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## LETTER TO THE EDITOR

# An unusual three-quantum yield from positrons annihilated in benzene

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**Abstract.** Features of positrons annihilated in benzene were investigated by the three gamma coincidences technique, together with lifetime spectroscopy and a magnetic quenching experiment. A high three-quantum yield was found; the result reveals the presence of two positron bound states: a 'relaxed' positronium system and a more complex one.

The time annihilation spectra of positrons in liquids, when collected with high-resolution spectrometers, can show up to four components [1]. The longest one ( $\tau_4$ , of the order of a few ns), is attributed to the decay of a bound positron–electron pair, similar to positronium *in vacuo* but with an electron density at the positron,  $|\psi(0)|^2$ , which is generally modified with respect to the same quantity *in vacuo* by a factor  $\eta_4$ :

$$\eta_4 = |\psi(0)|^2 / |\psi(0)|_{\text{Ps vac}}^2.$$

Henceforth, such a pair will be called quasi-positronium (QPS). With regard to the 'intermediate' component ( $\tau_3$ , of the order of 1 ns), its origin is still under debate. Similar features can be found in the spectra of several polymers; however, in some cases, it was ascertained that  $\tau_3$  must be attributed to the decay of a positron bound system [2, 3]. Recently, Bisi *et al* [4] performed positron studies in liquid benzene and naphthalene (air degassed) using lifetime spectroscopy. They resolved the time annihilation spectra into four components and showed, through a magnetic quenching experiment, that  $\tau_4$  in liquid naphthalene comes from the decay of a 'relaxed' ortho-QPS ( $\eta_4 < 1$ ). An analogous result was found in air degassed benzene by Billard *et al* [5]; these authors deconvolved their time annihilation spectra into only three components due to the limited resolution of their lifetime apparatus. On the other hand, no certain conclusion was reached by the Bisi group on the nature of  $\tau_3$ ; actually, the magnetic quenching technique cannot be applied to investigate this problem because of the low abundance of the intermediate component with respect to  $\tau_4$ .

With the purpose of obtaining more insight into this subject, we measured the three-quantum yield in benzene, since this method can give information on the features of the positron bound systems in condensed matter; the investigation was completed through magnetic quenching measurements.

The positron source— $\text{Na}^{22}$  from a carrier-free neutral solution—was deposited between two identical Kapton foils (1.08 mg cm<sup>-2</sup> thickness); for lifetime spectroscopy, the assembly was placed within a glass cell containing the liquid benzene (Analar from BDH), which was degassed by the standard free-thaw vacuum technique.

Table 1. Lifetimes (in ns) and relative intensities in air-degassed liquid benzene.

$\tau_1$	$I_1$	$\tau_2$	$I_2$	$\tau_3$	$I_3$	$\tau_4$	$I_4$
$0.148 \pm 0.018$	$18.1 \pm 2.2$	$0.416 \pm 0.007$	$37.4 \pm 1.0$	$1.18 \pm 0.18$	$5.3 \pm 0.8$	$3.26 \pm 0.02$	$39.2 \pm 0.8$

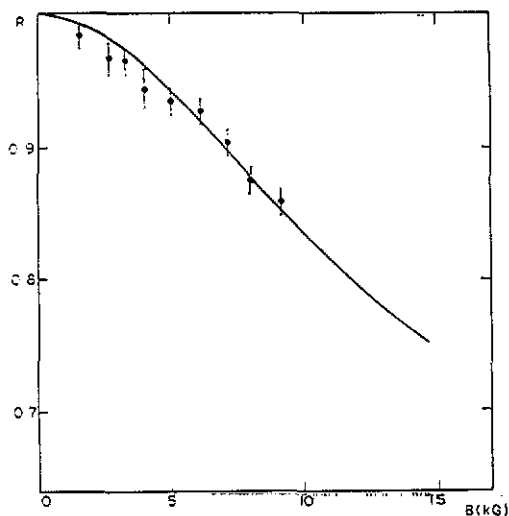


Figure 1. Quenching ratio  $R$  as a function of the magnetic field (in kG). Full curve: calculated quenching ratio with  $\eta_4 = 0.71$

The lifetime spectrometer has already been described by us [6]. We limit ourselves to mentioning the resolution, which was 190 ps; this value was raised to 320 ps by inserting light pipes for the magnetic quenching experiment. The lifetime spectra were analysed through the Positronfit Extended Program [7].

As far as the triple coincidence experiment is concerned, we used a 'double sandwich' configuration, with the positron source covered by two aluminium disks and inserted into the cell containing benzene, or, vice versa, with the two metal disks externally mounted on the cell. In this way we obtained a symmetrical configuration with regard to the scattering events concerning the photons travelling into the media. The measurements in benzene were alternated with those in aluminium, which was taken as a reference. The ratio of the spurious events to the true coincidences was 0.23, in the metal. The absolute counting rate in the metal resulted in 2.45 coincidences  $\text{min}^{-1}$ . Further information on the apparatus can be found in [8].

The time annihilation spectrum in benzene consists of four components, in accordance with [4]; the results are displayed in table 1.

In the magnetic quenching experiment the ratio  $R$  between the annihilation events in a prefixed time interval was measured in the presence as well as in the absence of the field (details on the data analysis procedure can be found in [6]). In figure 1 the shape parameter  $R$ , calculated between 3.7 ns and 19 ns where  $\tau_4$  is the only significant component, is reported as a function of the field. The data confirm that  $\tau_4$  arises from the decay of a 'relaxed' ortho-QPS, with  $\eta_4 = 0.71 \pm 0.06$ .

After this preliminary study on  $\tau_4$ , we can obtain information on the nature of  $\tau_3$  by analysing the data from the triple coincidence experiment. In fact, when QPS is formed

in matter, the three-quantum annihilation yield,  $P$ , is raised with respect to the same quantity  $P_0$  in a medium in which positrons are annihilated freely—typically a metal. The ratio  $P/P_0$  is given by the following expression:

$$P/P_0 = 372I_4[\eta_4\Gamma_t + \Gamma_p/372/\eta_4\Gamma_t + \Gamma_p + \frac{1}{3}\Gamma_p/372/\eta_4\Gamma_s + \Gamma_p] + (1 - \frac{1}{3}I_4)$$

where  $\Gamma_s$  and  $\Gamma_t$  are the PS intrinsic decay rates from singlet and triplet sublevels, respectively,  $\Gamma_p$  is the pickoff annihilation rate. We remark that  $P/P_0$  does depend on  $\eta_4$ . The measured  $P/P_0$  in benzene is  $3.61 \pm 0.10$ ; this value is higher than that expected by assuming a single QPS, associated with  $\tau_4$ , and indicates the existence of another positron bound system, which obviously correlates with the presence of the intermediate component. Such a system reveals interesting properties, which cannot be immediately explained within the usual frame. In fact, if we assume for simplicity that  $\tau_3$  represents the decay of a second ortho-QPS system, we can find its relative contact density,  $\eta_3$ , by generalizing the expression of  $P/P_0$  to include the contribution to the three-quantum yield from  $\tau_3$ . As a result, a value of  $\eta_3$  much greater than unity is obtained ( $\eta_3 = 4.50 \pm 0.95$ ), which would correspond to a 'compressed' atom. We also note that even if we assume  $\eta_4 = 0.84$ , as reported in [5], we find  $\eta_3 = 1.85 \pm 0.80$ , which is still greater than unity. In our opinion, however, it is hard to support this hypothesis; actually,  $\eta_3$  is higher than the maximum predictable value for a compressed QPS (1.3 [9] or 1.68 [10], depending on the adopted model); furthermore, it is difficult to justify the formation of such a compressed atom in a low-density medium.

An alternative explanation could be the formation of a positron complex with the molecules or radicals of the host medium. These kinds of interaction were studied by Schrader [11]; according to his calculations, the  $C_6H_6$  molecule cannot bind a positron or PS; instead, both  $e^+C_6H_5$  and  $(e^+e^-)C_6H_5$  are stable against dissociation. We observe that the radical  $C_6H_5\cdot$  can be created in the spur by the positron itself during the slowing down process. The lifetimes of  $(e^+$ -radical) complexes have never been calculated [12]; however, Kurtz and Jordan [13], on the basis of general arguments, estimated lifetimes of  $e^+$  binding molecules to be of the order of 0.1 ns. This value is one order of magnitude lower than the lifetime of our intermediate component, and makes its derivation from the positron complex  $e^+C_6H_5$  rather unrealistic. On the other hand, the lifetime of  $(e^+e^-)C_6H_5$  could be longer than that of  $e^+C_6H_5$ , in analogy with the lifetime of QPS with respect to free positrons. For these reasons, we relate the presence of  $\tau_3$  to the complex  $(e^+e^-)C_6H_5$ , whose stability has been predicted theoretically [11]. To support this hypothesis, we recall that Schrader [12] showed that the  $(e^+e^-)$  system results highly localized on the carbon missing the hydrogen; furthermore, we note that the wavefunction describing the positronic molecular orbital of such a system can be expected to be quite different from the hydrogenoid QPS wavefunction. These observations can justify the strange value of  $\eta_3$ , if we treat the  $(e^+e^-)$  structure bound to the radical as if it was the usual QPS with its characteristic contact density. It is interesting to remark that analogous results were found recently in some polymers, showing a lifetime spectrum with an intermediate component, and characterized by C-H bonds [14]. Systematic investigations with combined experimental techniques could shed more light on the stimulating problem concerning the detection of positron complexes.

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