Positron annihilation and high-temperature superconductivity

by M. Peter

After a brief review of the theory of positron annihilation techniques and of experimental principles, we give examples of the successful determination of electron momentum density and Fermi surfaces in alkali metals, transition elements and compounds, and cerium. We then discuss the application of positrons in superconducting oxides. So far the best results have been obtained in $YBa_2Cu_3O_{7-x}$, with confirmation of calculated band structure and observation of discontinuity at the Fermi energy.

Introduction

Nearly every speaker at the IBM Europe Institute Workshop on High-Temperature Superconductivity has begun his contribution by expressing his gratitude to Professors Bednorz and Müller for their invitation and for having given such a decisive impulse to the study of superconductivity. The magnitude of their discovery can be inferred from the fact that not only the field of superconductivity but also many other domains in science and technology are currently undergoing rapid change influenced by the breakthrough in superconductivity. Positron annihilation is one of these domains. To some of the new questions raised, positron annihilation may give answers, and in doing do, may improve its own method.

• *Positron annihilation*

Positrons, when brought inside a sample, annihilate with electrons at a rate proportional to the electron density: The interaction cross section is given by the classical electron radius R_{el} [1]:

$$RATE = \pi R_{el}^2 c n_{el} . \quad (1)$$

Here, n_{el} is the local electronic density. More precisely, the rate is proportional to the integral over the product of positron and electron density. The lifetime given by our formula is relatively long, of the order of several hundred picoseconds. Careful studies by several investigators [2] show that in many cases these lifetimes suffice for the positrons to reach thermal equilibrium (within picoseconds) before decay.

This is true for positrons generated by the usual sources (^{22}Na, ^{64}Cu), which are emitted with considerable energies, and *a fortiori* for slow positron beams [3]. The main difference between beams and isotopic sources is the penetration of positrons: In copper oxides, positrons of ^{22}Na penetrate over 100 μm; with slow beams (energy of several volts), penetration can be limited to well-defined surface layers [4].

• *Decay modes*

During positron–electron annihilation, energy, momentum, and angular momentum are conserved. Single-photon decay is thereby prohibited. In the most common mode, a positron combines with an electron of opposite spin to form two photons. The small difference between the momenta of these photons equals the momentum of the electron–positron system. If the momentum of the positron probe is small, or is known, the momentum of the electron(s) can be inferred.

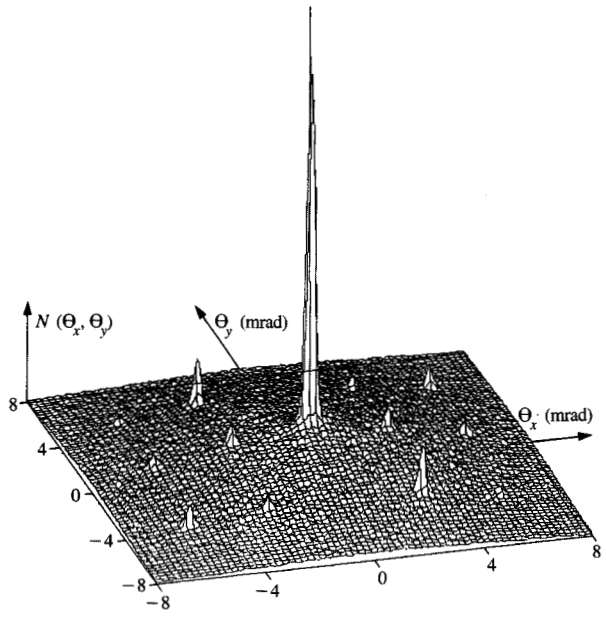

Figure 1

2D-ACPAR of positronium in quartz. Center of gravity momentum is zero (high peak); satellites mark projection of reciprocal lattice points. Soft hill due to core electrons in quartz.

Higher-order modes also exist; they imply decay with the production of an odd number of gammas in the triplet configuration, or an even number of gammas in the singlet configuration. The probability of these processes decreases with powers of the fine-structure constant alpha [5].

The spin-dependence of the process implies that with polarized positrons it is possible to measure the polarization of the electron gas [6]. Positrons from beta decay are partially polarized due to parity violation [7], so it has, for instance, been possible to show the polarization in a single crystal of Ni [8].

The formation of positronium (Ps) is of particular interest. It resembles hydrogen, its momentum distribution showing as a narrow peak. In vacuum, the binding energy of Ps is 6.8 V; in the presence of an electron gas, the binding energy is shielded out. Ps is therefore not found in defect-free metals, but in insulators [9] it is easily observed. Since the center-of-mass momentum of Ps is small, Ps gives rise to a narrow peak in the angular correlation of positron annihilation radiation (ACPAR) which may serve for calibration of the 2D apparatus [10] (**Figure 1**).

A general review of positrons in solids was given by Hautojärvi [11]; a theoretical formulation of the positron in a many-electron system is due to Ferrell [12]. There we find that the positron–electron interaction is given by

$$H(\mathbf{p}) = \int \mathbf{dx} \exp(-i\mathbf{px})\hat{\psi}_-(\mathbf{x})\hat{\psi}_+(\mathbf{x}). \tag{2}$$

In the independent-particle model (IPM) this leads to an angular momentum density distribution, such as that measured by positron annihilation followed by 2γ emission:

$$\rho^{2\gamma}_{\mathrm{IPM}}(\mathbf{p}) = const \sum_j \left| \int \mathbf{dr} \exp(-i\mathbf{pr})\psi_{-_j}(\mathbf{r})\psi_+(\mathbf{r}) \right|^2. \tag{3}$$

In a periodic crystal with Bloch functions, this can be written as

$$\rho^{2\gamma}_{\mathrm{IPM}}(\mathbf{p}) = const \sum_{\mathbf{k},j} \sum_{\mathbf{G}} n_j(\mathbf{k}) |A^{\pm}_j(\mathbf{k}, \mathbf{G})|^2 \delta(\mathbf{p} - \mathbf{k} - \mathbf{G}), \tag{4}$$

with

$$A^{\pm}(\mathbf{k}, \mathbf{G}) = \sum_{\mathbf{G}'} A^{+}(\mathbf{G}')A^{-}(\mathbf{k}, \mathbf{G} - \mathbf{G}'). \tag{5}$$

Here $n_j(\mathbf{k})$ is the occupation of state $\mathbf{k}$ in band j, and $A^{\pm}(\mathbf{k}, \mathbf{G})$ are the coefficients in the expansion of the periodic Bloch functions of $\psi\mathbf{k}_-(r)\psi_+(r)$.

Lock, Crisp, and West (LCW) proposed a folding procedure which makes evident the occupation number [13]:

$$\hat{\rho}^{2\gamma}_{\mathrm{IPM}}(\mathbf{p}) = \sum_{\mathbf{G}} \rho^{2\gamma}_{\mathrm{IPM}}(\mathbf{p} - \mathbf{G}). \tag{6}$$

If the positronic wavefunction is constant, the folded density becomes (in the IPM case)

$$\hat{\rho}^{2\gamma}_{\mathrm{IPM}}(\mathbf{p}) = const \sum_{\mathbf{k},j,\mathbf{G}} n_j(\mathbf{k})\delta(\mathbf{p} - \mathbf{k} - \mathbf{G}); \tag{7}$$

i.e., it is a periodic function in the reciprocal lattice space, and is determined by the occupation number of the Bloch electrons. This means in particular that occupied bands make approximately flat contributions.

Both in the alkali metals and in transition metals, expression (7) gives a first estimation in reasonable agreement with momentum distributions observed with positrons. The effects of the positron wavefunction, as well as correlation effects, in these cases are minor corrections. In the oxides the situation appears to be more complex.

• *Correlations*

The problem of correlation between the positron and the electron gas, and the influence of this correlation on positron measurements, have been the object of much work; for studies of the case of nearly free electrons, we refer to a recent paper by Stachoviak and references therein [14]. For the case of transition metals, Jarlborg and Singh have proposed a simple model [15] which successfully explains why correlations enhance the annihilation probability for

certain bands, while for other bands there is de-enhancement. The basic idea is to consider the change of the electronic wavefunction in the field of the positron, shielded within the correlation sphere. One finds decreasing effects in higher densities, giving enhancement which decreases with energy in the *d*-band.

The effects of electron–electron interaction are reviewed, for instance, by Berko [16].

Experimental methods

Positrons are used in three different experiments.

First, studying the *lifetime* of the positron in the solid gives a measure of the electronic density. In very clean and regular samples, the positron comes to rest in the crystalline solid, but impurities, domain walls, or phase boundaries will trap positrons, often in regions of lower density. As a result, several lifetimes are observed in these cases. Lifetime measurements are widely practiced as a means of materials testing [17].

Second, studying the *energy* of positron annihilation radiation measures the Doppler shift and gives information on the momentum density defined in relation (3). However, with this method one obtains the density in function of only one momentum component, which means a one-dimensional projection of the full three-dimensional information. The advantage of the method is its rapidity. Sometimes one extracts a shape parameter S, related to the width of the momentum distribution; S can be used to compare polycrystalline samples (above and below T_c, for instance).

Third, studying the *angular correlation of positron annihilation radiation* in one or two dimensions (1D-ACPAR and 2D-ACPAR) is the most powerful method: It results in one- and two-dimensional projections of the momentum density. From these projections, the full three-dimensional momentum distribution can be reconstructed with good accuracy, using algorithmic methods familiar from tomography [18]. In each event, the momentum of the annihilation pair $\mathbf{p}$ is carried away by the two gammas of momentum $\mathbf{p}_1$ and $\mathbf{p}_2$. The longitudinal component gives rise to the Doppler shift, and the transverse component gives rise to small angles between the nearly antiparallel quanta:

$$\mathbf{p}_x = mc\Theta_x, \qquad \mathbf{p}_y = mc\Theta_y. \tag{8}$$

For electrons with Fermi velocity, the angle is of the order of milliradians; therefore, precise measurements with two-dimensional detectors are required. Many groups use scintillation detectors (Anger cameras) for this purpose; in Geneva we use wire chambers with lead converters according to Manuel et al. [19]. These wire chambers have turned out to be very reliable and well adapted to the ^{22}Na sources currently used. Should more intense sources become available (enriched ^{22}Na, ^{64}Cu, beams), faster detectors might be required.

ACPAR experiments in metals

Equation (4) shows that the observed momentum density will have a quasi-periodic structure in the reciprocal lattice. In the case of constant ψ_+, each occupied Bloch function will have Fourier components at $\mathbf{p}_n = \mathbf{k} + n\mathbf{G}_0$, the total weight of these components being one by closure, which is the basis of LCW folding. We therefore expect to find an image of the Fermi surface in the first zone, repeated in the next zones with ever weaker amplitudes [for constant positron density, the amplitude is the Fourier transform of the Wannier function, as can be deduced from Equation (5)]. In the case of *alkali metals*, this structure was indeed observed, in very good agreement with the IPM model [20] (**Figure 2**).

Recent work in *transition metals* has been summarized by Manuel et al. [21]. Results exist for V (**Figure 3**), Nb, Mo, and Mo–Nb alloys, Cr both paramagnetic and antiferromagnetic, and ferromagnetic Fe and Ni, with observation of the momentum–spin density [8]. It is clear that agreement with band structure calculations is less good in the transition metals than in alkali metals. Enhancement factors must be used, and in some cases, adjustments of the energies of the different bands have been tried. This poses the question of the "legitimacy" of band calculations in this context. Such calculations should almost always give correct energies and densities, as shown by Kohn [23]. The potentials which are commonly used for band calculations are, however, obtained under certain simplifying assumptions, such as the local density functional approximation or even the atomic sphere approximation; removal of these restrictions should lead to the "correct" potential, and hence to good density and energy. Even then, however, a good momentum density and good excited states are not guaranteed, and special efforts must be made to estimate the electron–positron correlation effects. In view of this, it is a rather remarkable experimental fact that band calculations do achieve remarkably good predictions of the Fermi surfaces observed, and that these results can still be improved with relatively transparent correlation corrections [15]. On the other hand, it is not surprising that in the *rare earths* the difficulties with agreement increase [24].

- *Oxides*

The momentum distribution of $Na_{0.64}WO_3$, a tungsten bronze, was measured by Akahane et al. and analyzed following the method of Chiba [25, 26]. If the density of a *nonconducting oxide* is subjected to LCW folding, then, in the simplest hypothesis (IPM and constant positron wave function), a flat distribution should result. Such a distribution was seen, for instance, in MgO (A. A. Manuel, private communication). On the other hand, Wachs et al. have seen small deviations of closed-band behavior in NiO which they ascribe to nonuniformity of the positron wave functions [27].

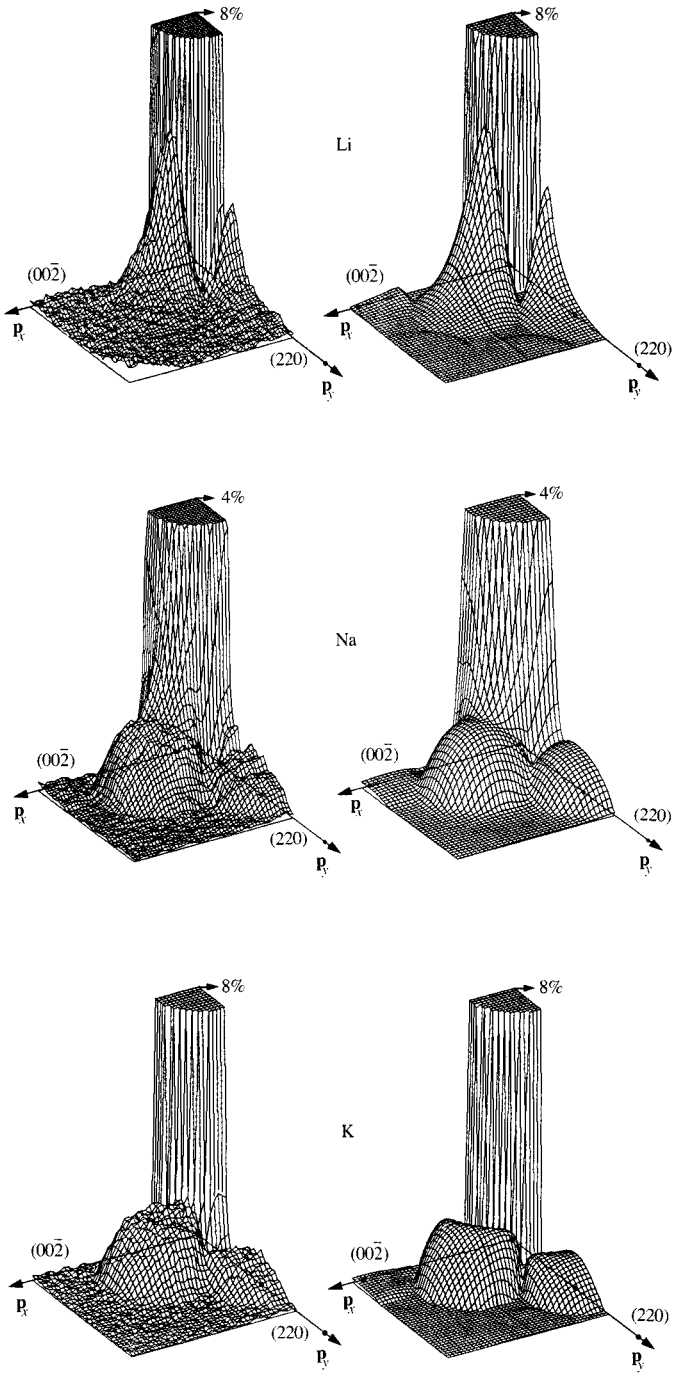

Figure 2

2D-ACPAR of Li, Na, K: left, experiment; right, band structure calculation.

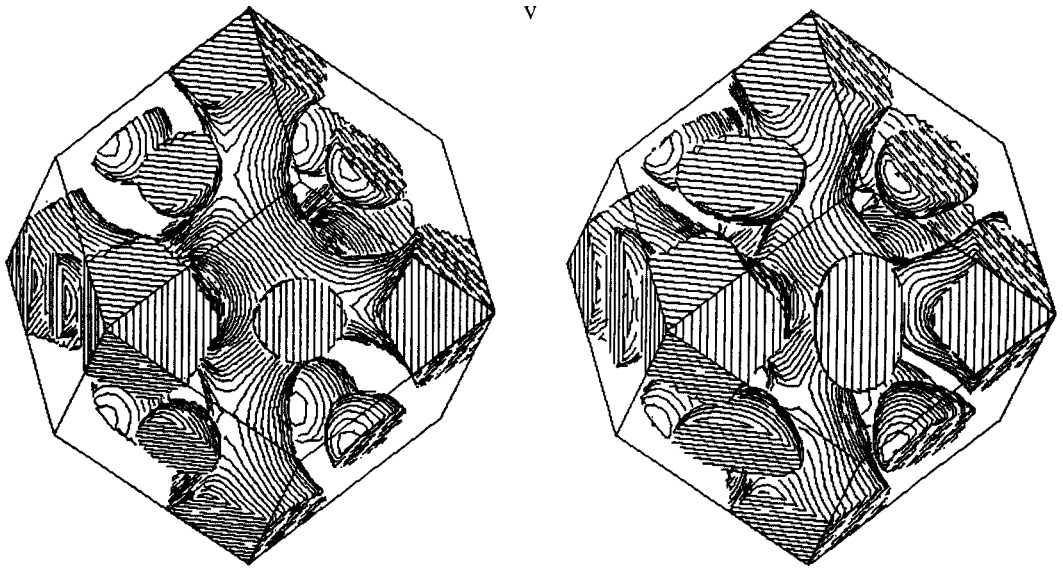

Figure 3

Fermi sphere of vanadium: left, reconstructed from 2D-ACPAR; right, from band calculation by Jarlborg et al. [22].

• *Metallic superconducting compounds*

ACPAR techniques work as well with compounds as with elements, and even with disordered alloys [28]. This is an advantage over the de Haas van Alphen (dHvA) technique, where the electronic lifetime must exceed the cyclotron period, requiring very pure samples and/or very strong magnets, as well as low temperatures.

The A15 phases, which held the record for highest superconducting transition temperature T_c before the advent of the oxides, had been studied carefully with ACPAR (**Figure 4**). References and a short summary of these studies may be found in [21]. In these complex compounds also, remarkable agreement between observed ACPAR and band structure calculations (in particular also linear muffin-tin orbital (LMTO) calculations [30]) was obtained. Certain features around the Γ point, corresponding to electrons on the linear chains, appeared in all high-temperature A15 compounds; the interaction of these electrons produced pronounced phonon anomalies; positron, neutron, Raman data, and band calculations suggest that these compounds are BCS superconductors with high density of states and high electron–phonon coupling, and not-too-strong exchange coupling [31]. Thus the positrons gave us a rather detailed and conventional picture of what were the high-T_c materials before the oxides. We were thereby somewhat misled in our appreciation of the possibility of increasing T_c, but at least we had a technique which enabled us to study the amazing new oxides.

Positrons in the superconducting oxides

In view of all the detailed information which positrons have delivered about metallic superconductors, there is an obvious interest in applying these same techniques to the new oxides. As this paper is being written, there is still no consensus as to the mechanism responsible for high T_c in oxides. Few believe that electron–phonon coupling can achieve pairing in the new temperature range. On the other hand, many expect to find a BCS-like state, stabilized by a new pairing mechanism (plasmons, superexchange, etc.) to be relevant in a band-like metal, leading to a BCS-like state, perhaps with short coherence length, and with a quasi-instantaneous interaction which shifts attention from the density of states at E_F to the total number of charge carriers. In contrast, there are those who feel that in the proximity of a Mott transition the band picture is inadequate, and who aim to establish a very different description of the RVB type. We have quoted some of our preferred authors on the subject in our contribution to ICPA-8 [32], and would like to mention the contributions to that conference of Varma, of Aharony, and of Emery, as well as the famous work of

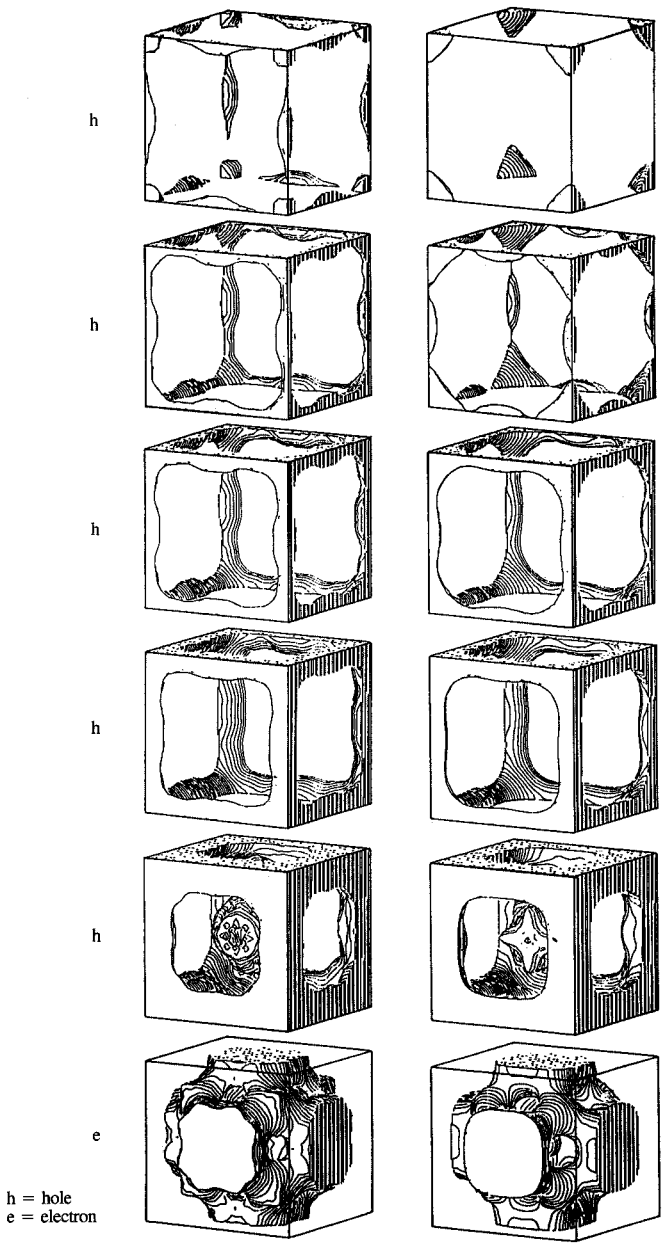

Figure 4

Fermi surfaces of Nb_3Sn extracted from reconstructed 3D occupation density [29]: left, experiment; right, theory.

Kitaoka et al. which focuses our attention on the oxygen holes but leaves the magnetic fluctuations on the copper site as a puzzle [33].

What can 2D-ACPAR contribute to the solution of these questions? In principle, a very accurate map of the momentum density should reveal the modifications of electronic density due to the superconducting transition which are expected to occur close to the Fermi surface, provided there is a Fermi surface, and provided there is sufficient resolution [34, 35]. In practice, momentum resolution of ACPAR machines is limited, and no effects directly attributable to superconductivity have yet been

firmly established. Lifetimes and shape parameters have been seen to vary around T_c (for references see [32]) but with wide variations between authors. For reliable data, and *a fortiori* for 2D-ACPAR, single crystals of sufficient quality and size are indispensable. For the work in Geneva, Damento et al. [36] contributed single crystals; later specimens were grown by W. Sadowski in collaboration with E. Walker and H. Scheel. For the different methods used for crystal growth, see [36–45].

• *ACPAR in* $(La_{(1-y)}Sr_y)_2CuO_4$

Concerning this class of substances, we cite the papers of Tanigawa et al. [46] and of Wachs et al. [47].

The latter work concerns samples with $y = 0$, hence semiconductors involving basically full bands. Therefore, no Fermi surface is seen. The electronic momentum density is modeled by a ligand-field Hamiltonian in fair agreement with measurement. The fit is based on a linear combination of a Cu *d* state with oxygen hole states. The description is therefore appealing to intuition. Its success is not surprising in view of the fact that, for full bands, a description based on Wannier functions is equivalent to one based on Bloch functions, and that the model includes several adjustable parameters.

The work of Tanigawa gives data for $y = 0$ to $y = 9\%$ and claims to see similar Fermi surfaces in both samples. Tanigawa also finds agreement with band calculations, provided the Fermi energy is adjusted downward. If this interpretation is taken literally, obvious contradictions with transport properties would have to be explained.

The uncertainty thus created touches on the most potentially useful contribution that positrons can make to the understanding of the oxides: Is there a Fermi surface? How big is the discontinuity at the Fermi energy? The results on La_2CuO_4 underline the fact that, particularly in the compound, the structure visible in the positron data, after LCW folding, is largely due to causes other than occupation number and Fermi surface; positron wavefunction, and eventually correlation effects, play an important role.

• *ACPAR in* $YBa_2Cu_3O_{7-x}$

Measurements in this compound have been reported by the Geneva group [32, 48], by the Argonne group [49], and by the Tsukuba group [50]. Among the theoretical calculations we mention three late references and, of course, the earlier work referenced therein [51–53].

It is fair to say that measurements and interpretation generally agree in this substance. The detailed analysis by Bansil and coworkers of the data of the Argonne group points the way. They confirm the calculated band structure and see the discontinuities at the Fermi energy. Their measurements were taken along the *c*-axis and therefore give results practically without reconstruction.

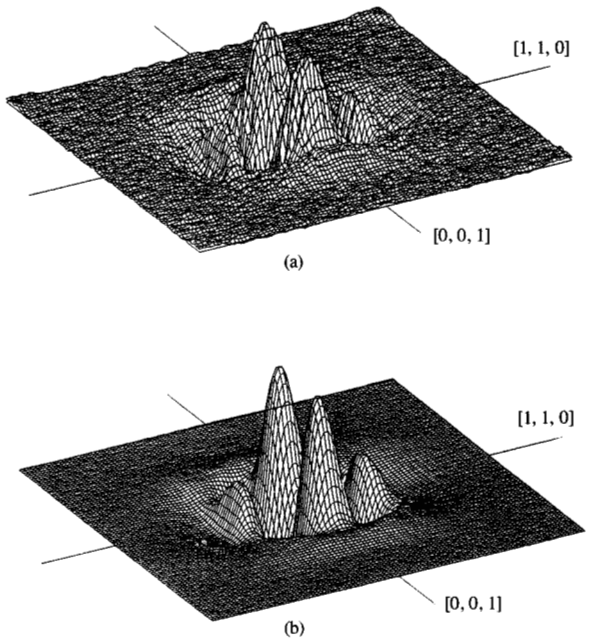

Figure 5

Momentum density in $YBa_2Cu_3O_{7-x}$, unpublished measurements by the Geneva group: (a) superconducting sample; (b) insulating sample.

The Geneva group has thus far measured perpendicular to the *c*-axis and has less detailed results. We tried to compare the momentum distribution between conducting and nonconducting samples. The result, suggested by the intermediate data shown in **Figures 5** and **6**, and apparently confirmed by later measurements, is that the *anisotropic* part of the momentum distribution in **p** space is twice as large in the insulator as in the conductor, and that after LCW folding the anisotropic part (in **k**-space) is twice as large in the conductor as in the insulator. This experimental result can be interpreted in terms of an important contribution due to a Fermi surface which is overcome in the insulator by other sources of anisotropy, but becomes dominant after folding.

Conclusion

Positron annihilation, and particularly 2D-ACPAR, has been successfully used to observe Fermi surfaces in many "conventional" metallic superconducting elements and compounds, and to find significant agreement with band structure calculations. In La_2CuO_4, after LCW folding, a structure is observed both in conducting and insulating

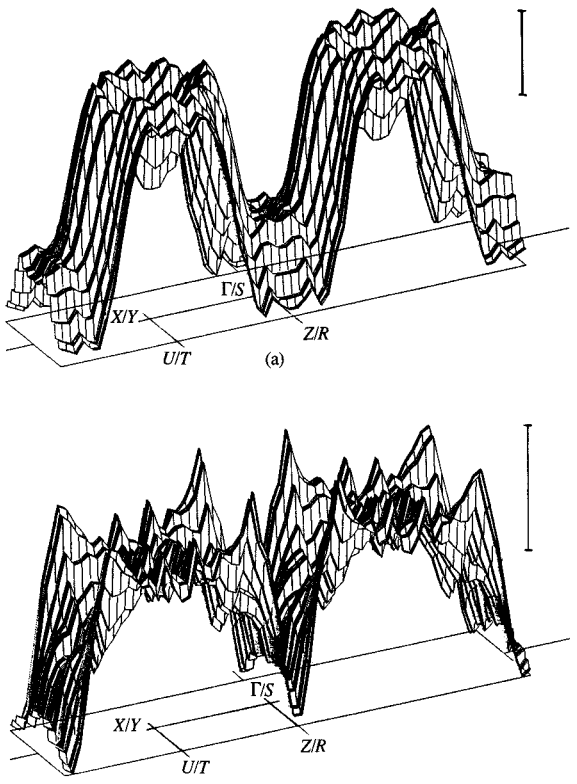

Figure 6

Same measurements as in Figure 5 after LCW folding (see text).

samples; the interpretation of these structures is still in doubt, but underscores that positron wavefunction and possible correlations make larger contributions than in the metallic case. $YBa_2Cu_3O_{7-x}$ has been more widely studied, and the best results strongly suggest the presence of a Fermi surface and a band structure in agreement with the calculation of Bansil et al. The need for further confirmation of these results, and extension to the other superconductors, is obvious.

Note

Recent measurements have been performed at Geneva [54] on the same YBCO single crystal, made either metallic ($\sim O_7$) or insulating ($\sim O_6$) by heat treatment. In *p*-space, one notes a narrowing of the 2D-ACPAR when moving from the metallic to the insulating phase, as noticed, for example, by von Stetten et al. [55] for ceramic samples. This narrowing may be understood [56] as the effect of a localization of the positron wavefunction in sites made empty on the CuO chains. (These effects have recently been reviewed by Manuel [57].)

After folding, these two measurements give similar LCW distributions. A similar result has also been obtained by Tanigawa et al. [46] in Sr–La–Cu–O. These two results lead us to think that positron effects are predominant and that Fermi surface features are small. This hypothesis is consistent with the recent work of Bansil et al. [53] and with band structure calculations of the 2D-ACPAR using LMTO by Jarlborg et al. [58].

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