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# Positron annihilation and superconductivity

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*In honor of Martin Peter's 60th birthday.*

*Abstract.* We discuss how positron annihilation may or not be used to study superconductivity. A special attention is given to results obtained on recently discovered high- $T_c$  superconductors.

This short discussion of connections between superconductivity and positron annihilation [1, 2, 3] is triggered by two evident reasons. The first is the recent discovery of high- $T_c$  superconductors [4]. This event has considerably modified the situation in solid state physics: since one year, almost all available techniques have been used to investigate the very scheming properties of these materials. The second reason is that experimental solid state physics has greatly benefited from the large progress in electronics and computing sciences. Consequently, more precise investigations can be made. Positron annihilation does not make exception to this trend. New powerful equipments have open access to many areas of research. Naturally, they are applied to the new superconductors and start to produce interesting results which have to be discussed in the light of what has been done in the past.

The two dimensional angular correlation of the positron annihilation radiation (2D-ACPAR) is a typical example of this phenomena. The first 2D-ACPAR machine was developed at Brandeis University by S. Berko and his collaborators. It is made of arrays of scintillators. In its first stage, this apparatus was only producing some slices of the 2D distribution [5], but it was soon upgraded to deliver complete 2D distributions [6]. During this period, under the impetus of M. Peter and A. P. Jeavons, a very fruitful collaboration was established between CERN, and the University of Geneva, to develop high density proportional chambers [7]. A prototype angular correlation system gave its first results in 1978 [8] and was superseded later by a more efficient apparatus [9]. Besides multicomputers and wire chambers, use of Anger cameras was early made, with great profit, by R. N. West at the University of East Anglia [10, 11]. today, about ten 2D-ACPAR machines are in operation around the world [12]. All of them follow from one of the three paths mentioned above.

We shall now focus on superconductivity itself. We start first by discussing how positron annihilation and superconductivity met in the past. In the fifties, it was thought that, after a rapid slowing down, positron form positronium and annihilate from the positronium ground state [13]. Because the superconductive state is characterized by inability of the system of electron pairs to exchange energy with the lattice system or with impurities, the conversion of the long-lived triplet state of positronium will not be able to be efficiently converted to the singlet state and consequently an increase of the positron lifetime was expected [14]. It is now well established that positronium formation do not occurs in metals because the correlation between a positron and a single electron cannot last for times longer than the inverse of the plasma frequency ( $10^{-16}$  s) [15]. Large changes of the mean lifetime observed in superconducting lead [16] are now attributed to annihilations with liquid helium and no change of the lifetime has been observed neither in superconducting lead [17] nor in any other metals until the beginning of 1987. Concerning angular correlation measurements, no difference between superconducting and normal states were detected in lead [18]. This negative result is well understood in terms of the BCS theory when one consider what is the consequence of the superconductive condensation on the electron momentum distribution. Following de Gennes [19], one writes the BCS state as

$$\tilde{\phi} = \prod_{\vec{k}} (u_{\vec{k}} + v_{\vec{k}} a_{\vec{k}\uparrow}^{\dagger} a_{\vec{k}\downarrow}^{\dagger} a_{-\vec{k}\downarrow}^{\dagger}) \phi_0 \quad (1)$$

where  $\phi_0$  is the vacuum state. Minimizing (1) leads to the energy gap at the Fermi energy and simple properties of the superconducting state. The momentum distribution is deduced from the probability of finding an electron condensed into the state ( $k\alpha$ ):

$$\langle \tilde{\phi} | a_{\vec{k}\alpha}^{\dagger} a_{\vec{k}\alpha} | \phi \rangle = v_{\vec{k}}^2 = \frac{1}{2} \left( 1 - \frac{\xi_{\vec{k}}}{\sqrt{\xi_{\vec{k}}^2 + \Delta_{\vec{k}}^2}} \right) \quad (2)$$

where  $\xi_{\vec{k}}$  is defined by

$$\xi_{\vec{k}} = \frac{\hbar^2 k^2}{2m} - E_F \quad (3)$$

As noticed by De Genes,  $v_{\vec{k}}^2$  can in principle be measured by Compton profile or positron annihilation. In the case of the non interacting electron gas,  $v_{\vec{k}}$  has the trivial form

$$v_{\vec{k}} = \begin{cases} 1 & \xi_{\vec{k}} < 0 \\ 0 & \xi_{\vec{k}} > 0 \end{cases} \quad (4)$$

In the superconducting state, the distribution function  $v_{\vec{k}}^2$  is continuous. The transition region, where  $v_{\vec{k}}$  goes from 1 to 0, has an energy width and a momentum width  $k$ :

$$\frac{\hbar^2}{2m} [(\vec{k}_F + \delta\vec{k})^2 - k_F^2] \simeq \Delta \quad (5)$$

$$\delta_k = \frac{m\Delta}{\hbar^2 k_F} = \frac{\Delta}{\hbar v_F} = \frac{2}{\pi \xi_0} \quad (6)$$

where  $\xi_0$  is the coherence length of the superconductor. With  $k_F \sim 1 \text{ \AA}^{-1}$  and a  $\xi_0 \sim 10^4 \text{ \AA}$ ,  $\delta k/K_F \sim 10^{-4}$  in lead, well below the resolution of any ACPAR measurement. On the other hand, Nb<sub>3</sub>Sn, with  $\xi_0 \sim 50 \text{ \AA}$  has been early elected as a good candidate to detect the smearing of the Fermi break when going to the superconducting state. Faraci and Sapdoni have investigated this A15 compound with 1D-ACPAR measurements [20] and claimed they were able to observe the first experimental redistribution of  $k$ -space states at the superconducting transition. A careful look at their data reveals that this statement is based on the point at 7 mrad of the curve measured at 77 K, this point introducing a discontinuity in their curve. Nothing similar has been reported from recent 2D-ACPAR measurements by Hoffmann et al. [21].

Beside a smearing of the Fermi break, an other expected manifestation of the superconductivity has been pointed out by Perkins and Woll [22]. By calculating, as a function of time, the thermalisation of a positron which loses energy by exciting quasiparticles out of the BCS ground state, they came to the conclusion that the minimum positron energy shall change sharply when the metal goes to the superconducting state. This is easily understood since, when the energy of the positron drops in the range of the gap, its energy loss rate is cut off, owing to the presence of the energy gap, while at energies higher than the gap, the energy loss rate is larger than in the normal state, owing to the increased density of states just above the energy gap. Therefore they conclude from this mechanism to a lake of thermalisation of positrons, if the lifetime of the positron is three times larger than a critical  $t_0$  given by:

$$t_0 = \frac{m\hbar E_F}{2\pi m^* \Delta^2} \quad (7)$$

where  $m^*$  is the effective mass of the positron and  $E_F$  the Fermi energy. Conversely, the positron minimum energy should be smaller in the superconducting state than in the normal state if its lifetime is smaller than  $t_0$ . Perkins and Woll have estimated to few Kelvin the change of the positron temperature at the time of its annihilation in classical superconductors. Effects on high-T<sub>c</sub> superconductors will be discussed elsewhere but, to our knowledge, these have not been observed until now. To terminate this section let us mention the work of Tripathy and Bhuyan [23]. These authors have calculated lifetime of positron in superconducting aluminium, evaluating the annihilation rate in the high density limit. They found the very long lifetime ( $10^{-5}$  s) and their result converge to the value observed in the normal state when  $\Delta = 0$ . It is not physically clear why the lifetime should be so long in the superconducting state. Nevertheless, these authors made the suggestion that long lifetimes would be searched more efficiently with a dual parameter spectrometer, i.e. a setup in which lifetime of annihilation pairs having a specific momentum (the Fermi momentum in the

present case) may be selected (A review of lifetime techniques is given in Refs. [24] and [1]).

Before to focus on high- $T_c$  superconductors, let us mention what results were obtained by positron annihilation in Chevrel phases, in the superconducting oxide  $\text{BaPb}_{(1-x)}\text{Bi}_x\text{O}_3$  [25] and in A15 compounds. Properties of Chevrel phases have been reviewed by Fischer [26]. In these phases, lifetimes measurements have shown that positron are partially trapped by the channels which are intrinsic to the crystal structure [27]. It has also been observed that the intensity of this lifetime component decreases with increasing  $x$  in  $\text{Cu}_x\text{Mo}_6\text{Se}_{7.6}$ . This is coherent with the fact that Cu atoms obstruct the channels when  $x$  increases, diminishing the trapping accordingly. Charge transfer from Cu to  $\text{Mo}_6$  clusters has been established on the basis of 1D-ACPAR measurements, interpreted with a simple free electron model [28]. The same model has been used to establish correlations between  $T_c$  and the Fermi momentum [29]. In the superconducting oxide  $\text{BaPb}_{(1-x)}\text{Bi}_x\text{O}_3$ , positron lifetime measurements have been reported [30] for various  $x$  concentrations. Annihilation characteristics display anomalies correlating the dependence of  $T_c$  with the concentration of Bi. These results are interpreted as a manifestation of the nucleation and growth of the superconductive phase. In this model, interface boundaries, which are supposed to comprise electrons defects, are potential sites of positron trapping.

Superconducting properties of A15 materials have been reviewed by J. Muller [31]. One of the main question was to understand the variation of  $T_c$  in these materials. It was clear very early that independent families of linear chains, the principal characteristic of the  $\beta$ -W structure, may be at the origin of superconductivity. Labbe and Friedel [32] have outlined the role of these chains, which may give a high density of state at the Fermi energy. The key point was to know if the Fermi level of a particular A15 compound lies at a peak of the density of states and if the interchain coupling was strong enough to smear these peaks. M. Weger was one of the pioneers in the field [33] and recognized very early that positron annihilation would be an efficient way to determine Fermi surfaces (did they present planar sections?) and electron momentum distributions. A clear answer was obtained by a joint effort of precise band structure calculations and precise 2D-ACPAR measurements. The first A15 compound to be extensively investigated on this way was  $\text{V}_3\text{Si}$  [34, 35]. It was found that linear muffin tin orbitals (LMTO) band structure calculations gives the Fermi energy with a precision of a few millirydbergs [35]. One of the most strong hypothesis in this study was that the role of the positron wave function is minor and therefore one can neglect the deformation of the electron momentum distribution sampled by positrons. This effect was not known precisely in compounds and is recognized to play significant role in transition metals [36, 37], as well as correlation effects [38, 39]. It is only recently that this hypothesis has been proved to be valid, in the case of A15 compounds [21]. This result has been made possible by the method, due to Singh and Jarlborg [40], to calculate the two-photon momentum density within the framework of the LMTO band structure calculation. For sake of completeness, it has to be clearly stated that  $\text{V}_3\text{Si}$  has first been studied, using



2D-ACPAR (with a lower resolution) by the group of S. Berko [34] and that the results are in agreement with the results obtained at Geneva. 1D-ACPAR measurements [41] have also suggested that tight-binding band structure calculations provide a possible explanation of most of the structure appearing in the measured curves.

Some other A15 compounds have been investigated [42, 21]. As an example, we show in Fig. 1, the Fermi surface of  $\text{Nb}_3\text{Sn}$ , extracted from positron annihilation measurements [21]. One point should be made clear: positron annihilation experiments does not give the number of bands crossing the Fermi energy. Consequently, it is not possible to determine the Fermi surface sheets without a guess, or an input from band structure results. But, and it is the important point to keep in mind, once this value is fixed, it is possible to determine the shape of the Fermi surface sheets purely from the two photon momentum distribution reconstructed from 2D-ACPAR measurements. The solution is unique (due to the convention used to label the bands according to increasing energy) and comparison with the calculated Fermi surface is ruthless: any difference of the Fermi surface topology will be clearly reflected. The Fermi surface topology is independent of the algorithm of reconstruction used to deduce the two photon

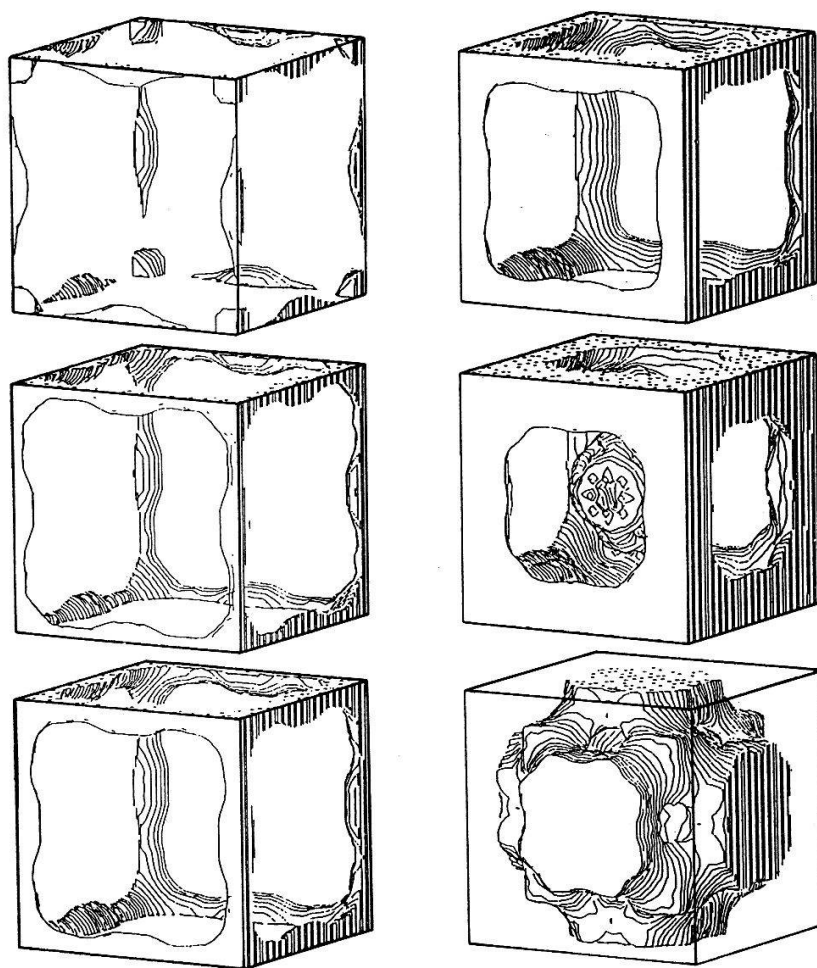


Figure 1  
Fermi surface of  $\text{Nb}_3\text{Sn}$ , as reconstructed from positron annihilation measurements [21].

momentum distribution from 2D-ACPAR. The most widely used schemes are the filtered back projection [43, 34], the Fourier transform [44] and the Cormack's method [45, 46]. Let us mention that Mijnen [47] has developed a very powerful reconstruction algorithm for 1D-ACPAR data which was used before the appearance of 2D camera. To terminate with A15 compounds, let us mention that an estimation of the parameters entering in the BCS relation of  $T_c$  has been made for Nb-Al alloys on the basis of 1D-ACPAR measurements [48]. This analysis is not very precise, being based on the free electron gas model.

The recent discovery of high- $T_c$  superconductivity by Bednorz and Muller [4] has deeply modified the situation. Many positron annihilation results have already been published [49, 50, 51, 52, 53, 54, 55], both from  $\text{La}_{(2-x)}\text{Sr}_x\text{CuO}_4$  and  $\text{YBa}_2\text{Cu}_3\text{O}_7$  [56]. All these measurements reveal some effect at or in the vicinity of the superconducting transition. Positron lifetimes, Doppler broadening, as well as 2D-ACPAR measurements are reported. Most of these works were performed on sintered samples and their analysis is made difficult by the trapping of positrons in voids, grain boundaries and internal surfaces. The only work performed on single crystals available to us so far are 2D-ACPAR distributions measured at Geneva [54], from samples grown by Damento et al. [57]. In the actual situation (everything is evolving very rapidly in this field!) one may draw the following conclusions: measurements seems to indicate that positrons are partly trapped at empty sites due to oxygen deficiencies [50, 51, 52]. The abnormal behaviour observed around  $T_c$  is generally interpreted as a manifestation of the disappearance of the superconducting state: not only the superconducting gap [52] may be responsible but also a change of the electronic structure [50] is thought to play an important role. The fact that 2D-ACPAR results [54] does not reveal significant changes in the electronic structure at the superconductive transition lead us to seriously consider the possible existence of a structural transition taking place at this temperature. This possibility has also been pointed out by Ishibashi et al. [49] who have measured Doppler broadening profiles. Some other experimental indications support this hypothesis: precise neutron diffraction [58] has revealed an anomaly of lattice parameters at  $T_c$ . Thermal expansion data lead to the same conclusion [59, 60]. An electronically driven structural anomaly in the vicinity of  $T_c$  is also suggested by sound velocity measurements [61].

Our actual thinking is that, beyond the fraction of positron trapped by oxygen vacancies, a part of free positrons probes the electron momentum density of  $\text{YBa}_2\text{Cu}_3\text{O}_7$ . It is likely that trapped positrons give an isotropic contribution to 2D-ACPAR, as found in Al single crystals [62], which is not interfering much with the electron momentum distribution. In Fig. 2 we show 2D-ACPAR results from  $\text{YBa}_2\text{Cu}_3\text{O}_7$ , folded back in the first Brillouin zone [63]. These distributions can be interpreted as the once integrated occupation number. On the same figure are presented equivalent distributions [54] calculated from the Fermi surface topology obtained by APW band structure [64]. The agreement is good proving that the Fermi surface topology is reflected by positron annihilation in high- $T_c$  compounds. The same conclusion has apparently been drawn by Tanigawa et al.

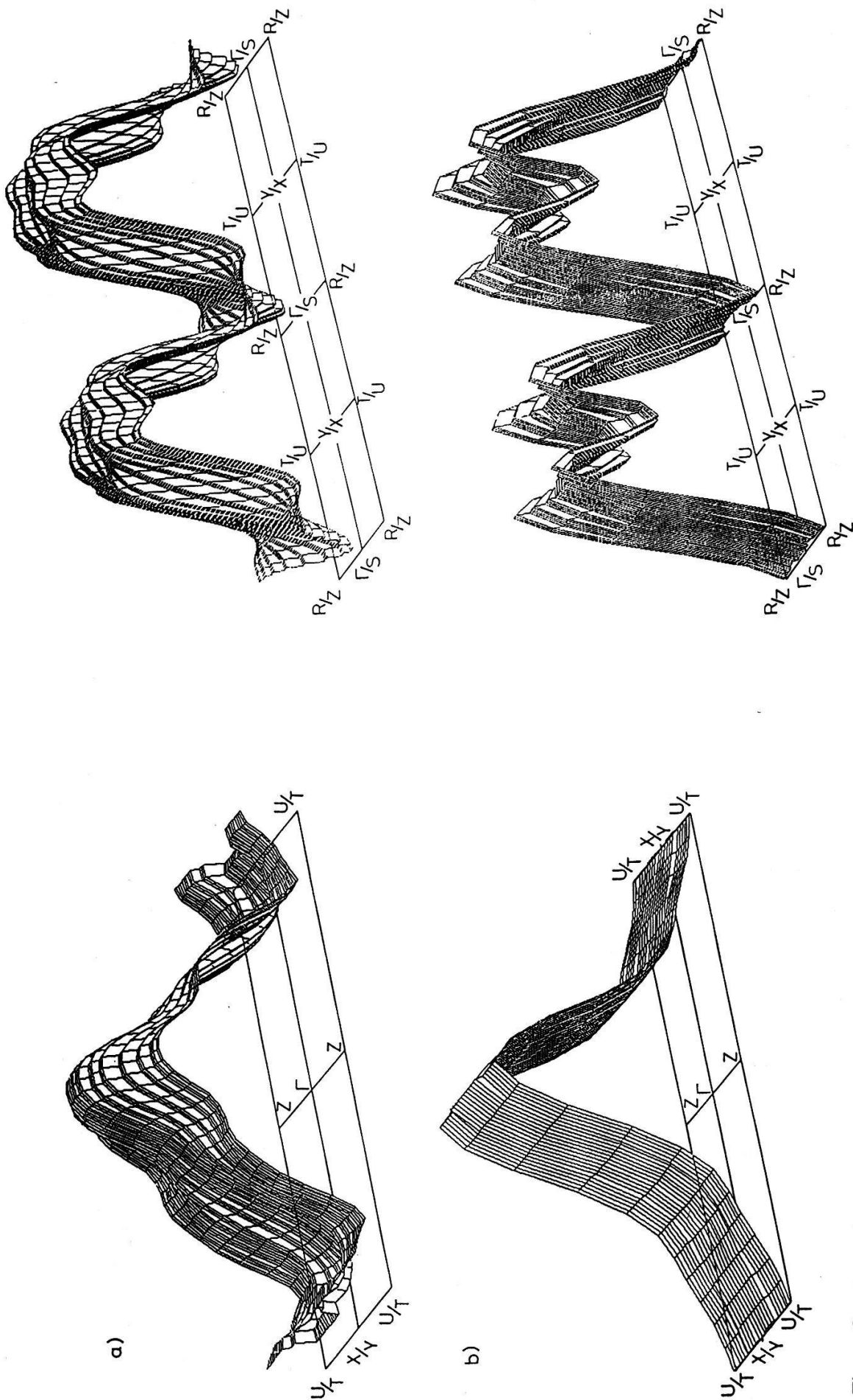


Figure 2

Once integrated electron occupation number in  $\text{YBa}_2\text{Cu}_3\text{O}_7$ . On the upper line are shown results obtained from 2D-ACPAR measurements at 77 K. Lower line shows distributions calculated from the calculated Fermi surface topology [64]. The projection is along [100] and [110] for left and right column respectively.



[65], for  $\text{La}_2\text{CuO}_4$ . Nevertheless, it is not clear why, despite the large amount of structural vacancies in  $\text{YBa}_2\text{Cu}_3\text{O}_7$ , positron annihilation reflects the topology of the Fermi surface. Calculations of the positron and electrons wave functions are now required to discuss in details experimental results.

To conclude, a direct observation of the superconductivity (smearing of the Fermi surface or a change in the thermalisation of positrons) has not been clearly established until now. Even for the results obtained in high- $T_c$  ceramics more detailed analysis are needed and a possible structural transformation may force us to interpret positron annihilation data on an other way!

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