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Three quantum annihilations in silicone fluids

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Abstract. Measurements on the three photon decay component in silicone fluids indicate that there is a detectable three photon yield contrary to what is observed in most other liquids. Degassing of the fluids increases the three photon yield. There is little or no effect on the three photon yield in silicone fluids with change in molecular weight.

1. Introduction

Positron annihilation studies in different materials have been made quite extensively by many groups and a very good collection of the results is now available in tabular form (Hogg *et al* 1968). Most of these data have been obtained from lifetime measurements and angular correlation experiments and very little data exist on the direct observation of the three quantum decay of positronium and those that do exist are only in a few oxides and gases. Probably it was thought that in liquids the triplet state cannot exist long enough to emit three photons and that the triplet state is quenched completely by pick-off or other mechanisms so that all annihilations in liquids take place via two photon annihilations. Probably because of this, few three quantum annihilation experiments have been attempted and only in a few liquids. From our earlier work on positron lifetime measurements in silicone fluids (Sen and Patro 1969a) and in tetrachlorides (Sen and Patro 1969b) it was observed that the substitution of carbon (core) by silicon greatly enhances the triplet positronium formation.

In our earlier attempt to observe the effect of molecular weight on τ_2 and I_2 in silicone fluids†, though a small change was found in τ_2 no such change could be detected in I_2. As the origin of I_2 is pick-off from the triplet state, a small percentage decrease (of the order of 2%) in I_2 will appear as a 2% three photon decay and may easily be detected in an experiment aimed at detecting three photon decays and not as a change in I_2 if the total positronium formed is constant. Thus, the results of previous workers (Gray *et al* 1967, Sen and Patro 1969a and Tao and Chuang 1969) that there is no change in I_2 because of the presence of dissolved oxygen can be tested. Therefore, it was felt worthwhile to measure the three photon annihilation in silicone fluids normal and degassed and measure any change due to degassing and change in the molecular weight.

† We follow the general convention of positron annihilation lifetime measurements and denote τ_1 and I_1 as the lifetime and intensity attributed to free annihilation ($\tau_1 \simeq 0{\cdot}3$ ns) and τ_2 and I_2 as the lifetime and intensity ($\tau_2 \simeq 1$–3 ns for condensed matter) attributed to orthopositronium decaying by the pick-off process.

2. Experimental techniques

Three quantum annihilation estimates were made by using the method of three-to-two-photon relative yields. For these measurements a Ge(Li) detector assembly with a resolution of 3 keV was used. The source used was ^{22}Na deposited and sandwiched between two 0·3 mil thick nickel foils. The silicone fluids used were dimethyl polysiloxane fluids of different viscosity grades. The details regarding formulae, molecular weights and structure of these fluids have been already given (Sen and Patro 1969a) and hence will not be repeated here.

Degassing of these fluids was achieved by heating the sample under study to approximately 180 °C for an hour or so and then cooling the sample and inserting the source in the sample without disturbing the vacuum. The standard vacuum–freeze–thaw-technique for degassing was not used since the present method had been tested earlier and is also very convenient.

The three photon relative yields have been determined with respect to those of aluminium where it is known that no three photon annihilations take place (to be exact only 0·27% 3γ). The standard technique of using Al outside while taking the sample run and sample outside while taking the Al run was adopted to minimize the scattering and selfabsorption effects. Both methods 'valley-to-peak' and 'peak-to-peak' were used to get the three photon estimates as was done by previous workers (Bussolatti and Zappa 1964, Gainotti *et al* 1964a, 1964b and Sen and Patro 1969c). Normalization of the two runs was made from Compton distribution (over 240 channels) of the 1·28 MeV nuclear gamma ray from the ^{22}Na source. The boundaries used for the valley are from 380 keV to 480 keV. In every run at least 80 000 counts were accumulated in one channel at the peak with an expansion of 1·5 keV per channel.

3. Results and discussions

The degassed silicone fluids showed quite high abundances in three photon annihilations ($\simeq$3·0–4·0%). Typical gamma ray spectra between 350 keV and 520 keV, obtained using the Ge(Li) detector, for degassed 50 cSt silicone fluid and for aluminium are displayed in figure 1 with peak counts normalized. The valley of the silicone fluid curve is clearly seen to be higher than that of the Al curve showing the presence of the three photon annihilations from the fluid. The results of three-to-two photon relative yields obtained from valley-to-peak and peak-to-peak methods for all the degassed samples studied have been collected in table 1. The percentage of three photon relative yields in all the undegassed samples lies between 1 and 2% with an error of $\pm$0·3% and has not been included in table 1. The valley-to-peak percentages have been obtained by normalizing the valley of degassed 350 cSt to 3·6% which has been obtained directly from the peak-to-peak method.

The results clearly show that there is no correlation between the triplet positronium formation and the viscosity or molecular weight of the sample. Another result of this work is that the presence of dissolved oxygen in the sample decreases the three photon emission. In the present investigation, a finite change in the three photon percentage between undegassed and degassed silicone fluids, though quite small (ie from 1–2% to 3·0–4·0%) has been observed and this small change was not seen previously (Sen and Patro 1969a) in the large pick-off component ($\simeq$40–45%) probably because of the inherent difficulty in observing a small variation in a large quantity. There are a

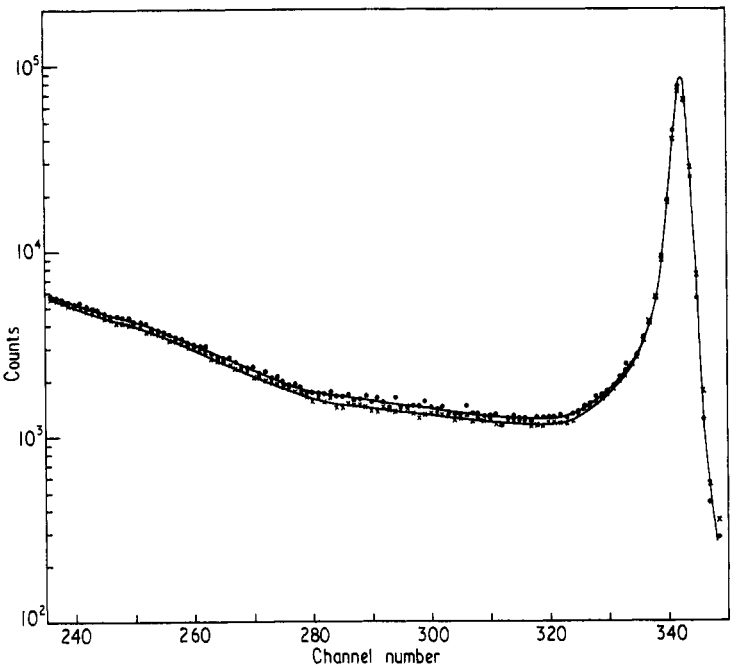

Figure 1. Annihilation spectra obtained with source sandwiched in degassed silicone fluid (50 cSt) in Al outside (●), and Al in silicone fluid outside (×), using the Ge(Li) detector set-up. 1 channel = 1·5 keV.

Table 1. The results of three photon relative yield measurements using a Ge(Li) detector assembly

Sample (degassed)	Viscosity (cSt at 25 °C)	Percentage of three photon relative yields	
		valley-to-peak†	peak-to-peak
DC 200	10	3·4 ± 0·3	3·7 ± 0·4
DC 200	50	3·8 ± 0·3	3·5 ± 0·4
DC 200	100	3·4 ± 0·3	3·7 ± 0·4
DC 200	350	3·6 ± 0·3	3·6 ± 0·4
DC 200	1000	3·3 ± 0·3	3·4 ± 0·4
DC 200	100000	3·8 ± 0·3	3·9 ± 0·4
DC 200	1000000	3·0 ± 0·3	3·2 ± 0·4
$SiCl_4$ (undegassed)		2·5 ± 0·3	2·0 ± 0·4

† The value of 3·6 % for 350 cSt sample has been taken from the peak-to-peak ratio method.

few liquids where the pick-off component is more than 40 %. They are toluene (≃45 %), heptane (≃40 %) and benzene (≃41 %). But in all these liquids the three photon percentage is less than 0·5 %, as observed in the present work, indicating that practically all the triplet state had been quenched to the singlet state. Figure 2 shows the gamma ray spectra taken with toluene and Al samples. The two spectra practically overlap each other giving evidence of the absence of three photon annihilations from the toluene sample.

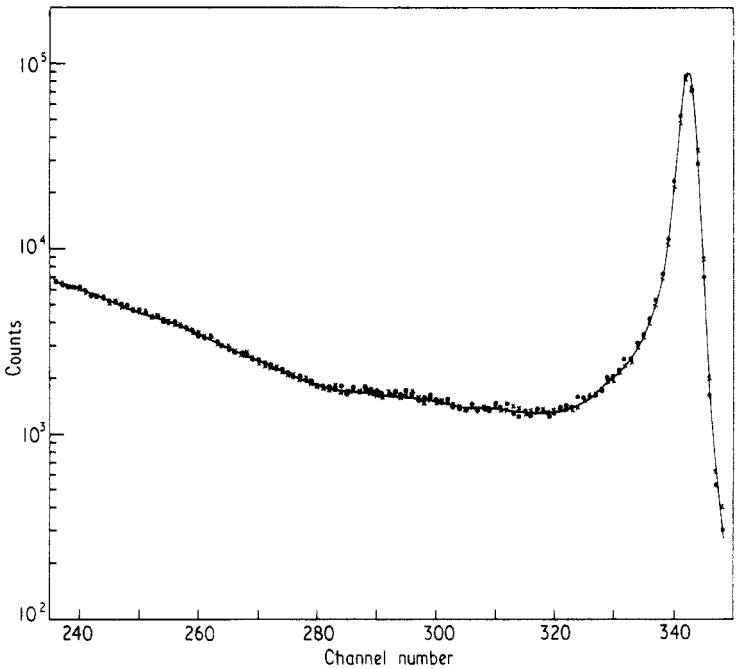

Figure 2. Annihilation spectra obtained with source sandwiched in toluene in Al outside (×), and Al in toluene outside (●), using the Ge(Li) detector set-up. 1 channel = 1·5 keV.

The fact that such high three photon annihilations take place from silicone fluids, including $SiCl_4$ (see table 1), may be very significant and worth investigating. In showing such high three photon percentages these silicone fluids behave somewhat similarly to oxides. Large cavities may be present in these liquids to allow the positronium to exist long enough to emit three photons. Unlike oxides, where surface effects are very important, in liquids these effects play no role. Our results merely support our previous finding that silicone fluids remain special in showing such high triplet positronium formations. Experiments are being performed to measure directly the singlet and triplet lifetimes in these silicone fluids and will be reported later.

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