# Positron annihilation in high-$T_c$ superconductors: present status

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# Positron annihilation in high-$T_c$ superconductors: present status

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**Abstract.** Positron annihilation has been extensively used to study high-$T_c$ superconductors during the last two years. Positron-lifetime, doppler-broadening and angular-correlation measurements have been reported and significant results have been obtained. We review this effort, focusing on the investigation of the electronic structure of these materials. We also try to establish the fundamental issues, with the present actual knowledge of high-$T_c$ superconductivity.

## 1. Introduction

The discovery of high-$T_c$ superconductivity by Bednorz and Müller (1986) has stimulated a large effort in all areas of solid state physics. Many kinds of experiments have been performed to study newly-discovered high-$T_c$ materials and various theories are proposed to explain the unusual properties of these superconducting oxides. One of the most comprehensive sources of information on this subject is the proceedings of the 'Interlaken Conference' (Muller and Olsen 1988). In the present paper, we review research on high-$T_c$ compounds using positron annihilation. A description of the method may be found in Brandt and Dupasquier (1983) and Hautojärvi (1979). The reader is referred to a previous paper (Manuel 1988) for a discussion of positron annihilation in 'classical' superconductors.

There are two fundamental questions concerning the new class of materials. The first is to know what mechanism governs their properties: BCS-phonons, polarons, excitons spinons and holons or other excitation? The second concerns the ultimate properties of these materials: $T_c$, $H_{c2}$, $I_c$, mechanical and corrosive properties are crucial for the possible applications. There is no final response to these questions yet, but they have to be kept in mind when discussing the potentialities of any method of investigation, and we shall later address these questions during our discussion of the results of positron annihilation investigations.

Our review of the literature is divided into three parts. In § 2, we discuss the behaviour of the positron in cuprates. Is the positron localised in some region of the lattice unit cell? What is the effect of oxygen vacancies on the annihilation parameters? In § 3 we discuss the possible ways of observing some manifestations of the superconducting transition and what we can learn about the mechanism itself. In § 4, we focus on the

**Table 1.** Positron lifetimes $\tau_i$ and their respective intensities $I_i$ in orthorhombic $YBa_2Cu_3O_{7-x}$ at room temperature.

| Author and reference | $\tau_1$ (ps) | $I_1$ (%) | $\tau_2$ (ps) | $I_2$ (%) |
|---|---|---|---|---|
| Teng *et al* 1987[a] | 185 | 93 | 533 | 7 |
| Jean *et al* 1987 | 139 | 63 | 220 | 30 |
| Usmar *et al* 1987[b] | — | — | 210 | 94 |
| Ishibashi *et al* 1988[c] | 95 | 45 | 210 | 55 |
| Bharathi *et al* 1988b | 50 | 5 | 190 | 95 |
| Mandal *et al* 1988 | 153 | 67 | 333 | 28 |
| Balogh *et al* 1988 | 146 | NA | 220 | NA |
| Wang *et al* 1988 | 201 | NA | 850 | NA |
| Harshman *et al* 1988 | 165 | 100 | — | — |
| Moser and Henry 1988[d] | 178 | 100 | — | — |
| Hong Zhang *et al* 1988a | 174 | 89 | 338 | 10 |

[a] At 106 K.
[b] At 160 K.
[c] At 200 K.
[d] Single crystal.

question of the electronic structure of high-$T_c$ superconducting compounds: are they strongly correlated systems or band-like metals?

## 2. Behaviour of positrons in high-$T_c$ superconducting materials

The behaviour of positrons in high-$T_c$ materials has been studied both theoretically and experimentally. Since the annihilation rate is determined by the overlap of electron and positron wavefunctions, a precise determination of these wavefunctions is required. Calculations have all been performed on the basis of the approximation of independent particles. In $La_2CuO_4$, Jean *et al* (1988) have shown that the positron is wrapped around the Cu(1)O(1) spine at the interstices of the bonds. In $YBa_2Cu_3O_7$, more calculations are available (Bharathi *et al* 1988a and 1989, Jarlborg *et al* 1988, Szoteck *et al* 1988, Turchi *et al* 1988 and von Stetten *et al* 1988). They show that the positron is rather delocalised and forms 'chains' along the ordered rows of vacant sites in the plane containing the Cu–O chains. Von Stetten *et al* (1988) and Jarlborg *et al* (1988) obtain a positron density distribution which is very small in the *Y* plane, despite the large empty space in the lattice. This is due to the charge transfer which confers a positive charge to this region, repulsive to the positron. The 2D delocalised state of the positron has also been proposed by Szotek *et al* (1988) from a positron band structure calculation. Therefore, if one assumes that the band picture is valid for describing electronic and positronic states in $YBa_2Cu_3O_7$, one may conclude that the positron density distribution, while not uniformly distributed in the lattice cell, is delocalised.

Experimentally, the behaviour of the positron in a solid may be investigated by different methods: positron lifetime, Doppler broadening and angular correlation of the annihilation radiation. We shall now focus on measurements of the positron lifetime because they are very sensitive and widely used.

We summarise in table 1 typical lifetime values reported for the 1–2–3 Y–Ba–Cu compound. In this table we have shown results obtained at room temperature,

temperature effects being discussed in § 3. The results reported in table 1 are from sintered samples having a small oxygen deficiency, i.e. samples undergoing a superconducting transition. Lifetime spectra are analysed assuming one, two or three different lifetimes $\tau_i$ of relative intensities $I_i$. The number of lifetime components is a delicate quantity to determine. It is fixed on the basis of a chi-squared test between the lifetime spectrum and a model spectrum. In metals, $\tau_1$ is the lifetime of positron annihilating with the bulk material, $\tau_2$ and $\tau_3$ are components due to annihilations of positrons trapped in defects (vacancies, dislocations, etc). Positron lifetimes depend of the electron concentration at the site of the positron, hence $\tau_2$ and $\tau_3$ are longer than $\tau_1$. Usual lifetimes spectrometers have a time resolution of 250–300 ps, which is of the order of magnitude of the positron lifetime. It is therefore rather difficult to separate lifetime components which differ by less than 20 ps.

At first inspection, table 1 seems to suggest a large dispersion of the results obtained in $YBa_2Cu_3O_7$. This means neither that the method is inadequate nor that it is delicate in its interpretation, as it is a very well established experimental tool which gives well characterised and reproducible results (MacKenzie 1983). The dispersion of the results reflects the complexity of the study of superconducting oxides. The preparation of samples is easy but their characterisation is extremely difficult. The granularity of the powder used to produce sintered samples, temperature treatments, homogeneity and the presence of an eventual second phase are factors which certainly play an important role in the dispersion of the results. Even if samples have a high $T_c$, exhibit a sharp superconducting transition and have a metallic-type resistivity, it does not preclude large effects due to the above-mentioned factors. The main message of the confrontation of these experimental values is to warn us not to draw hasty conclusions from individual studies. Considerable effort has to be expended to obtain reliable and well characterised results. A possible explanation for the dispersion of the positron lifetimes has been proposed by Usmar *et al* (1988). These authors have observed an important variation of lifetime with the microstructure of their samples and conclude that the positron behaviour in $YBa_2Cu_3O_7$ may depend critically on the twin density of the material studied.

We now summarise the results and conclusions obtained so far about the positron behaviour. Jean *et al* (1987) resolved three positron lifetimes in all their spectra. The weak long-lived component $\tau_3$ was easily identified as ortho-positronium annihilation at the interfacial spaces or surfaces among crystals. Their shorter lifetime $\tau_1$ (see table 1) has a typical value for positrons annihilating in bulk metals. It does not show significant temperature dependence. They have attributed the intermediate lifetime component $\tau_2$ to annihilations of positrons lying in unoccupied lattice sites (often called oxygen vacancy sites, with the risk of confusion with vacancies present on the Cu–O chains of oxygen deficient samples). Later, Usmar *et al* (1987) observed aging effects on the annihilation parameters: room temperature lifetime measurements performed at two-day interval gave different results, stressing the extreme sensitivity of positron annihilation to surface deterioration. Aging effects have also been reported by Wang *et al* (1988), who outline the important role played by defects in the superconducting state.

The question of the charge state of oxygen vacancies has been addressed by some authors. Bharathi *et al* (1988b) have studied the variation of annihilation parameters with oxygen concentration, quenching samples from various temperatures to vary the concentration of oxygen. Assuming that all Cu ions exist in a '2+' state, charge neutrality implies positively charged O vacancies in the orthorhombic phase and neutral ones at $O_{6.5}$. Positrons are therefore less effectively trapped in the orthorhombic phase and their wavefunctions overlap more efficiently with electrons of neighbouring atoms. According to Bharathi *et al* (1988b), this model explains why $\tau_2$ in the orthorhombic phase is smaller than in the tetragonal phase. Balogh *et al* (1988) have a different opinion: assuming that Cu(1) sites are occupied by $Cu^{3+}$ ions, the authors think that the weak trapping they observe is due to the presence of neutral trapping centres, probably oxygen vacancies.

The trapping of positrons in $YBa_2Cu_3O_{7-x}$ has also been discussed by Smedskjaer *et al* (1988c) who conclude that, for small values of $x$, almost none of the positrons are trapped, while an increasing fraction of the positrons become trapped for increasing $x$. Their data suggest that oxygen vacancies act as trapping centres, excluding positively charged anion vacancies. For low vacancy concentrations, they evaluate the specific positron trapping cross section to be $1.5 \times 10^{11}\ s^{-1}$ in Cu(1)–O(1) layers which have chains of unoccupied crystallographic sites. This result is four orders of magnitude less than for a vacancy in a typical metal. It has been confirmed by Balogh *et al* (1988). Hong Zhang *et al* (1988a) and Xiany Zhou *et al* (1988) came to the same conclusion. These are important results for the study of electronic structures by positron annihilation: they suggest that the method is able to probe electrons from the bulk material, opening the possibility of investigating the electronic structure, at least in the case of small $x$.

Positron-annihilation Doppler-broadening spectroscopy has been used by Hong Zhang *et al* (1988b) to monitor the movement of oxygen vacancies. They found an activation energy of $0.8 \pm 0.1$ eV for the migration of oxygen vacancies in the orthorhombic phase of the Y–Ba–Cu–O compound.

The number of results for other compounds is less than for Y–Ba–Cu–O. Let us mention the study of Semedskjaer *et al* (1987), who have investigated the behaviour of oxygen vacancies in $La_{2-x}Sr_xCuO_{4-y}$. Shape parameters in $HoBa_2Cu_3O_7$ have been measured by Mandal *et al* (1988) who conclude that the substitution of Ho for Y does not alter appreciably the annihilation characteristics. The effect of doping of $YBa_2Cu_3O_7$ with Sr has been investigated by Huang *et al* (1988). These authors conclude that the number of oxygen vacancies increases with the strontium concentration.

## 3. Does one observe manifestations of the superconducting transition with positron annihilation methods?

In this section, we discuss the temperature dependence of positron annihilation characteristics, mainly the shape parameter of the Doppler-broadened annihilation line. This last quantity, as well as the distribution of angular correlation of the positron annihilation radiation, gives the momentum of the positron–electron pair. The contribution of the positron to the total momentum of the pair is usually negligible because the positron is thermalised prior to its annihilation.

A theory of positron annihilation in superconductors has recently been presented by Barnes and Peter (1989). The reader is referred to Manuel (1988) for a discussion of the expected manifestations of the superconductivity on positron annihilation characteristics. Here we restrict ourselves to mentioning only the two most important, related to the presence of a gap at the Fermi level: (i) a possible lack of thermalisation of the positron at the annihilation time and (ii) a smearing of the break at the Fermi momentum in the electron momentum density distribution (see also Brovetto *et al* 1987).

The shape parameter $S$ of the annihilation curve is defined as the ratio of the area of the central part to the total area under the curve. There is no established criterion to define the central area of the curve, therefore a comparison of absolute values of $S$ obtained by different authors is not possible. Nevertheless, the variation of the shape parameter with temperature is not affected by a specific criterion, and variations with temperature can be compared.

We show in figure 1 various temperature variations of the shape parameter $S$ for superconducting samples of $YBa_2Cu_3O_{7-x}$. It is very interesting to note that all measurements show some feature at $T_c$, despite the widely differing temperature variations obtained.

The first report of a peculiar behaviour of the shape parameter around $T_c$ is due to Ishibashi *et al* (1987) (see also Doyama *et al* 1987). Jean *et al* (1987) were the first to observe that $S$ does not show any anomaly in non-superconducting samples, while a significant variation is observed at $T_c$ (an increase at the onset in their case) in superconducting samples. These authors have concluded that a change in the electronic structure occurs at $T_c$, the electronic density near oxygen vacancies being higher in the superconducting state. Usmar *et al* (1987) also observed the same difference between superconducting and insulating samples in the variation of the shape parameter $S$ with temperature. They have suggested that the increase of $S$ at $T_c$ may be linked to a ‘condensation’ of electrons into unoccupied crytallographic sites (O(5) and O(6)). The idea that the electronic density at the site of the positron is higher in the superconducting state is also shared by Ishibashi *et al* (1988), on the basis of lifetime results correlated against the superconducting transition, and by Sundar *et al* (1988a, b), who suggest that this effect may be due to a dimerisation of oxygen ions in the superconducting state of $YBa_2Cu_3O_7$. The fact that the shape parameter of non-superconducting samples does not exhibit anomalies has also been reported by von Stetten *et al* (1988) for $YBa_2Cu_3O_{7-x}$ and by Jean *et al* (1988) for $La_{1.85}Sr_{0.15}CuO_4$. Moreover these authors report evidence for a common high-temperature superconducting effect in the 1–2–3 and 2–1–4 systems by comparing their data with an older set (Harshman *et al* 1988). They obtain the same variations of the positron lifetime with reduced temperature. In both cases they observe an increase of the lifetime in the superconducting state when the temperature decreases. This indicates, in contrast to results mentioned previously, a reduction of the sample electron density in the superconducting state. Jean *et al* (1988) conclude that this trend is consistent with a localised ion scheme. For completeness, let us also mention that Chanalambous *et al* (1988), Zhonjin *et al* (1987), Zhu Jingsheng *et al* (1988) and Teng *et al* (1987) have respectively reported anomalies in lifetime spectra and Doppler lines which until now have not always been reproduced and which are difficult to interpret.

A discussion of the variation of the shape parameter in terms of a particular superconducting mechanism is made difficult by the large variety of results. Smedskjaer *et al* (1988b) have claimed that the large positive temperature dependence of the shape parameter they observed may be consistent with a BCS-like theory, if an energy band characterised by a small dispersion crosses the Fermi level. From our point of view, it is very hazardous to conclude this on the basis of the results available today. If it is widely accepted that the positron annihilation characteristics are sensitive to the superconducting transition in oxides, it is very difficult safely to correlate observations with a particular mechanism or even with the presence of a gap at the Fermi energy. Moreover it has to be proved that anomalies at $T_c$ are not due to a cause only indirectly related to the superconducting state. The following (not exhaustive) list of causes has been given by von Stetten *et al* (1988): (i) $e^+$–$e^-$ correlations changes due to the superconducting pairs; (ii) changes in the positron trapping probability due to (i) or due to changes in vacancy screening below $T_c$; (iii) structural change below $T_c$; (iv) large anharmonic motion of the O(4) oxygens; (v) changes in the electronic density near the Cu–O plane.

We finally mention some peculiar variations of the shape parameter observed at temperatures above $T_c$ in $YBa_2Cu_3O_{7-x}$. The most interesting one is the very sharp

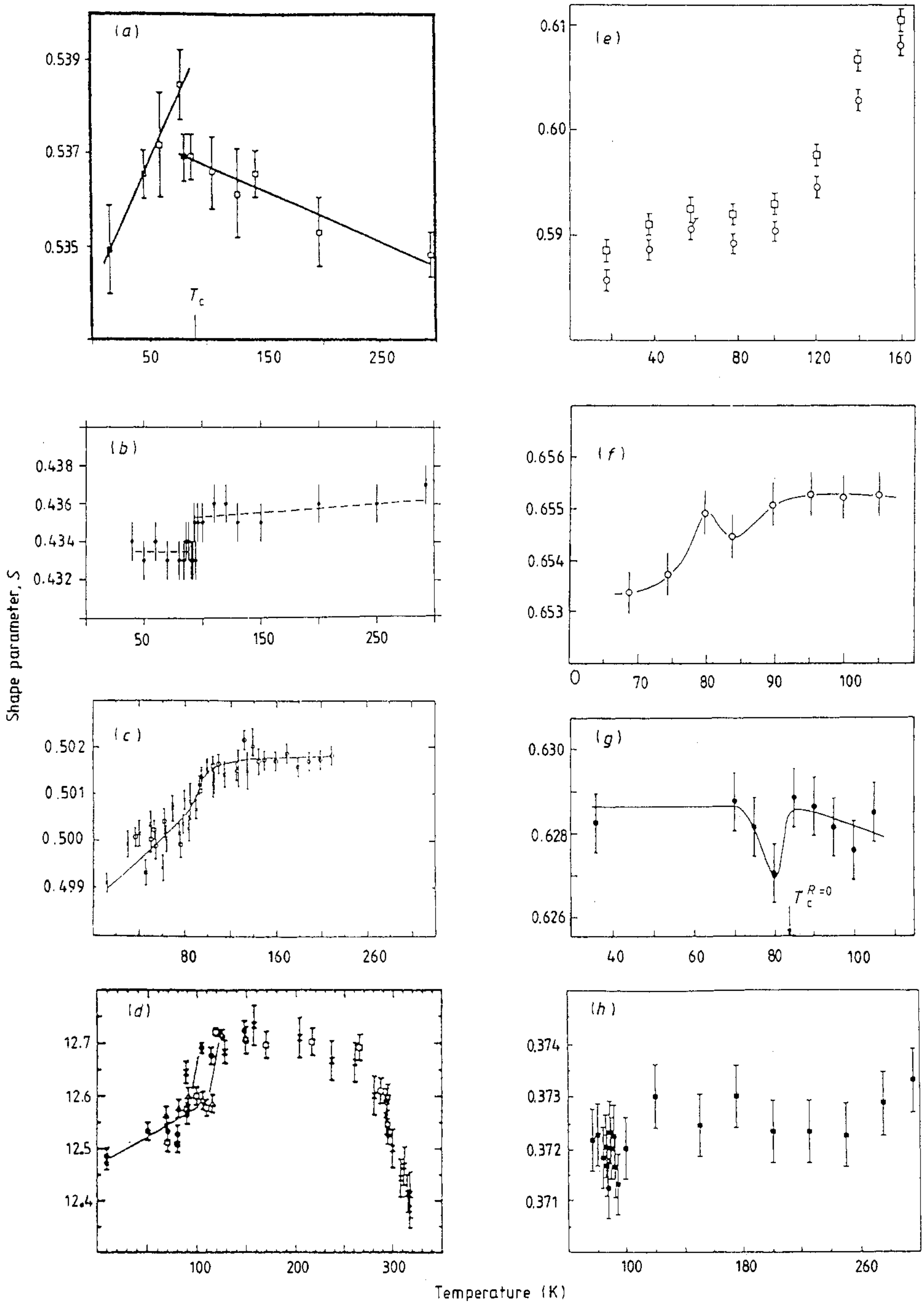


**Figure 1.** Shape parameters, $S$, of the Doppler-broadened positron annihilation curve in superconducting $YBa_2Cu_3O_{7-x}$. (*a*) Smedskjaer *et al* (1988c), (*b*) Jean *et al* (1987), (*c*) Usmar *et al* (1987), (*d*) von Stetten *et al* (1988), (*e*) Hoffmann *et al* (1988a), (*f*) Ishibashi *et al* (1987), (*g*) Mandal *et al* (1988), (*h*) Wang *et al* (1988).

discontinuity at 240 K observed by Balogh *et al* (1988). Brusa *et al* (1988) have also observed a fall of the shape parameter above 240 K. What happens at this temperature is not well understood but anomalies in many physical properties have been reported. Balogh *et al* (1988) propose either the presence in their sample of a second phase undergoing a phase transition at this temperature or an interaction between the samples and water or water vapour. Brusa *et al* (1988) suggest a shallow positron trapping in oxygen vacancies, inhibited at low temperature by variations of the local charge. A decrease of the shape parameter above 100 K has been reported by von Stetten *et al* (1988) in some samples. It is also interpreted as thermal detrapping of positrons from shallow traps. Lynn *et al* (1988) have also obtained a similar decrease above 230 K and mention that an additional structure seems to occur around 240 K in a number of different samples, its origin being unclear at present.

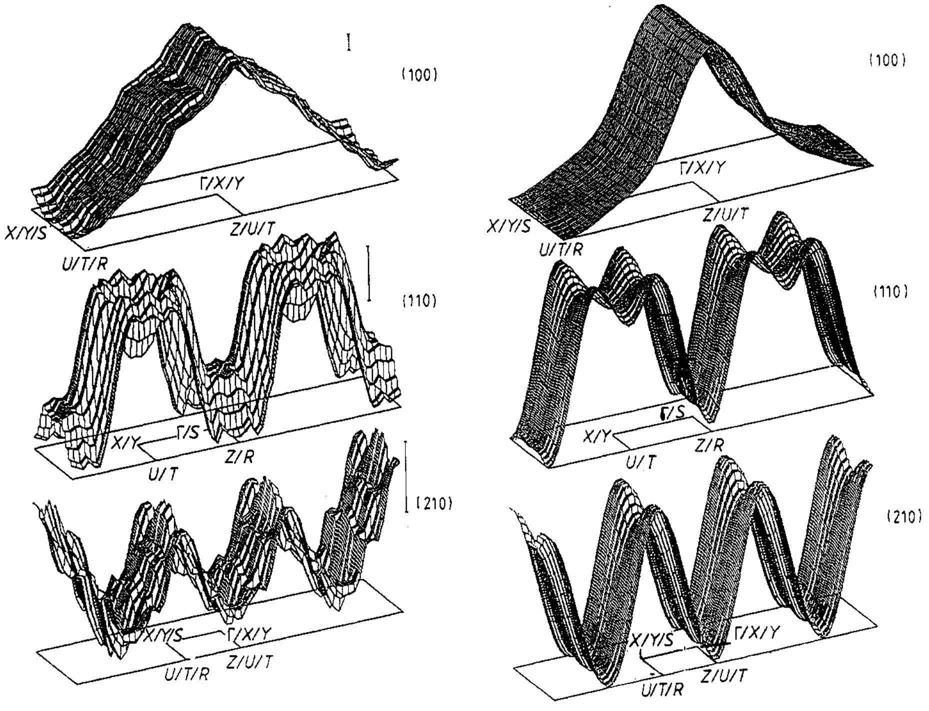
## 4. Studies of the electronic structure of superconducting oxides

The question of the electronic structure of the superconducting oxides is fundamental. Actually two possibilities are considered: (i) the ‘classical’ picture of a metal described by a band structure and characterised, among other things, by a Fermi surface; (ii) that strong correlations are dominant and an ionic picture would be adequate to describe superconducting oxides.

Positron annihilation, and more precisely measurements of 2D angular correlations, is a perfectly satisfactory method for investigating electronic structures. Both the 1–2–3 and 2–1–4 systems have been studied. Since recent reviews are available on this subject (Peter 1988, Peter *et al* 1988a, b), we only survey this field briefly.

The first point to make is that studies of electronic structures have to be performed on single crystals. Despite the small number of studies of the behaviour of positrons in single crystals of superconducting oxides, one may state (or hope) that the situation in these cases is less confused than in ceramics. A second point in favour of these studies appears in the light of the results reviewed in § 2: in samples with low oxygen deficiency, it is highly probable that positrons are delocalised and will therefore probe the electronic properties of the bulk materials. A more delicate point is that the majority of data analyses are based on the Lock–Crisp–West (LCW) theorem (Lock *et al* 1973), which is exact only if (*a*) the positron wavefunction is a constant and (*b*) electron–positron correlations are negligible. In metals and intermetallic compounds a variable positron wavefunction can be introduced into the calculations and it is well accepted that correlation effects do not play an important role. In superconducting oxides, one has less experience and some degree of caution is required in the use of the LCW theorem. Friedel and Peter (1989) have recently discussed this point.

The positron group at Livermore (Wachs *et al* 1988a, Turchi *et al* 1988) has studied the electronic structure of semiconducting $La_2CuO_4$ without resort to LCW and explains fairly well the experimental anisotropies of the measured 2D distributions of angular correlations with an LCAO-molecular orbital scheme. This result suggests strong electron localisation and indicates the importance of the covalency in this compound. Tanigawa *et al* (1987, 1988) have interpreted their measurements in the same compound using LCW. They came to the conclusion that, despite a negative slope in the resistivity of their sample, there is a Fermi surface consisting of two sheets, in agreement with band-structure results if one shift the Fermi level by a few tens of mRyd. Wachs *et al* (1988b)



(*a*) (*b*)

**Figure 2.** Distribution of the 2D angular correlation of the positron annihilation radiation from $YBa_2Cu_3O_{7-x}$ for three different crystal orientations. Each distribution represents a projection of electron occupation density. (*a*) Measurements, (*b*) models derived from band-structure calculations. (From Hoffmann *et al* 1988b.)

have stressed out the failure of LCW in NiO and suggested that a similar failure may occur in $La_2CuO_4$.

In $YBa_2Cu_3O_{7-x}$, the Geneva group (Hoffmann *et al* 1988a, b) was the first to report measurements of 2D angular distributions. The authors have investigated three different crystal orientations. As shown in figure 2, they obtained a good agreement with the Fermi surface calculated by Yu *et al* (1987) and, in early measurements (Peter *et al* 1988b), some evidence that the signal attributed to the Fermi surface is strongly attenuated in a non-superconducting sample. These two facts suggested that there is a Fermi surface in $YBa_2Cu_3O_{7-x}$. Smedskjaer *et al* (1988a) also came to this conclusion on the basis of the first measurement in the *ab* plane. The comparison with a calculation of the electron–positron momentum distribution (Bansil *et al* 1988) gives a good agreement, adding hence some credence to the existence of a Fermi surface in $YBa_2Cu_3O_{7-x}$.

Recent measurements have been performed at Geneva (Hoffmann 1989) on the same 1–2–3 single crystal, made either metallic ($\sim O_7$) or insulating ($\sim O_6$) by heat treatment. The narrowing of the 2D angular correlation curve noted by von Stetten *et al* (1988) for ceramic samples has been reproduced, but the two measurements give similar LCW distributions, suggesting that positron effects are predominant and that Fermi surface features are small. This hypothesis is consistent with calculations of the 2D-ACPAR distribution by Jarlborg *et al* (1989).

## 5. Conclusions

We have attempted to show the results obtained by positron annihilation on high-$T_c$ superconductors. The behaviour of the positron has been discussed and it has been concluded that this particle is delocalised in materials free of oxygen vacancies, while significant trapping may occur in oxygen deficient samples. The temperature dependence of the annihilation parameters systematically shows an anomaly at $T_c$, but it is difficult to ascribe it to a direct effect of superconducting electrons and, *a fortiori*, to a particular pairing mechanism.

The electronic structure of $La_2CuO_4$ and $YBa_2Cu_3O_{7-x}$ is at present a matter for debate. Independent measurements on $YBa_2Cu_3O_{7-x}$ led to the conclusion that a band-structure description of this material is in agreement with positron annihilation data. But comparison of results from insulating and metallic samples seems to indicate that positron effects are important and complicate the analysis of the data. Therefore, the existence of a Fermi surface is not yet certain.

Progress in high-$T_c$ superconductivity is very rapid. It is difficult to give an up-to-date overview. We have tried to cover the literature, but our work is certainly not complete. Further information will be found in the proceedings of the Eighth International Conference on Positron Annihilation (Dorikens *et al* 1988), held just after the present workshop†.

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