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THE POSITRONIUM ATOM IN ORGANIC LIQUIDS

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An experimental study has been made of the correlation curves and time spectra of positron annihilation in a number of organic liquids. The results obtained are interpreted on the basis of the bubble model, according to which positronium atoms, interacting with molecules of the liquid, repel them, forming a "bubble"—a potential well of a certain depth and radius. These two parameters have been calculated on the basis of data obtained by two methods (correlation and time). Practically identical values are obtained in the two cases, which favors the model assumed (the well radius in hydrocarbons, alcohols, and bromides is 4–4.5 Å; the well depth in hydrocarbons, alcohols, and bromides is 0.7–1 eV.

I. INTRODUCTION

In discussion of the system consisting of an atom of positronium and a liquid, we can distinguish two extreme cases:

1. Weak interaction of the positronium atom with the medium. The change in the liquid structure as the result of this interaction can be neglected.
2. Strong interaction. In this case the repulsion between the positronium atom and the liquid turns out to be so great that the formation of a cavity in the liquid as the result of this repulsion, and localization of the positronium atom inside the cavity, can lead to a state with a free energy lower than the state without change of the liquid structure. In this case the bubble model is evidently applicable to description of the behavior of the positronium.

The principal idea of the model is that a stable configuration of the medium- (positronium atom) system is achieved as the result of equilibrium which occurs, on the one hand, as the result of repulsion of the positronium atom from the surrounding molecules and, on the other hand, as the result of compressive forces of the bubble, arising as the result of surface tension.

In the work of Ferrell,[1] who used the bubble model to calculate the probability of pick-off annihilation of positronium in liquid helium, a result was obtained in good agreement with experiment. He assumed that the well depth was infinite and that inside the bubble there is saturated vapor in which the positronium annihilation occurs. However, recently Stewart and Briscoe[2] checked the applicability of Ferrell's model, studying correlation curves in a number of compressed gases. The radii of the wells, calculated according to the model presented by Ferrell,[1] turned out to be 30% smaller than those obtained by Briscoe and Stewart from analysis of correlation curves. Roellig[3] perfected Ferrell's model, showing that helium molecules cannot enter inside the bubble and that the pick-off annihilation of positronium is due to its quantum-mechanical leakage outside the bubble—a well of finite depth.

In this paper the applicability of the bubble model to organic liquids is studied. This study is carried out by two means: for each material, values of the depth and radius of the well are determined initially from analysis of the momentum distribution of singlet positronium atoms, which was obtained after appropriate analysis of the correlation curve (the correlation method), and then by means of the bubble model and experimentally obtained probabilities of positronium pick-off annihilation (the time method).

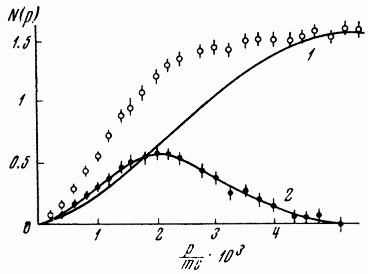
II. EXPERIMENTAL PART

Measurements of the angular correlation of annihilation γ rays and of the lifetime of the long-lived component of the time spectrum of annihilation in the ten organic liquids listed in the table were carried out in the apparatus described in detail previously.[4,5]

The angular resolution function of the equipment for study of the annihilation radiation angular correlation is well approximated by a Gaussian distribution with a mean-square deviation $\sigma = 1$ mrad. The isotope Na^{22} with an activity of 5 millicuries was used as a source. The accidental-coincidence background and the contribution from positron annihilation in the chamber walls together did not exceed 3% of the total number of coincidence pulses. The number of coincidences in the peak of each curve was at least 2000. The resolution of the time equipment was 1 nsec or better, and one channel of the time analyzer corresponded to 0.1 nsec.

In the work in the time equipment, all materials studied were subjected to necessary processing to remove dissolved oxygen, which was not necessary in the work with angular correlations, since the presence of oxygen does not affect the shape of the narrow compo-

Material	Correlation method			Time method				
	kR	R, Å	U, eV	kR	R, Å	U, eV	$\lambda_p/N \times 10^{14}$ sec^{-1}	Z_{eff}
Hexane	2.2	4.5	0.75	2.27	4.6	0.78	5.56	38
Decane	2.25	4.2	0.90	2.28	4.3	0.90	9.28	62
Dodecane	2.25	4.2	0.94	2.28	4.2	0.94	11.5	74
Hexadecane	2.25	4.1	0.91	2.27	4.1	0.98	15.2	98
Ethanol	2.2	4.3	0.77	2.27	4.4	0,81	2.27	20
Butanol	2.2	4.3	0,77	2.27	4.3	0.90	4.65	32
Octanol	2.2	4.2	0.81	2.28	4.1	0.94	8,21	56
Iso-propanol	2.3	4.4	0.94	2.27	4.45	0.89	3.72	26
C_3H_7Br	2.2	4.4	0,84	2.12	4.1	0.87	4.60	20
$C_{10}H_{21}Br$	2.25	4.0	1,0	2.19	3.7	1.1	12.9	68
Relative error	±7%	±7%	±14%	±2,5%	±2.5%	±5%	±2%	—



Momentum distribution of parapositronium atoms: O—experimentally obtained total momentum distribution L(p—; ●—experimentally obtained momentum distribution of parapositronium atoms N(p) (the narrow component of the correlation curve). 1—curve for the broad component M(p) calculated by the Coulson-Duncanson method; 2—momentum distribution curve for parapositronium atoms calculated from Eq. (6).

nent of the correlation curve, but is important just in consideration of the momentum distribution of singlet positronium atoms.

The correlation curves of annihilation γ-ray coincidences give complete information on the nature of the distribution of $f(p_z)$ projections of the momenta of the center of mass of the annihilating e^+e^- pairs on the Z axis, which is perpendicular to the XY plane and which passes through the source (the point O) and the collimating slit of the fixed γ-ray counter (parallel to the X axis and perpendicular to the Y axis). Since in work with ordinary liquids the samples studied are completely isotropic, by proceeding from the shape of $f(p_z)$ it is easy to obtain also (without any additional experiments) the distribution function L(p) for the total momenta of the center of mass of the annihilating e^+e^- pairs.

In order to separate the narrow component of the correlation curves, it is necessary to know the shape of the wide component. Following Kerr et al.,[6] we calculated the shape of the wide component M(p) by the Coulson-Duncanson method.[7] The distribution obtained was matched with that part of the experimental L(p) curve where the contribution of the narrow component can clearly be neglected—in the momentum region satisfying the relation $5 \times 10^{-3} \le p/mc \le 7 \times 10^{-3}$ (m is the electron mass). Further, from the experimentally obtained L(p) curve we subtracted the matched M(P) distribution, and the result obviously represents the narrow component of the momentum distribution M(p), i.e., the momentum distribution of atoms of parapositronium, which are shown as an illustration for decane in the figure.

III. THEORETICAL PART

1. Correlation method. Following the arguments of Roellig,[3] we will assume that as the result of the exchange interaction a bubble is formed around the positronium atom in the liquid, whose properties relative to this atom can be approximated by a spherical well with rectangular walls, of depth U and radius R.

We will write the positronium wave function for this case in the momentum representation:

$$a(p)=\frac{C}{pR/\hbar}\,\frac{(\varkappa R)^2+(kR)^2}{(\varkappa R)^2+(pR/\hbar)^2}\left[\frac{\sin^2(kR-pR/\hbar)}{kR-pR/\hbar}-\frac{\sin(kR+pR/\hbar)}{kR+pR/\hbar}\right]. \tag{1}$$

Here $-\kappa = k \cot kR$,

$$C^2=\text{const}\cdot R^{-1}\left(1-\frac{1}{2kR}\sin 2kR+\frac{\sin^2 kR}{\varkappa R}\right)^{-1}, \quad k=\sqrt{\frac{4mE}{\hbar^2}},$$

and E is the positronium energy (with respect to the bottom of the well).

The momentum distribution of the atoms of parapositronium (for the natural condition of spherical symmetry of the wave function) is written in the form

$$Q(p)=4\pi p^2|a(p)|^2. \tag{2}$$

For comparison of Eq. (2) with experiment, it is necessary to take into account the Gaussian angular resolution function of the equipment. The experimentally obtained momentum distribution then acquires the form

$$N(p)=Bp\frac{d}{dp}\int\limits_{-\infty}^{\infty}\exp\left[-\left(\frac{p}{mc}-\theta\right)^2\Big/2\sigma^2\right]A(\theta)d\theta, \tag{3}$$

where $A(\theta)$ is the narrow component of the correlation curve for zero width of the collimator slits,

$$A(\theta)=B'\int\limits_{\theta}^{\infty}\frac{1}{q}Q(q)dq=B'\int\limits_{\theta}^{\infty}q|a(q)|dq. \tag{4}$$

Substituting (4) into (3) and performing the differentiation, we obtain

$$N(p)=B''p\int\limits_{-\infty}^{\infty}\exp\left[\left(-\frac{p}{mc}-\theta\right)^2\Big/2\sigma^2\right]\left(\frac{p}{mc}-\theta\right)\int\limits_{\theta}^{\infty}q|a(q)|^2dq\,d\theta. \tag{5}$$

Performing an integration by parts in Eq. (5), we finally obtain

$$N(p)=B'''p\int\limits_{0}^{\infty}\exp\left[-\left(\frac{p}{mc}-\theta\right)^2\Big/2\sigma^2\right]\theta|a(\theta)|^2d\theta, \tag{6}$$

where $a(\theta)$ is written according to Eq. (1) for $p = \theta mc$.

The parameters kR and R entering in Eq. (6) were determined by the method of least squares so as to provide the best agreement of a function N(p) of the form of Eq. (6) with the experimental momentum distribution of singlet positronium atoms obtained by the means described in section II.

2. Time method. According to Roellig,[3] the probability of pick-off annihilation of positronium in one molecule of the medium (λ_p/N, N is the number of molecules per unit volume) is written in the following way:

$$\lambda_p/N=\pi r_0^2 cZ_{\text{eff}}P(kR), \tag{7}$$

where P(kR) depends only on the parameter indicated in the parentheses and represents the probability of penetration of the positronium atom outside the well:

$$P(kR)=\sin^2 kR/(1-kR\operatorname{ctg}kR). \tag{8}$$

As an upper-limit estimate of the number of electrons in which triplet positronium annihilates (Z_{eff}), we can take the number of valence electrons of the atoms in the molecule.

In order to provide a minimum energy of the liquid-(positronium atom) system, the following condition should be satisfied:[3]

$$\frac{dE}{dR}+\frac{d}{dR}(4\pi R^2\gamma)=0, \tag{9}$$

where γ is the surface tension.

Relations (7), (8), and the boundary conditions provide the possibility of determining the quantities kR, R, and U listed in the table by another independent method from the values of λ_p/N obtained by us.

IV. DISCUSSION OF RESULTS

In the table we have given the values of kR, R, and the potential-well depth U obtained by the time and correlation methods, practically identical values being obtained in the two cases. The agreement of the results permits the conclusion to be drawn that the bubble model apparently describes correctly the behavior of the positronium atom in organic liquids.

As can be seen, the two basic methods of physicochemical investigation of positronium (correlation and time) can be used to determine the characteristics of the potential field effectively acting on the positronium atom in the liquid. The insignificance of the difference in the parameters describing the potential well—the bubble—in different organic liquids is explained by the closeness in the values of surface tension and repulsion potential between the liquid and the positronium atom for these materials.

The applicability of the bubble model to description of the behavior of a positronium atom in the liquids studied indicates that here a case of strong interaction is observed (see section I). However, if the surface tension of the liquid is sufficiently large, the repulsion between the positronium atom and the liquid will not provide formation of a bubble, and the bubble model will become inapplicable.

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