A comparison of the temperature dependence of electron-positron momentum density characteristics in $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$, $YBa_2Cu_3O_7$ and $La_{1.85}Sr_{0.15}CuO_4$

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Abstract. The temperature dependence between 2 and 300 K of the positron–electron annihilation lifetime and the electron momentum distribution from the Doppler-broadened annihilation γ-ray energy spectrum were measured in the high-temperature superconducting material $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$. A temperature dependence is observed for the positron lifetime, below the critical temperature (124 K), suggestive of a dramatic decrease in the positron–electron overlap resulting from superconductivity. The magnitude of the observed change in the positron lifetime is the largest that we have seen for any high-T_c material, although the temperature dependence of the positron lifetime is similar to what we recently reported for $YBa_2Cu_3O_7$ and $La_{1.85}Sr_{0.15}CuO_4$. The temperature dependence of the deduced electron momentum distribution indicates a complex but small variation in the number of low-momentum electrons available to annihilate with the delocalised positrons. This new experimental result further confirms our earlier suggestion of a common physics involved in high-T_c superconductivity for perovskite materials and provides evidence for the formation of superconducting electron states which are anti-correlated with the delocalised positron wavefunction for all classes of the new high-T_c materials.

There is no universally accepted mechanism (see, e.g., [1]) to explain the observation of superconducting materials with critical temperatures greater than 30 K [2, 3], the upper limit which the conventional electron–phonon interaction can explain. The positron is particularly useful for probing the changes in electronic structure which may be associated with the superconducting mechanism [4–8]. Positron annihilation studies have shown that there is a change in the temperature dependence of the various positron annihilation indications below the critical temperature in the perovskite superconductors [4, 6], while no such change is observed in conventional BCS superconductors [9]. Our experiments with $YBa_2Cu_3O_7$ [5, 6] exhibit a variety of temperature dependences which are a consequence of the state of the positron and the defect trapping sites in the material (oxygen vacancies). We have discovered that a positron in an undisturbed Bloch state couples in a recognisable but peculiar way to the superconducting mechanism itself.

The recently discovered perovskite materials with T_c greater than 120 K [10, 11] contain Tl–O and Bi–O layers between three Cu–O layers. The question is whether or not the same 'general' behaviour reported by us for positron annihilation in

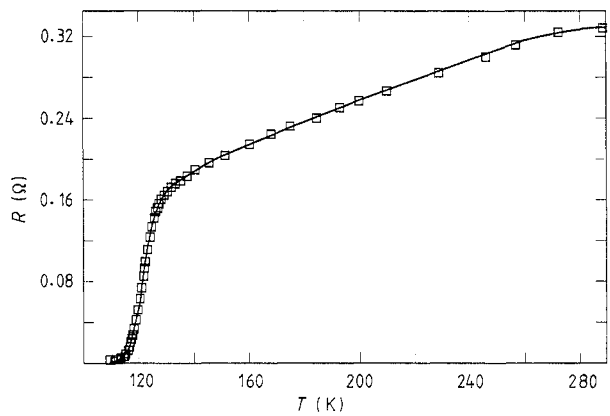

Figure 1. Resistance R against temperature T for $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$.

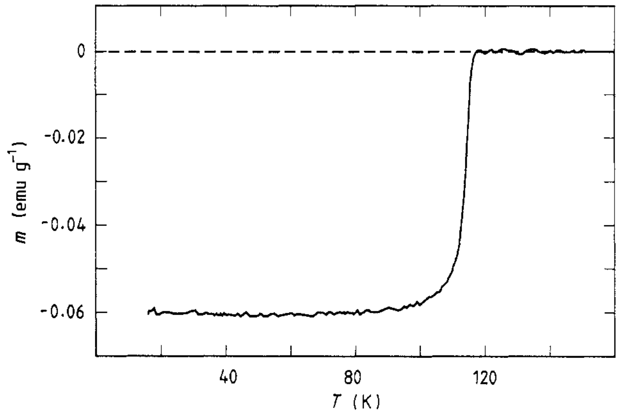

Figure 2. DC magnetisation m against temperature T for $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$. The external field equals 30 Oe.

$La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_7$ [5] would be observable for these larger-unit-cell materials as well. In this paper, we report the first experimental results on the positron lifetime and S-parameter of the Doppler-broadening energy spectra in $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$.

Polycrystalline samples of $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ were synthesised by a direct solid state reaction method [12]. X-ray powder diffraction pattern measurements displayed the $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ structure. Using four-point resistance and DC susceptibility, the onset temperatures were found to be 124 K and 120 K, respectively (figures 1 and 2). Two sintered polycrystalline samples 5 mm in diameter and 2 mm thick with a grain size of 1–5 μm were used in the present study.

The positron source (a deposition 1 mm in diameter of about 35 μCi of $^{22}NaOC_2H_5$ on an approximately 1.1 mg cm^{-2} Ni foil) was sandwiched between the two samples. The temperature of the sample was controlled and monitored with an accuracy of about 0.2 K. The positron lifetime measurements were performed using a standard fast–fast coincident circuit incorporating BaF_2 scintillators. The resolution function of the lifetime spectrometer was determined to be a sum of two Gaussians with widths (FWHW) of 250 ps

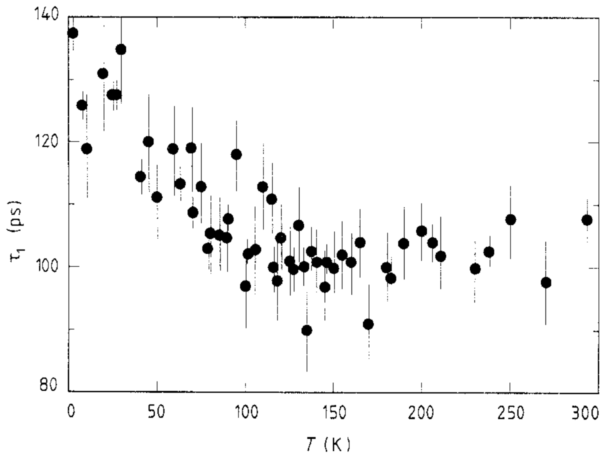

Figure 3. Positron lifetime τ_1 against temperature T for $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$. ($T_c$ = 124 K).

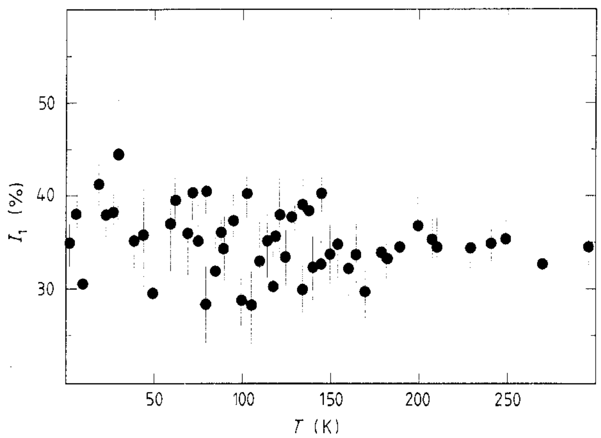

Figure 4. Intensity I_1 against T for $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ (T_c = 124 K).

(80%) and 300 ps (20%) by fitting the known ^{207}Bi lifetime (186 ps). The lifetime spectra were analysed using the PATFIT program [13]. We have used methods similar to those in [14] to determine a source correction specific to the Ni foil (about 5.7% with the lifetimes, 0.13 ns (70%) and 0.41 ns (30%)). This correction was made in the analysis of all the lifetime spectra. Each lifetime spectrum contained 1×10^6 or 4×10^6 counts, collected at a rate of 10^6 counts h^{-1}. All the lifetime spectra obtained exhibit three lifetime components with a χ^2 per degree of freedom of 1.00 ± 0.10.

The Doppler-broadening energy spectra were measured by using an intrinsic Ge detector with a resolution of 1.5 keV at 512 keV and an energy dispersion of 40 eV/channel. The Doppler-broadening spectra are reported as a parameter S which is taken as a ratio of the sum of the counts in the central region (±0.9 keV) to the total counts in the 511 keV peak. Each spectrum contains 1.5×10^6 counts collected over a period of 5 min.

Three positron lifetimes are resolved in the analysis of the lifetime spectra. The longest lifetime is 1.0–1.2 ns with an intensity of 1.0–2.3%. This lifetime is attributable to ortho-positronium (triplet positronium) annihilation. The long lifetime and intensity

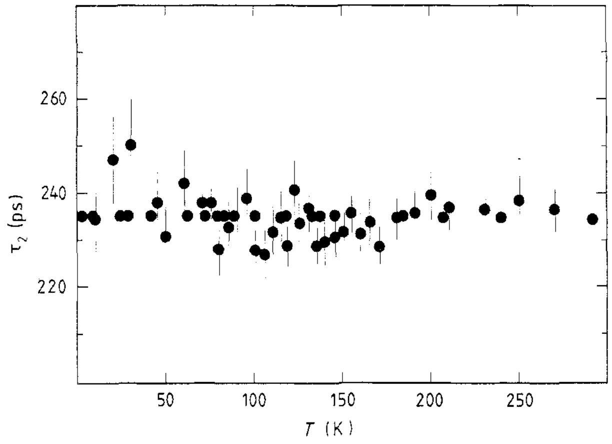

Figure 5. Positron lifetime τ_2 against temperature in $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ (T_c = 124 K).

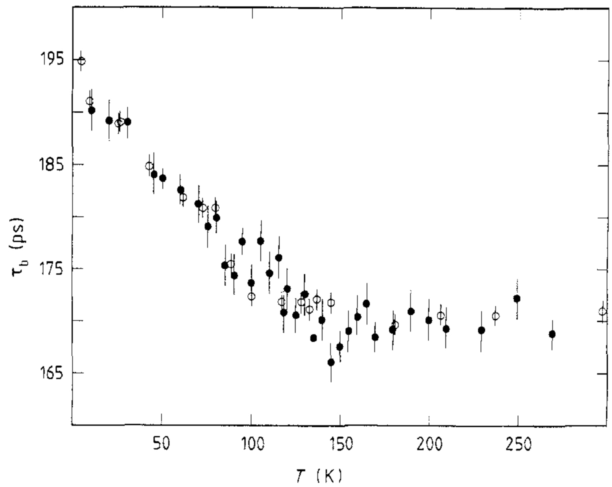

Figure 6. Positron bulk lifetime τ_b for $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ where the bulk lifetimes were deduced from a simple trapping model (equation (1)): ○, data from high statistics (4 × 10^6 counts); ●, data from medium statistics (10^6 counts).

do not vary systematically with temperature, and therefore we do not discuss this component here. The two shorter-lived positron lifetimes, one 100–140 ps and the other about 235 ps, are related to the annihilation of the positron in the bulk of the material.

Figures 3 and 4 show the temperature variations of the shortest-lived positron lifetime and its corresponding intensity with respect to temperature. τ_1 decreases monotonically from 138 ps at 2 K to 100 ps near T_c, while I_1, the intensity of this component, remains at 35 ± 5% from 2 to 298 K. As shown in figure 3, τ_1 remains nearly constant (100 ± 10 ps) for $T > T_c$. The second lifetime, τ_2 (figure 5), is found to be nearly constant (236 ± 10 ps) for 2 K < T < 298 K.

Two positron lifetimes indicate two positron states in the materials under study. A value of 236 ps for τ_2 is typical of a positron lifetime for defects or imperfections of the perovskite superconducting materials [4–6], while a value for τ_1 of 100–138 ps is rather

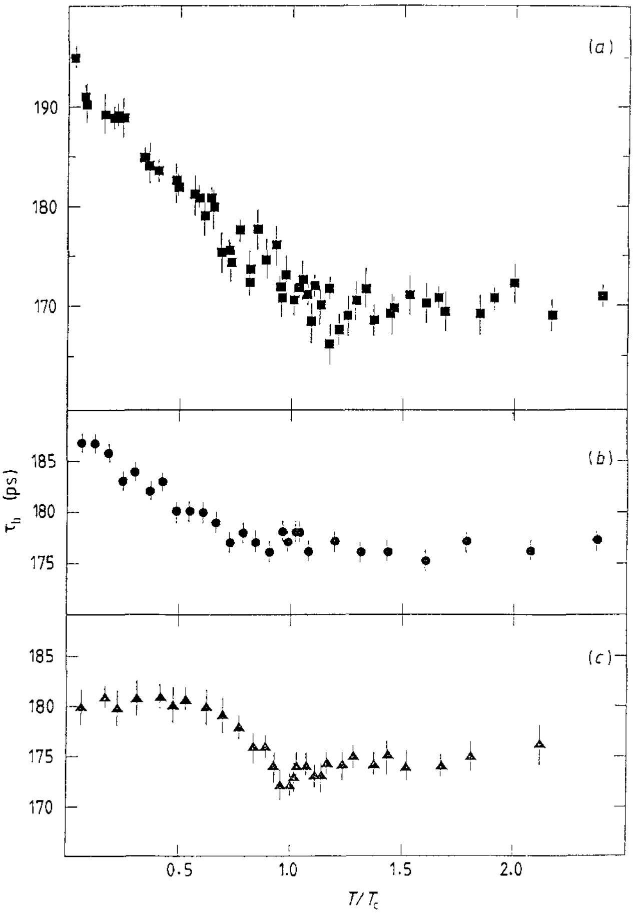

Figure 7. A comparison of the positron Bloch state lifetimes τ_b for three classes of perovskite superconductor: (*a*) $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ (T_c = 124 K); (*b*) $YBa_2Cu_3O_7$ (T_c = 84 K); (*c*) $La_{1.85}Sr_{0.15}CuO_4$ (T_c = 33 K).

too short for the bulk lifetime in the perovskite materials. However, if the positron is trapped at a vacancy defect as a result of a transition from the Bloch state, one expects to observe a short lifetime for τ_1. We invoke a two-state trapping model to interpret the two observed positron lifetimes. In this model the positron occupies a delocalised state from which it may annihilate or make a transition to a trapped state. According to this model, the delocalised positron lifetime is expressed as

$$\tau_b = (I_1\tau_1^{-1} + I_2\tau_2^{-1})^{-1} \qquad (1)$$

where I_1 and I_2 are the observed probabilities corresponding to the observed positron lifetimes τ_1 and τ_2, respectively. A plot of the deduced positron bulk lifetime against T is shown in figure 6.

We note the strong temperature dependence of τ_b for $T < T_c$, but not for $T > T_c$. This temperature dependence for τ_b is consistent with our previous determination of the Bloch state lifetime in $YBa_2Cu_3O_7$ [6] and in $La_{1.85}Sr_{0.15}CuO_4$ samples [5] where only single lifetimes were observed. A comparison of these Bloch state lifetimes against T is shown in figure 7. The present data for the $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ material suggest that

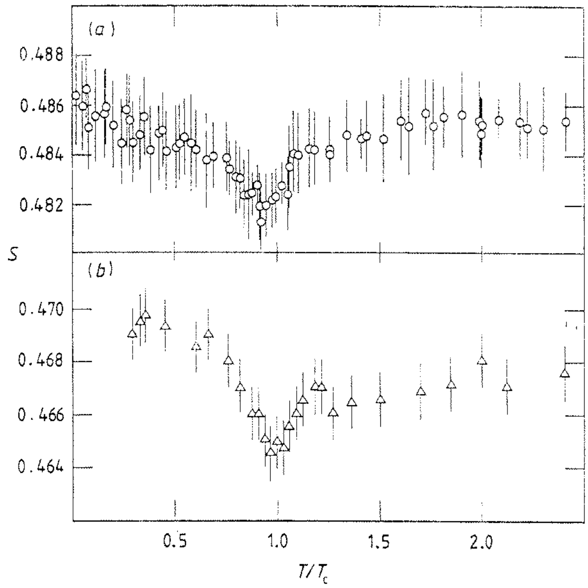

Figure 8. Doppler-broadening parameter S against temperature T for $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ (T_c = 124 K) and (*b*) $La_{1.85}Sr_{0.15}CuO_4$ (T_c = 33 K). The data for $La_{1.85}Sr_{0.15}CuO_4$ were from [5].

the trapping sites for the positron are remote from the regions of the crystal where the positron Bloch state can sample the changes associated with superconductivity. The most likely location for the Bloch state positron is in the vicinity of the Cu–O planes where it would be sensitive to the changes associated with superconductivity. We are assuming that the observed short lifetime arises from a delocalised positron Bloch-like state. Further, we know that the Bloch state of the positron will preferentially sample the more electronegative region of the crystal, giving rise to an almost 2D distribution of the position [5].

It is important to compare the relative changes in τ_0 for these three systems, i.e. $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_7$ and $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ in the superconducting state. We calculate the difference between τ_b for the normal state and τ_b extrapolated to $T = 0$ in the superconducting state. The changes in τ_b are 8 ps, 12 ps and 24 ps for $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_7$ and $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$, respectively. The large lifetime change in τ_b observed for $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ appears to indicate that our earlier speculation [6] about a possible response to a lattice dilation is unlikely to be correct. The temperature response of τ_b and the magnitude of the change between T_c and 0 K may be indicative of details of the electronic structure which are a key to the superconducting mechanism in perovskite materials. We might need to consider the possibility that the superconducting electrons are drawn from more tightly bound electrons as a consequence of a redistribution, resulting in a lowered electron–positron overlap [5]. The increase in τ_b reflects a decrease in effective electron density as seen by the positron in its delocalised state suggestive of a correlation with the opening of the superconducting gap [6]. The possibility that the positron Bloch state itself is modified cannot be ruled out. There is also a variation between the Bloch state lifetimes of the three materials for $T > T_c$, i.e. in the normal state. This variation may be indicative of the increased number of carriers, resulting from additional and more closely spaced Cu–O layers.

There is complementary evidence for the observed coupling of the positron to the superconducting phase transformation provided by the measurement of the electron momentum distribution obtained from Doppler-broadening experiments. The very similar temperature dependences of the parameter S deduced from the annihilation γ-ray energy spectra for $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$ and $La_{1.85}Sr_{0.15}CuO_4$ are shown in figure 8. Overall, we observe an increase in S (narrower momentum distribution) as the sample temperature goes to zero. However, in the vicinity of T_c the results indicate an increase in high-momentum electrons, coincident with the increase in the positron lifetime. With decreasing temperature the character of the momentum distribution 'returns' to one similar to that observed in the normal state, indicating the suggested redistribution. We remark, however, that the relative change in the parameter S in the superconducting state is approaching the sensitivity limit of this technique. We expect to observe a higher sensitivity by using the angular correlation of annihilation radiation technique. Such experiments are in progress at several laboratories including our own. One must keep in mind that the small changes in momentum distribution are accompanied by a large change in electron density distribution, and thus a redistribution of the positron wavefunction might need to be included in a self-consistent model.

In conclusion, we have observed the largest positron lifetime variation (24 ps) associated with the superconducting state by measuring the positron lifetime in $Tl_{2.2}Ca_2Ba_2Cu_3O_{10.3+\delta}$. This temperature dependence further supports our previous finding of a common superconducting positron interaction mechanism for the perovskite materials [5]. Consideration of both the lifetime and the Doppler data suggests the anti-correlation of positrons with electrons, as well as a possible coincident electron structure redistribution as T is decreased below T_c.

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