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Application of age-momentum correlation measurements for studies of the formation of positronium and its reactions in liquids

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Abstract

The age-momentum correlation measurements were applied to the study of annihilation characteristics of positrons and positronium (Ps) in benzene, carbontetrachloride and those mixtures. The observed lifetime spectra and Doppler broadening profiles were decomposed into two-components. The inhibition of the Ps formation by carbontetrachloride was observed in a momentum dependence of the lifetime and in a time dependence of the S parameter.

1. Introduction

It is well known that positronium (Ps) is formed when energetic positrons are injected into liquids. The formation of Ps is usually detected by the long-lived component in lifetime spectra of positrons. Analysis of the lifetime spectra enables us to obtain information on chemical reactions between Ps and molecules. However, since the lifetime spectrum for liquids is the superposition of various annihilation modes, the decomposition of the short-lived components is difficult. Carbontetrachloride is known to be an efficient inhibitor of the Ps formation, and the inhibition of the Ps formation was studied by the analysis of the long-lived component in lifetime spectra.¹⁻³ It has been suggested that the age-momentum correlation measurement is a powerful tool to separate some coexistent annihilation modes.⁴ Thus, in the present paper, we applied the age-momentum correlation measurements in order to study annihilation characteristics of positrons and Ps in benzene, carbontetrachloride and their mixtures.

2. Experimental

The specimens used in the present experiment were benzene, carbontetrachloride and their mixtures. These specimens were degassed and transferred into cylindrical glass vessels. The age-momentum correlation spectra were measured by the system constructed at the University of Tsukuba.⁴ The obtained lifetime spectra were decomposed into two components by using RESOLUTION⁵. The Doppler broadening profiles were characterized by the S parameter, where the central region of the spectrum was defined from ~ 510.5 keV to ~ 511.5 keV.

3. Results and Discussion

Figures 1 and 2 show the momentum dependence of the mean lifetime, τ_M , and the time dependence of the S parameter for benzene, carbontetrachloride and those mixtures, respectively. For carbontetrachloride, the momentum dependence of τ_M and the time dependence of S were nearly flat. This means that positrons annihilate from only one state in carbontetrachloride. Since the obtained lifetime is shorter than that corresponding to the pick-off annihilation of ortho-Ps, it can be concluded that Ps is not formed in carbontetrachloride. For benzene and benzene-carbon tetrachloride solutions, the value of τ_M at $|\Delta E| = 2 \sim 4$ keV and that of S at $t = 0$ ns were found to increase. Figures 3-6 show the momentum dependence of the first lifetime, τ_1 , and that of the second component, I_2 , for each specimen. Figure 7 shows the time dependence of the S parameter corresponding to the short-lived component for these specimens, where the values denoted by S_{τ_1} , S_{τ_2} , S_{tot} are the average values of S for the short-lived

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Fig. 1. The momentum dependence of τ_M for benzene carbontetrachloride solutions.

Fig. 2. The time dependence of the S parameter for benzene carbontetrachloride solutions.

Fig. 3. The momentum dependence of the lifetime of the short-lived component, τ_1 , and the long-lived component, I_2 for benzene.

Fig. 4. The momentum dependence of τ_1 and of I_2 for 0.02 vol.% CCl_4 in C_6H_6 .

Fig. 5. The momentum dependence of τ_1 and of I_2 for 0.2 vol.% CCl_4 in C_6H_6 .

Fig. 6. The momentum dependence of τ_1 and of I_2 for 1.0 vol.% CCl_4 in C_6H_6 .

Fig. 7. The time dependence of the S parameter corresponding to the short-lived component for benzene carbontetrachloride solutions.

between benzene and carbontetrachloride. From Figs. 4-6, it can be seen that addition of carbontetrachloride causes a decrease in the dips of both τ_1 and I_2 at $|\Delta E| = 0$ keV. This corresponds to the decrease in the intensity of the mode with the narrowest momentum distribution and the shortest lifetime, that is, the decrease in the intensity of self annihilation of para-Ps. The overall decrease of I_2 also corresponds to the decrease in the intensity of the pick-off annihilation of ortho-Ps. In Fig. 7, the value of S at $t = 0$ ns was found to decrease with increasing the content of carbontetrachloride. This is also attributed to the inhibition of the Ps formation by carbontetrachloride. No drastic change in the S - t plots and in τ_1 - ΔE plots due to the reaction between positrons and carbontetrachloride was observed.

4. Conclusion

We have presented the application of the age-momentum correlation measurement for the study of the formation of Ps and its reaction in benzene, carbontetrachloride and those mixtures. From the measurements, it was found that at least five annihilation modes coexisted for benzene and benzene-carbon tetrachloride solutions. The inhibition of the Ps formation by carbontetrachloride was successfully observed in the time dependence of S corresponding to the short-lived component and the momentum dependence of τ_1 .

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component, that for the long-lived component and that for τ_M , respectively. In Figs. 3-6, the τ_1 - ΔE plots can be reproduced when $S_1 > S_2 > S_3 > S_4$ and $\tau_1, \tau_3 < \tau_2, \tau_4$, where S_i and τ_i are the characteristic value of S and the lifetime for the i -th annihilation mode, respectively. The first annihilation mode has the narrowest momentum distribution and the shortest lifetime. Thus, this component can be associated with the self-annihilation of para-Ps. The fourth component can be assigned to the annihilation of free positrons. The second and third components may be associated with the annihilation of para-Ps with a rather wide momentum distribution or the compound formation between Ps or positrons and molecules. Because the second lifetime is long (2.3-2.4 ns), I_2 is associated with the pick-off annihilation of ortho-Ps.

In Fig. 7, an increase in value of S at $t \approx 0$ ns and a dip in the S - t plot at $t \approx 3$ ns were observed. Those are attributed to the self-annihilation of para-Ps and to the annihilation of free positrons, respectively. From Fig. 7, it was found that the characteristic value of S for the annihilation of free positrons is smaller than S_{τ_2} . This suggests that the momentum distribution of electron-positron pairs observed by the annihilation of free positrons is broader than that by the pick-off annihilation of ortho-Ps. The results in benzene containing carbontetrachloride are intermediate