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Anomalous magnetic quenching of quasi-positronium in solid octadecane

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Abstract. Positron annihilation in liquid as well as in solid octadecane has been investigated by means of lifetime spectroscopy combined with magnetic quenching experiments. An anomalous magnetic quenching in solid octadecane was detected, which cannot be explained in terms of the usual Zeeman effect on positronium with a modified electron contact density at the positron. A technique to extract the value of the relative contact density without making use of magnetic fields is used. The measurements indicate that the longest lifetime component both in the liquid and solid phases must be ascribed to the decay of a positron–electron bound state.

1. Introduction

The $m = 0$ ortho-type ground state sublevel of a positron–electron bound pair in condensed matter, quasi-positronium (qPs), is quenched in the presence of a static magnetic field. This fact gives a useful technique for obtaining the electron density at the positron, which is the characterising quantity for qPs [1, 2]. However, anomalously large quenching effects at low fields were recently found in some polymers [3, 4] as well as in naphthalene [5]; they cannot be explained within the usual framework, which consists of the Zeeman effect on Ps, with the addition of a single parameter to account for the perturbation, induced by the medium, of the electron density at the positron in qPs [2]. In the present paper we report the results of a similar ‘anomalous’ effect (in the sense specified above) found in solid octadecane. We extended the research to octadecane in the liquid phase, with the purpose of studying any modification of the features of the magnetic quenching with respect to the solid phase; this could give some insight into the influence of the physical structure of the host medium on the Zeeman effect in qPs. Positron annihilation in solid octadecane was at first performed with the aim of investigating the origin of the longest lived component: in fact, its identification with qPs decay was uncertain because of the lack of a narrow peak, typical of para-qPs, in angular correlation measurements [6]. Such a matter can definitely be clarified by the present results.

The impossibility of obtaining the electron density through the usual technique [2] in the presence of anomalous magnetic effects induced us to search for methods capable of giving the electron density for qPs without magnetic fields being involved. We paid attention to para-qPs lifetime, which contains, in principle, the same information as the ortho-state. The first results are also reported in this paper.

Table 1. Lifetimes and intensities of the time annihilation spectrum of positrons in liquid and solid octadecane.

Phase	τ_1 (ns)	I_1 %	τ_2 (ns)	I_2 %	τ_3 (ns)	I_3 %
Liquid	0.169 ± 0.016	11.4 ± 0.2	0.463 ± 0.031	54.4 ± 0.7	2.919 ± 0.036	34.2 ± 0.6
Solid	0.133 ± 0.007	7.5 ± 0.2	0.331 ± 0.003	70.0 ± 0.8	1.504 ± 0.015	22.5 ± 0.6

2. Experimental details

Olefin-free solid octadecane supplied by Fluka was used (purity: 99%; molecular weight: 254.50; melting point: 27–29 °C); it was prepared by melting powders without any further purification.

The positron source, about 1 μCi of ^{22}Na from a carrier-free neutral solution, was deposited between two identical Kapton foils (1.08 mg cm^{-2} thickness) glued together and inserted in the specimen in the usual ‘sandwich’ configuration.

Measurements on solid octadecane were made by changing the sample every 48 hours to avoid possible radiation damage effects. The laboratory environment was controlled during the measurements in such a way as to maintain the temperature in the range 17–20 °C. Experiments on liquid octadecane were performed at 39 °C; a temperature controller ensured stability within ± 1 °C.

The lifetime apparatus was identical to that previously used by us; details can be found in [4]. The resolution, measured as the full width at half maximum of a ‘prompt’ curve from a ^{60}Co source, gave a result of 200 ps for lifetime measurements and 380 ps for the magnetic quenching experiments. All the spectra were analysed with the POSITRONFIT extended program [7]; a suitable correction for annihilations in the supporting foils was taken into account.

3. Results and discussion

The time annihilation spectra were at first analysed into three components without any constraints. Since the longest component τ_3 is magnetically quenched in both the liquid and the solid phases, as we show below, we attributed τ_3 to ortho-qPs decay. As a consequence, a short component arising from para-qPs must be present in the lifetime spectrum; therefore, we re-analysed the spectra with the constraint $I_1/I_3 = \frac{1}{3}$ (the ratio of the statistical weights of para- to ortho-qPs), yet letting τ_1 be free. Therefore, this component represents, in our framework, the decay of para-qPs. The results are displayed in table 1. The value of the middle component, τ_2 , in the liquid phase, strictly agrees with those found in other molecular liquids [8]; according to [9], we ascribe it to the annihilation of ‘free’, i.e. unbound, positrons, and we tentatively extend this interpretation to the corresponding lifetime in the solid phase. We remark that the values of τ_3 and I_3 do not depend on the analysis (free or constrained) carried out.

Magnetic quenching experiments were performed in both phases; the results are shown in the following figures. A shape parameter—the quenching ratio R —is plotted

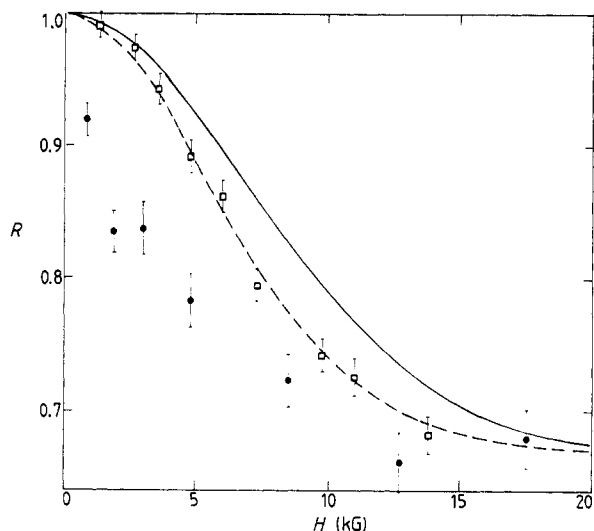


Figure 1. The quenching ratio R as a function of the magnetic field in solid (\bullet) as well as in liquid (\square) octadecane ($t_1 = 7.5$ ns, $t_2 = 14.5$ ns). Full curve: quenching ratio calculated for qPs in both phases with the same contact density as *in vacuo* ($\eta = 1$). Broken curve: best fit curve for the liquid phase ($\eta = 0.62$).

against the applied field as shown in figure 1. R is defined as the number of annihilation events occurring in a fixed time interval $t_2 - t_1$ at a given field, normalised to the same quantity at zero field. In our case the choice of $t_2 - t_1$ ensured that the only significant contribution to R was made by τ_3 . As is well known, in the presence of a static magnetic field, the $m = 0$ ortho-qPs sublevel is perturbed by the field and is progressively quenched [10]. Conversely, the $m = \pm 1$ sublevels are unaffected by the field. Therefore, if τ_3 arises from the decay of qPs, it follows that R monotonically decreases with the increasing field, attaining the asymptotic value $\frac{2}{3}$ when the $m = 0$ ortho-state is totally quenched. In contrast, if τ_3 does not represent the decay of ortho-qPs, R is insensitive to the field.

A glance at figure 1 immediately allows us to conclude that τ_3 is the lifetime of a bound positron–electron system; consequently, the extraction of the para-component from the time annihilation spectra is justified.

As mentioned in the introduction, there is no evidence of a narrow peak in the angular correlation curves for solid octadecane. A possible explanation is that the para-qPs component is broad and cannot be unambiguously identified as a ‘narrow’ one [11]; such a case would correspond to a localisation of the bound positron system, for instance at thermally produced defects—a situation already found in other molecular solids [12].

The technique adopted furnishes other information about the physical structure of qPs, which is generally modified in respect to Ps *in vacuo* because of the interactions with external electrons [13]. The result is in most cases a ‘swelling’ of the positron–electron bound pair in matter, that is, a lowering of the electron contact density at the positron, $|\psi(0)|_{\text{qPs}}^2$. We globally take this effect into account through the following, phenomenological, *ansatz*:

$$\eta = |\psi(0)|_{\text{qPs}}^2 / |\psi(0)|_{\text{Ps}}^2. \quad (1)$$

The decay rates from the singlet and triplet states can be written as [14]

$$\lambda_1 = \eta\lambda_s + \lambda_p \quad (2a)$$

$$\lambda_3 = \eta\lambda_t + \lambda_p \quad (2b)$$

where λ_s (8 ns^{-1}) and λ_t ($7.14 \mu\text{s}^{-1}$) are the intrinsic decay rates for Ps *in vacuo*, and λ_p is the pick-off decay rate.

We note that, in principle, the relative contact density can be determined from an inspection of a lifetime spectrum which shows both the para- and ortho-components.

The qPs fine structure interval ΔW results, in a first but very good approximation [15]:

$$(\Delta W)_{\text{qPs}} = \eta(\Delta W)_{\text{Ps}}. \quad (3)$$

The contact density η is generally obtained by using the Zeeman effect on qPs [1]. In fact, in the presence of a magnetic field the decay rates of the perturbed $m = 0$ triplet and singlet sublevels respectively result [2]:

$$\lambda'_3 = (\lambda_3 + y^2\lambda_1)/(1 + y^2) \quad (4a)$$

$$\lambda'_1 = (\lambda_1 + y^2\lambda_3)/(1 + y^2) \quad (4b)$$

where λ_3 and λ_1 are given by (2), $y = (\sqrt{1 + x^2} - 1)/x$ and $x = 4\mu_B H/\Delta W$ (μ_B is the Bohr magneton and H the applied field). Since R is a function of η related through equations (2) and (4) [4], a fit of the calculated quenching ratios to the experimental ones generally furnishes the value of η .

We note in figure 1 the entirely different behaviour of R at low fields for the solid phase in respect to the liquid one. The measured R -values for liquid octadecane are well fitted by the broken curve which is obtained for $\eta = 0.62 \pm 0.05$. The full curve represents the quenching ratio calculated for unperturbed qPs (that is, with $\eta = 1$) in both phases, since R is barely sensitive to the pick-off decay rate.

The dependence of the perturbed $m = 0$ ortho-qPs lifetime $\tau'_3 = 1/\lambda'_3$ on η (equation (6)) allows us to obtain another determination of the contact density in liquid octadecane [16]. τ'_3 can be directly extracted from the lifetime spectra which show four components in the presence of the field H . We imposed the constraint $I'_3(H) = I_3(H)/2$, according to the statistical weight of the $m = 0$ ortho-state with respect to the $m = \pm 1$ sublevels. Furthermore, we fixed τ_2 at the value obtained at zero field, since the lifetime of unbound positrons cannot show any magnetic effect. In figure 2 the experimental values of τ'_3 for liquid octadecane are displayed as a function of the field, together with the best fit curve—the broken one—obtained for $\eta = 0.69 \pm 0.06$. Such a value is in agreement, within the errors, with the previous determination and allows us to check the consistency of our fitting procedure. However, we consider the use of the quenching ratio R to obtain η to be more general. In fact, the extraction of τ'_3 is only possible when the qPs yield is quite large and, furthermore, when a single component in the lifetime spectrum is due to the decay of ortho-qPs, in the absence of the field. For instance, it would be very difficult to analyse a time annihilation spectrum in Teflon—in the presence of magnetic fields—where there is experimental evidence that the two longest components have to be ascribed to qPs decay [17, 18].

The results for solid octadecane shown in figure 1 are completely different. We remark that the overlap of the results is possible since the time interval chosen is the same in the two phases. For the solid one a different choice of t_1 would have produced better statistics, but prevented us from directly comparing the values of R in the liquid and solid specimens. The sudden fall of the quenching ratio at fields below 5000 G makes a reasonable fit for any value of η impossible. This anomalous behaviour can be observed from another point of view in figure 2, where the values of τ'_3 are plotted versus the applied field. The large quenching of τ'_3 , even at low fields, greatly decreases the number

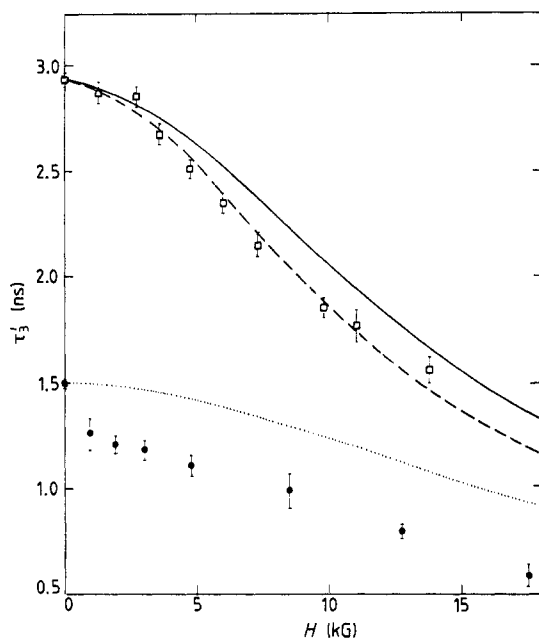


Figure 2. The lifetime of the perturbed ortho-qPs sublevel versus the applied field in liquid (□) as well as in solid (●) octadecane. Full curve: lifetime of $m = 0$ ortho-qPs in the liquid phase with $\eta = 1$; broken curve: best fit curve for the liquid phase ($\eta = 0.69$); dotted curve: lifetime of an $m = 0$ ortho-qPs in the solid phase with $\eta = 1$.

of annihilation events coming from the $m = 0$ ortho-sublevel in the time interval $t_2 - t_1$; the main contribution is due to the remaining $m = \pm 1$ ortho-states and the quenching ratio rapidly reaches its asymptotic value.

At present, we are not able to explain this result through a satisfactory physical model. Therefore, we limit ourselves to the following, qualitative consideration. The magnetic quenching effect on solid octadecane could be interpreted in terms of a stronger mixing of the $m = 0$ sublevels than that produced by the external applied field, as if an additional local field was present. Such a field could arise from the magnetic moments of the spur products. Since the diffusion times of particles outside the spur, as well as the recombination times [19], are much shorter than typical qPs lifetimes, no local fields will be ordinarily expected to act on qPs: the magnetic quenching effect would be generally explained in the framework of the usual theory, as happens in the liquid octadecane. However, qPs could interact with triplet excited states of the host structure created during the slowing-down of the positron itself (molecules or excitons, for instance); an applied magnetic field would orient their magnetic moments—otherwise randomly oriented—and the consequent local field would increase with the external one.

In this connection, it is interesting to observe that a magnetic quenching similar to that found in solid octadecane was detected in naphthalene. In this organic molecular material it is known that triplet excitons are formed even at room temperature and remain localised in the site of their formation; furthermore, their lifetimes can be much longer than that of qPs. Of course, further experimental investigation is necessary to test the foundation of the above argument; we only remark that interactions between positron and spur products were recently invoked to explain certain features of positron lifetime spectra in some alkali halides [20].

The anomalous trend of the quenching ratio R as well as of τ_3 does not allow us to obtain the relative contact density η by means of a single-parameter fit. Nevertheless,

the value of η can be, in principle, extracted from the para-component present in the time annihilation spectrum as explained above (equations (2a) and (2b)). By using for τ_1 and τ_3 the values displayed in table 1 we find $\eta = 0.70 \pm 0.07$ and $\eta = 0.86 \pm 0.09$ for the liquid and solid phase respectively. We note the satisfactory agreement among the three different determinations of η in the liquid octadecane. Unfortunately, we cannot at present compare the value of η in solid octadecane with others obtained through different techniques; however, the value found can give an initial idea about the perturbation on qPs in this structure. Obviously, the reliability of this method could increase in the future with the development of timing spectrometers with excellent resolution [21].

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