PHYSICAL REVIEW VOLUME 112, NUMBER 6 DECEMBER 15, 1958

Quenching of Positronium Lifetimes by Molecular Iodine*

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(Received August 4, 1958)

Measurements of the mean life of the τ_2 component for decay in solutions of iodine in normal heptane give a value of 10^{-17} cm^2 for the cross section of molecular iodine for annihilating ^3S positronium on collision. This effect is attributed to an enhancement of "pickoff" annihilation resulting from either a tendency to form positronium iodide or the high polarizability of molecular iodine. In quenching τ_2 from 2.49×10^{-9} sec to 1.32×10^{-9} sec the intensity I_2 of the long component was observed to vary over a range from 40% to 27%; however, the errors in I_2 are so large for the shorter lifetimes that it is impossible to tell from the data whether or not I_2 is influenced by the presence of iodine.

INTRODUCTION

TWO basic mechanisms which would cause quenching of the ^3S positronium lifetime in condensed materials have been suggested, "flipping" by spin exchange from the ^3S to the ^1S state[1] and "pick-off" annihilation with an electron bound to a neighboring atom.[2] The formation of positronium compounds has been mentioned as a possibility[3,4]; however, this can be regarded as a type of "pick-off" process.

The quenching of ^3S positronium in gases was studied by measuring three-gamma count rates and lifetimes as a function of gas pressure. Large cross sections, estimated to be around 10^{-16} cm^2, were found for NO, Cl_2, and Br_2. The quenching action of NO was attributed to spin exchange while the formation of positronium compounds was postulated for the halogens.[3]

The cross section of diphenyl picryl hydrazyl (DPH) for quenching the τ_2 component in benzene was determined by the investigation of two-gamma count rates[5] and lifetimes[6] for decay in solutions of DPH in benzene, the latter method yielding a value of $\sigma=1.2\times10^{-17}$ cm^2. Recent angular correlation studies indicate that this quenching is not due to spin exchange as was supposed, but is due instead to enhanced "pick-off"[4] which is probably caused by the formation of positronium compounds. Paramagnetic ions such as Mn^{++} have a much smaller cross section,[7] $\sigma\cong10^{-20}$ cm^2, and apparently lead to genuine spin exchange.[4]

It is the purpose of the present investigation to determine the cross section of iodine for quenching the τ_2 component. The method used consists of measuring τ_2 for various concentrations of iodine in normal heptane. If changes in the lifetime can be attributed entirely to the presence of molecular iodine, then $\lambda_2(N)-\lambda_2(0)=N\sigma v$ and $d\lambda_2(N)/dN=\sigma v$, where λ_2 is the annihilation rate, N the number of iodine molecules/cm^3, σ the cross section, and v the velocity of the positronium atom.

EXPERIMENTAL DETAILS

In measurements of this type, it is desirable that there be no interaction between the solute and solvent. Benzene was rejected for use as a solvent since part of the iodine molecules in solution become loosely attached to benzene molecules. Heptane was chosen because data on the solubility of iodine in heptane were readily available, iodine does not interact with heptane, and heptane has a suitable τ_2 component. The solutions were made up by mixing convenient volumes of saturated iodine in heptane solution and pure heptane.

The sample to be studied was contained in a rectangular tank milled out of cast Lucite. The source of positrons, Na^{22}, was deposited between two thin sheets of mica which were sealed at the edges and fixed to Lucite washers with shellac. This source holder slipped into two slots on the inside of the Lucite tank so that the source was held at the center of the sample. To complete the assembly a top was sealed on with vinylite cement.

The delayed-coincidence apparatus used in these measurements was described earlier[8] and has undergone only slight modification in the meantime. Liquid scintillators were replaced with crystals of diphenyl acetylene and a 3-mμsec fast-coincidence resolving time is now used instead of the former 2 mμsec in order to obtain a higher counting rate. With this arrangement, it takes about four hours to study a sample, i.e., to accumulate two complete sets of data. Evaporation during this time was less than 5%.

The correction for annihilation in the sample holder and calculations of the mean life and intensity of the long component have been discussed in a previous paper[8] and therefore will only be outlined here. The correction factor for annihilation in the mica films was

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* This research was supported in part by the U. S. Air Force Office of Scientific Research of the Air Research and Development Command and in part by the University of Texas Research Institute.

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[1] R. E. Bell and R. L. Graham, Phys. Rev. **90**, 644 (1953).

[2] R. L. Garwin, Phys. Rev. **91**, 157 (1953); M. Dresden, Phys. Rev. **93**, 1413 (1954).

[3] M. Deutsch, Progr. Nuclear Phys. **3**, 151 (1953).

[4] Robert de Zafra, Bull. Am. Phys. Soc. Ser. II, **3**, 229 (1958).

[5] T. A. Pond, Phys. Rev. **93**, 478 (1954).

[6] S. Berko and A. J. Zuchelli, Phys. Rev. **102**, 724 (1956).

[7] S. Berko and F. L. Hereford, Revs. Modern Phys. **28**, 303 (1956).

[8] Hatcher, Millett, and Brown, Phys. Rev. **111**, 12 (1958).

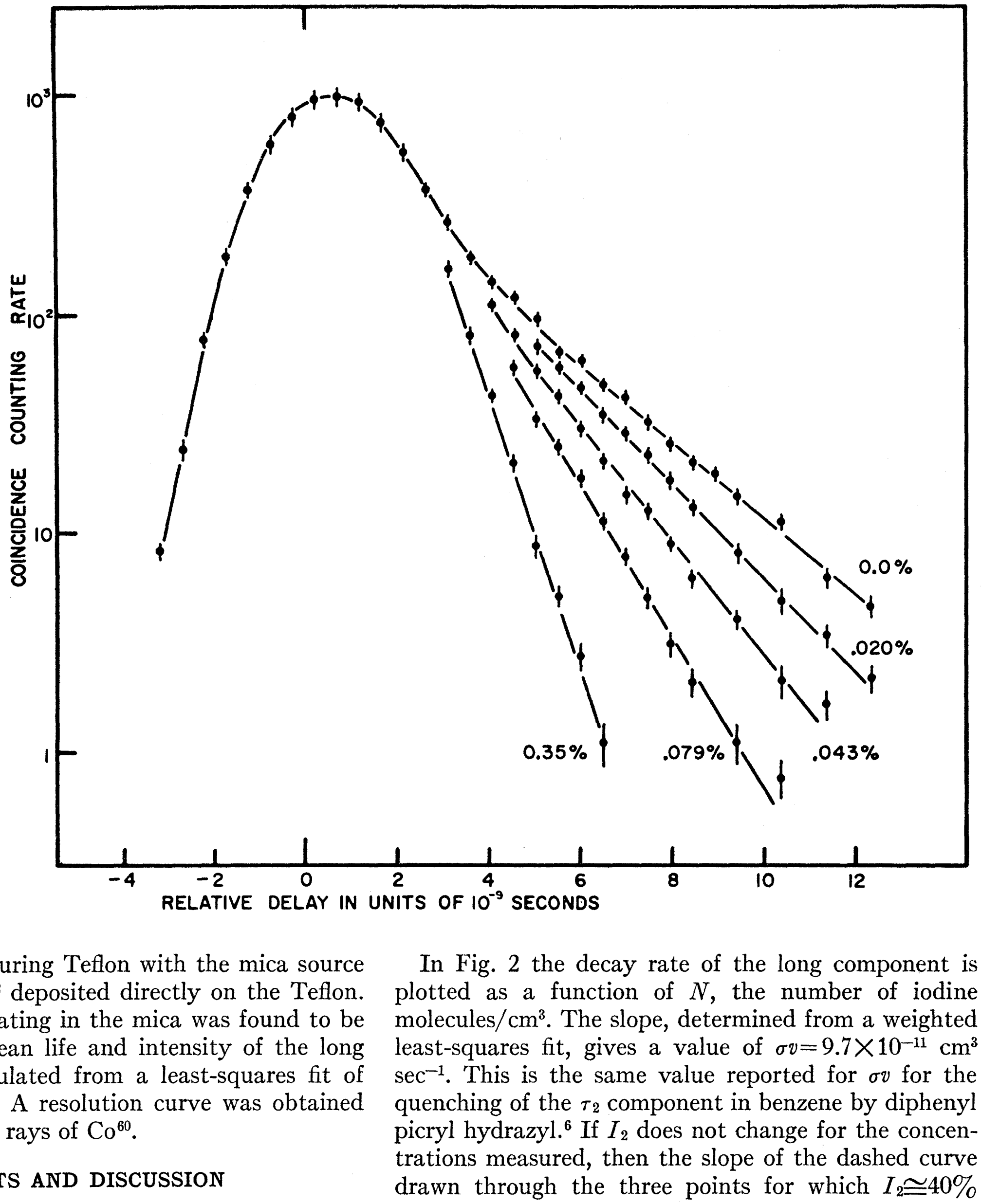

FIG. 1. The complete delayed coincidence curve for normal heptane and exponential tails for solutions of iodine in heptane. The numbers refer to the molecular percent of iodine in solution.

determined by measuring Teflon with the mica source and again with Na^{22} deposited directly on the Teflon. The fraction annihilating in the mica was found to be $(13.4\pm4)\%$. The mean life and intensity of the long component are calculated from a least-squares fit of the exponential tail. A resolution curve was obtained by using the gamma rays of Co^{60}.

RESULTS AND DISCUSSION

The strong dependence of τ_2 on the concentration of iodine can be seen in Fig. 1. The long component appears to be essentially quenched out for 0.35 mole % iodine in heptane solution, and there is a noticeable reduction in τ_2 for only 0.020 mole %, or when there is one iodine molecule in 5000 molecules of solution. The results from an analysis of the delayed coincidence curves are summarized in Table I. It will be seen that the intensity of the long component varies over a wide range of values. There appears to be a slight trend for I_2 to be reduced as N is increased; however, the errors in I_2 are so large for the shorter lifetimes that it is impossible to say for certain whether or not I_2 changes at all. The weighted mean I_2 was found to be 38.1%.

In Fig. 2 the decay rate of the long component is plotted as a function of N, the number of iodine molecules/cm³. The slope, determined from a weighted least-squares fit, gives a value of $\sigma v = 9.7\times10^{-11}$ cm³ sec⁻¹. This is the same value reported for σv for the quenching of the τ_2 component in benzene by diphenyl picryl hydrazyl.[6] If I_2 does not change for the concentrations measured, then the slope of the dashed curve drawn through the three points for which $I_2 \cong 40\%$ $(11.0\times10^{-11}$ cm³ sec⁻¹$)$ should be a better value of σv than the one obtained from the least-squares fit. The probable error is about 10^{-11} cm³ sec⁻¹. The cross

TABLE I. Results for positrons annihilating in solutions of iodine in heptane.

$M\%$[a]	$N(10^{18}\ \text{cm}^{-3})$	$\tau_2(10^{-9}\ \text{sec})$	$\lambda_2(10^{9}\ \text{sec}^{-1})$	$I_2\%$[a]
0.0	0.0	2.49 ± 0.1	0.402 ± 0.016	39.2 ± 3
0.020	0.822	2.03 ± 0.1	0.492 ± 0.024	40.0 ± 4
0.043	1.77	1.75 ± 0.1	0.570 ± 0.033	36.5 ± 5
0.060	2.47	1.63 ± 0.1	0.612 ± 0.038	27.4 ± 5
0.079	3.25	1.32 ± 0.1	0.758 ± 0.057	39.5 ± 9

[a] $M\%$ is the mole % iodine in the solution. $I_2\%$ is the intensity of the τ_2 component.

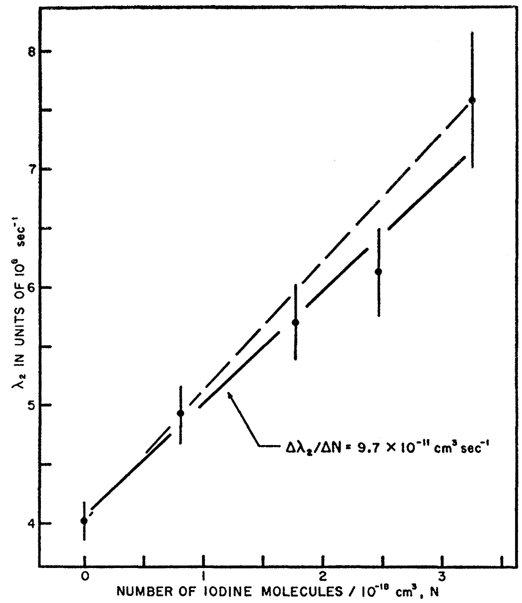

FIG. 2. A least-squares fit of λ_2 *versus* N yields a value of $\sigma v = 9.7 \times 10^{-11}$ cm³ sec⁻¹. The dashed curve is discussed in the text.

section is obtained by assuming that the positronium atoms move with thermal velocities. Taking the same value of v that has been used before,[6] it is found that $\sigma \cong 10^{-17}$ cm².

The forces between positronium and molecular iodine which cause quenching of the ^3S positronium lifetime are thought to arise from either the tendency of a positronium atom and an iodine molecule to form a bound system or the high polarizability of the iodine molecule, or both. More experimental work will probably be necessary before it can be said which of these factors is more important. The fact that NaCl in water alters the lifetime only slightly[9] seems to be evidence in favor of chemical forces between positronium and molecular iodine. The negative chlorine ion should have little inclination to undergo a chemical reaction with positronium since its outside shell is filled. On the other hand, it should be quite polarizable. It is possible that the Cl^- ion could be partially shielded from the positronium due to hydration effects, which would not be the case for Cl_2 or I_2.

A preliminary study carried on in this laboratory of water and a saturated solution of NaI in water indicates only slight differences in τ_2 and I_2. Thus, the present results seem to suggest that the quenching of ^3S positronium by molecular iodine is due primarily to chemical binding, although forces due to the polarizability of iodine cannot be ruled out. Investigation of the quenching of other halogens by both lifetime and angular correlation measurements should prove enlightening.

ACKNOWLEDGMENTS

The authors wish to thank Dr. Eldon Ferguson for helpful discussions and Mr. Jack Rogers for his assistance with the taking and analyzing of the data.

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[9] R. E. Green and R. E. Bell, Can. J. Phys. **35**, 405 (1957).