below) of the processed (unfolded) spectrum.

(ii) Minimisation of all other structures.

In establishing criterion (i) we recognised the possibility that the actual symmetry of the data might be lower than C_{4h} due, for example, to inequality of representation of the two lattice orientations in the twinned crystal. However, it is worth pointing out that, during our analyses, we found that the manifestations of C_{4h} symmetry were relatively insensitive to the detailed choice of underlying parameters while most other structures of lower apparent symmetry were strong and rapidly varying functions in the fitting parameter space and had magnitude comparable to that of the statistical noise. But,

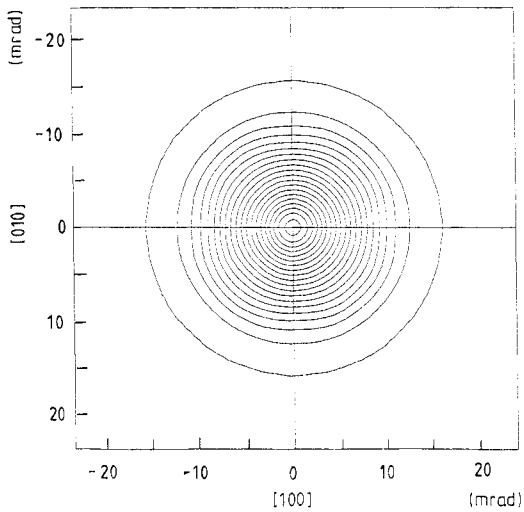

Figure 1. Contour plot of the 2D angular correlation curve for $YBa_2Cu_3O_{7-\delta}$ following subtraction of a 12% background contribution to account for annihilations in the specimen mount, correction for the momentum sampling function of the spectrometer and folding with the symmetry operations of the projected reciprocal lattice of the effective tetragonal structure of the twinned specimens. The scales show the tilt from the central position.

even after this optimisation procedure, we were left with a residual asymmetry. This, a slight inequality in the amplitudes of the major structures centred at the projections of reciprocal lattice points $\boldsymbol{G} = (11n)$ (see below), and antisymmetric in respect of reflection through either of the axes joining the diametrically opposed pairs of those points, persisted through all our analyses. Since the twin boundaries lie along these same directions it is possible that this asymmetry is real and is due to some positron annihilation in those strained interface regions. Nonetheless, since this effect was only of order one standard deviation in the centred corrected distribution, we chose to average it out of our final distribution by imposing the C_{4h} symmetry with folding. In that folding we adjusted the orientation of this distribution within the limits defined by the earlier noted $\pm 2°$ uncertainty in the crystal orientation with the criterion that the structures in the final folded anisotropy curve (figures 2) were the sharpest obtainable. Finally, all centres and axes having now been optimally defined, we generated, for further examination, a full set of fully symmetrised curves.

4. Results and discussion

The spectrum after correction and folding is shown, in the form of a contour diagram, in figure 1. The essential smoothness and comparative lack of structure is immediately obvious. This is a pattern not unfamiliar for positron–electron spectra of compounds comprising a multiplicity of electron bands (see for example Shiotani *et al* 1986; Wachs *et al* 1988) or even for simple oxides, such as NiO, (Wachs *et al* 1989). In order to see the finer details of the spectrum we subtracted 100% of the angular average of the folded

spectrum from itself and so obtained the anisotropy (figures 2(*a*) and 2(*b*)). The major structures in this distribution are the 4 positive excursions or higher momentum contributions (Berko 1979) centred on the (1, 1) points of the projected 2D reciprocal lattice.

It can be shown (Mijnarends 1979) that the electron–positron momentum density, for positrons annihilating from a fully symmetric ground state, carries the full point symmetry of the underlying crystal basis. A major contribution to that symmetry appears in the form of higher momentum components (or HMC). In the spectra for simple metals, an examination of the relative magnitudes of the various HMC (which may be described as the residual manifestations of the translational periodicity of the lattice) can considerably aid in the understanding of the nature of the underlying positron and electron states. In multi-element systems, where the lattice has a non-trivial basis or in any situation where the positrons may be annihilating from localised states of a symmetry far removed from that of the host lattice, an analysis of the HMC is less straightforward but may be even more useful. Here with a twinned orthorhombic ($a \neq b \neq c$) crystal we expect to find, following integration along the longitudinal c-axis, an apparent C_{4h} symmetry and HMC, to some degree, at all points in a projected (square) reciprocal lattice defined by the points $\boldsymbol{G} = (2\pi/a')(mi + nj)$ where $a' = (a + b)/2$ for which the stars will henceforth be denoted by $\{m, n\}$.

Examination of figures 2(*a*) and (*b*) discloses the strong HMC at the points $\{1, 1\}$. Slight indications of positive excursions are also discernible at the points $\{2, 2\}$ and $\{2, 0\}$ but barely above the noise. Most significantly there is little or no indication of appreciable effects at the points $\{1, 0\}$. Were this a single element system, one would expect HMC at all the points of the projected reciprocal lattice. However in a case that the positron wavefunction has preferential overlap with the electrons on one type of atomic site we could expect that the lattice defined by that particularly favoured site would be the one most germane to an HMC discussion. The present results follow this pattern and suggest, through the lack of any appreciable HMC at $\{1, 0\}$, a picture in which the metal ions play a minimal role and the oxygen sublattice is the relevant one. That the positron density should be greatest around the oxygen sites could be argued through considerations of atomic number, charge transfer and (Shiotani *et al* 1986) many-electron effects *a priori*. It is also the picture that emerges from an explicit bandstructure calculation (von Stetten *et al* 1988) and from our own complementary cluster calculations of $n_{ep}(\boldsymbol{p})$.

The momentum space density calculations were carried out within the independent particle model following the cluster approach pioneered by Chiba and Tsuda (1974) and since exploited by Chiba (1976) and others including Akahane *et al* (1985), Wachs *et al* (1988, 1989) and Turchi *et al* (1988). In this approach, the electron wavefunctions for a representative cluster of atoms of the solid are determined within the linear combination of atomic orbital (LCAO) molecular orbital (MO) formalism and the positron wavefunction is approximated using a variational procedure. Since it is clear from positron wavefunction calculations (von Stetten *et al* (1988)) that the positron density is greatest in the copper–oxygen chains, we chose a cluster consisting of a Cu(1) atom in a copper–oxygen chain surrounded by four O atoms; an in-chain O(1) 3.6661 au to either side and an O(4) 3.5064 au above and below (Capponi *et al* 1987). The basis set of electron wavefunctions for this cluster was obtained from Herman–Skillman calculations for each atomic constituent. Here we treated the atoms as ions so as to include the effects of charge transfer in the resulting basis wavefunctions. Specifically we assumed that the charge state of the Cu is 3^+ with a valence electron configuration of $3d^8$ and that of the O atoms is 2^- with a valence configuration of $2s^2\,2p^6$. We then form antibonding combinations of the Cu 3d and O 2s and O 2p basis functions while preserving charge neutrality for the cluster as a

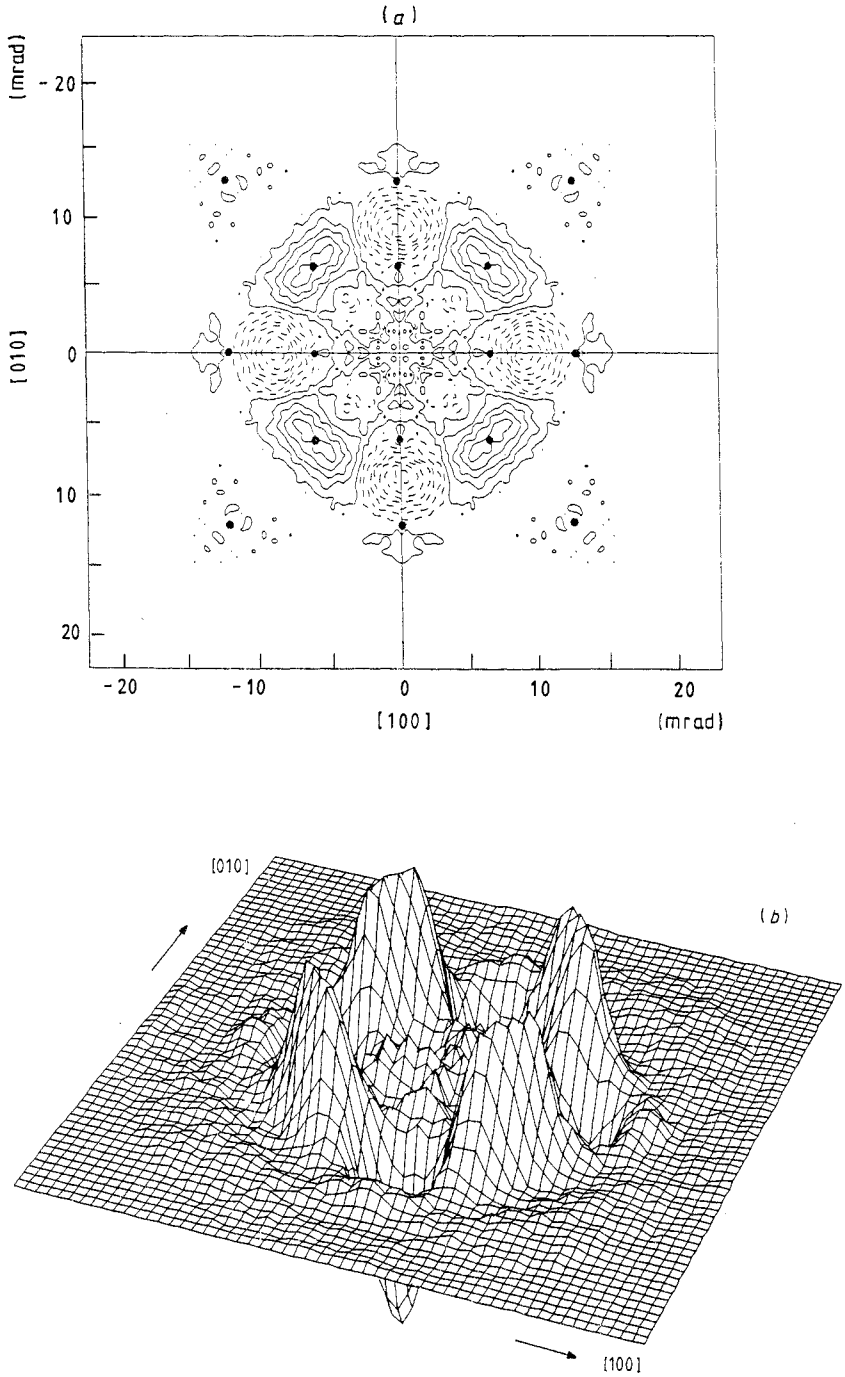

Figure 2. Contour (*a*), and isometric (*b*), views of the anisotropy of the spectrum of figure 1, as obtained by subtracting the angular average of the full spectrum from itself. In (*a*) the full and broken contours respectively represent levels of positive and negative density. The square lattice of superimposed filled circles is the projected reciprocal lattice. The amplitude of the positive excursions at (1, 1) is 2.5% of the peak (central channel) counts of the original spectrum shown in figure 1.

whole. The positron wavefunction was modelled as by Chiba and Tsuda (1974) as a constant amplitude from which is subtracted a superposition of Gaussians centred on each site and with characteristic widths determined by a variational calculation. The potential used for this calculation was obtained from a superposition of Herman–Skillman neutral atom charge densities for an entire $YBa_2Cu_3O_{7-\delta}$ unit cell. We would note here that the positron density resulting from this calculation was most concentrated around the oxygen atoms as expected from both our earlier arguments and the more sophisticated calculation of von Stetten *et al* (1988). The two-dimensional electron–positron momentum-space density was then obtained in the usual way (Berko 1979) from the sum of the Fourier transforms of the various products of the positron wavefunction and the electron wavefunctions.

The anisotropy of the final 2D momentum density for the copper–oxygen chains is shown, in the form of a contour diagram, in figure 3(*a*) and the further result after folding about the [1, 1, 0] direction to simulate the effect of the twinning is similarly presented in figures 3(*b*) and 3(*c*). The dominant and common feature in the data of figure 2 and that of figure 3 is immediately clear. Both possess major maxima centred close to the {1, 1} points. That the two differ in respect of the precise location and shape of these features is not surprising since the calculation is based on a simple cluster model which neglects the long range order and even next-nearest neighbour effects. The comparatively greater angular spread of {1, 1} maxima in the experimental results may nevertheless be significant. If, as earlier and tentatively suggested in § 3 some of the positrons are annihilating at the polarised (Mueller *et al* 1988) twin boundaries, a variety of effects (e.g. of the local crystal symmetry, local strains and, conceivably, regions of incoherence featuring oxygen vacancies) could be expected in the data. Irrespective of this or other possible complications, the agreement between the two curves is sufficient to suggest that the anisotropies in the experimental spectrum reflect, in the main, the spatial distributions of the electron and positron charge densities rather than fine details of the electronic band structure. The same conclusion emerges from the results of our applications of the LCW procedure.

We first applied the LCW transformation (Lock *et al* 1973; Lock and West 1975; Beardsley *et al* 1975), after removal of the 12% background due to the sample mount (see § 2), to the corrected and folded spectrum. The result (figure 4) is an almost completely structureless distribution with an amplitude variation entirely compatible with an interpretation in terms of residual positron and electron wavefunction effects and with no indications of rapid changes indicative of Fermi surface sections. This does not mean there are no Fermi surface effects. It may rather be that, as elsewhere (Shiotani *et al* 1986), those effects are simply obscured by the wavefunction residuals from not only the associated conduction bands but all the many other full bands contributing to the spectrum. The only solution to this problem is a calculation of the wavefunction residuals embracing not only band structure but also those many-body effects (Shiotani *et al* 1986) that may influence the distribution of positron density in the crystal. In the absence of that, the only obvious alternative is to attempt to isolate the more rapid variations in the momentum density by LCW folding the anisotropy rather than the total spectrum with the hope of thereby removing at least some of the wavefunction residuals. This is the approach of Smedskjaer *et al* (1988) which we also have followed, although we feel compelled to remark at this juncture that this procedure is only one of expedience: it has absolutely no theoretical justification in terms of the LCW theorem and the underlying physics.

The result of LCW folding the anisotropy spectrum is shown as figure 5. The form is not dissimilar to the LCW folded total spectrum (figure 4). The anisotropies in the original

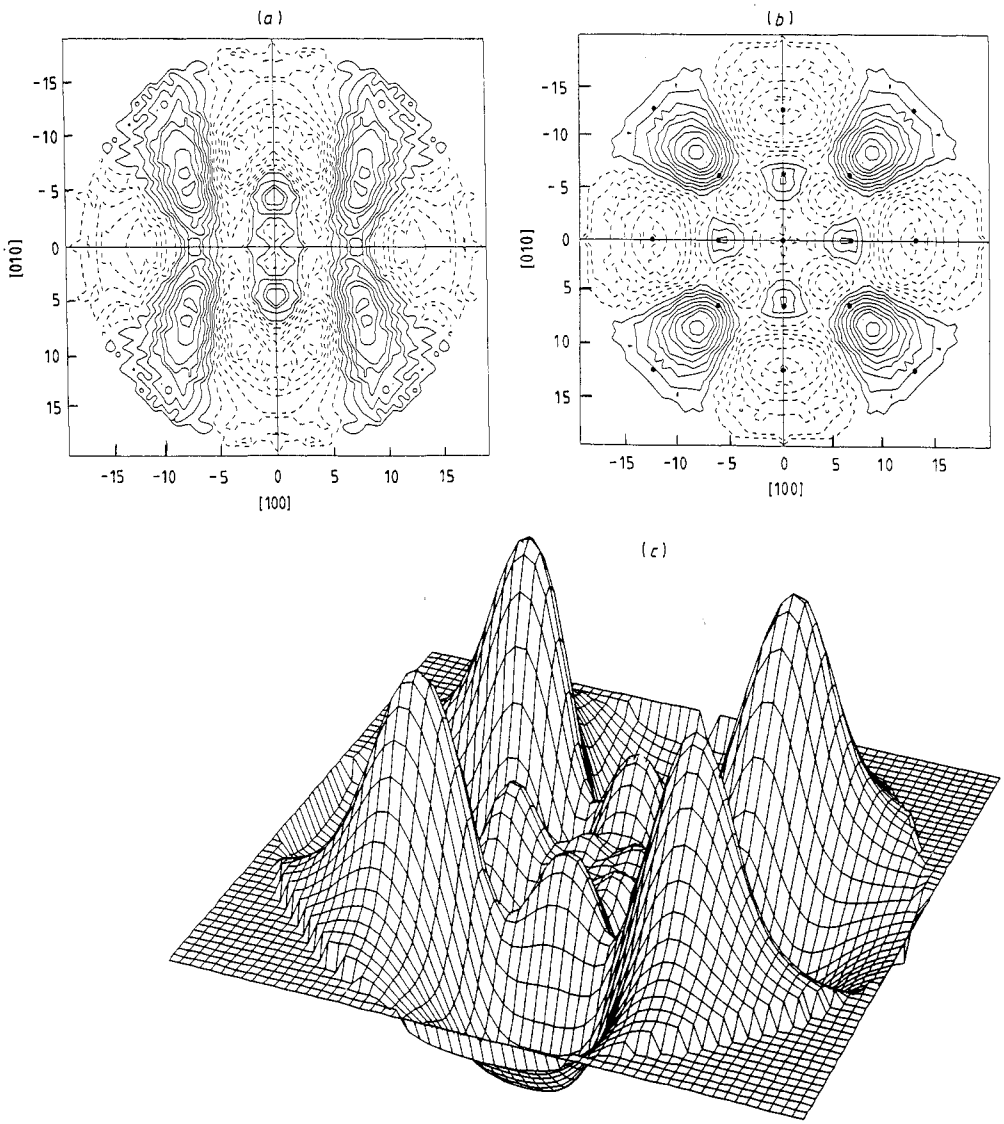

Figure 3. Anisotropy of the cluster calculation of the electron–positron momentum density in the copper–oxygen chains (*a*) before and (*b*), (*c*), after folding along the [110] directions to simulate the effect of twinning. Full and broken contours respectively represent levels of positive and negative density. In (*b*), as in figure 2, the reciprocal lattice points are marked as filled circles. In (*a*), (*b*), units are mrad, as for figure 1.

momentum density are largely eliminated consistent with our interpretation of them as wavefunction effects. Again there is no indication of the rapid changes which could signify the presence of a Fermi surface. This result is in close agreement with that of an almost exactly parallel measurement and analysis recently completed elsewhere (S Berko, private communication). It differs somewhat from the published result of Smedskjaer *et al* (1988) which did show rapid changes in the LCW treated spectrum and which were interpreted in terms of Fermi surface sections. We have also followed Bansil

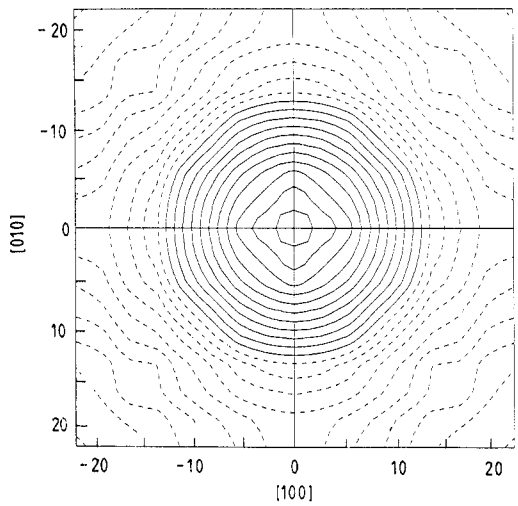

Figure 4. LCW folded spectrum of figure 1 (same units). The total amplitude of variation is 5% of the maximum (at centre). Full and broken contours represent levels of above and below average density, respectively.

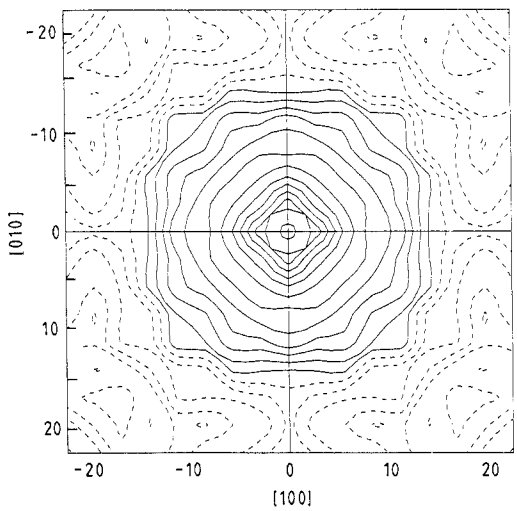

Figure 5. LCW folded anisotropy of figure 1 (same units). The total amplitude of variation is 1% of the maximum (centre). Full and broken contours respectively represent levels of above and below average density.

et al (1988) in computing the first derivative of sections through our measured momentum density in an attempt to identify, by this means, rapid changes in slope that might be indicative of Fermi surface.

Our derivative curves, generated by two-point differencing, are shown, without smoothing, for three high-symmetry sections, in figures 6(*a*), (*b*), and (*c*). In regard to their overall form these curves are in good agreement with the similar derivative plots

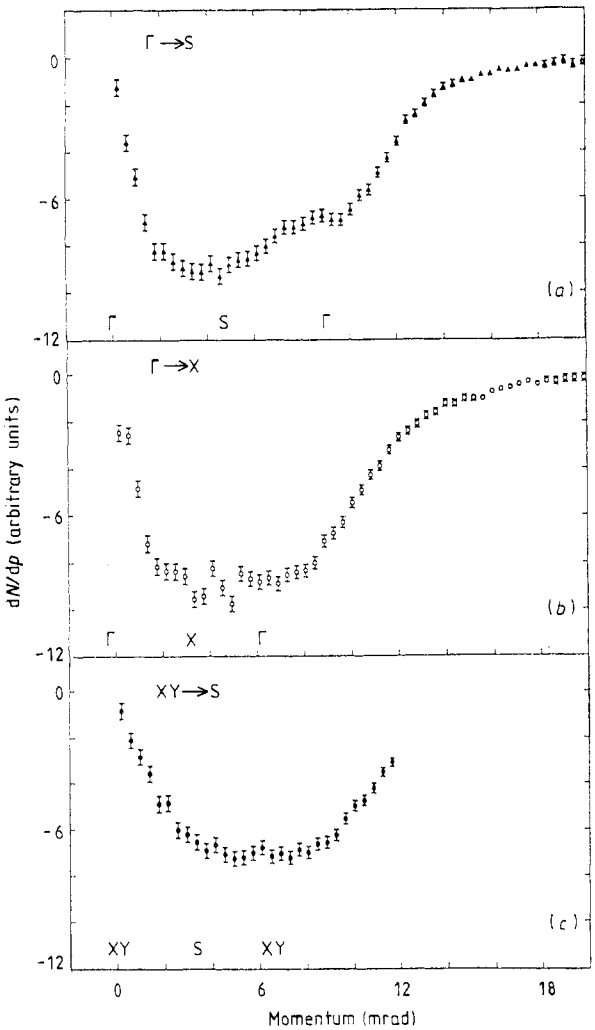

Figure 6. Derivative curves for principle sections of the measured spectrum along (*a*) $\Gamma \rightarrow$ S, (*b*) $\Gamma \rightarrow$ X, and (*c*) XY $\rightarrow$ S.

of Bansil *et al* (1988). In respect of the fine structures they are not. In both figures 6(*a*) and (*b*) there are short-range structures at or around 4 mrad but these are also present in our theoretical curves and, because of that, could not easily be attributed to other than the earlier discussed effects of the electron and positron wavefunctions. We do find other fine structures in some sections but little or no correlation between the structures as we move from section to section as would be the case were those structures to reflect a finite and continuous boundary between occupied and unoccupied regions of k-space.

5. Summary and conclusions

In this paper, we have reported the results of measurements of the 2D electron–positron momentum spectrum in $YBa_2Cu_3O_{7-\delta}$ of a precision significantly greater (approximately a factor of ten in total counts) than any other published results. We have observed

in those results a suggestion of symmetries incompatible with that of the underlying crystal and other specific effects that could suggest some of the positrons, at least, are annihilating in the twin boundaries. The major anisotropies of the measured spectrum reflect the spatial disposition of the copper–oxygen bonds and are consistent with a picture in which the positron–electron overlaps that determine annihilation are predominantly on the oxygen sites. Because of the twinning we are unable to distinguish between annihilations occurring in the copper–oxygen planes and in the copper–oxygen chains, but are prepared to accept the conclusions of von Stetten *et al* (1988) in this respect. We would note in passing that a predominance of annihilation on oxygen sites in the copper–oxygen chains is, in the context of most conventional band structure pictures, particularly favourable in regard to the eventual elucidation of any Fermi surface and/or the further search for understanding of the mechanisms underlying the superconductivity.

We have not, in this work, been able to establish the existence of a Fermi surface in $YBa_2Cu_3O_{7-\delta}$. We are not suggesting there is no Fermi surface but are merely asserting that the combination of the resolution, the sampling density and the statistics of our measurement has not allowed us to resolve one. The measurements of Smedskjaer *et al* (1988) were of lower statistical precision but were made on a finer sampling mesh and at higher intrinsic resolution. It is possible that these differences in the measurement parameters can explain the divergence in results and in the consequent interpretations. Our own analyses, however, suggest that the resolution of the structures predicted by the calculations of Bansil *et al* (1988) will require significantly better statistics. Specifically, our calculations suggest, in the context of an intrinsic angular resolution of the order of 0.7 mrad and a bin width of $(0.16\ \text{mrad})^2$, the need for a measurement embracing at least 5×10^8 total counts in the spectrum.

Acknowledgments

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