TEMPERATURE DEPENDENCE OF POSITRON ANNIHILATION

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

Figure Page

CHAPTER I

INTRODUCTORY SURVEY

One of the early achievements of the relativistic quantum theory was Dirac's (23) prediction that electrons could occupy negative as well as positive energy states(24). The positive and negative energy states correspond to what are now known as electrons and positrons respectively. At first the apparent absence of positrons was taken as evidence for the failure of Dirac's electron theory. Two years later however the positron was detected in a cloud chamber experiment (1). In this instance it occurred as a consequence of pair production. Still later it was discovered that unstable nuclei could decay by positron emission (29), thus providing a ready source of positrons for experiments. The mass of the positron was found to be equal to that of the electron and its charge was of equal magnitude but opposite in sign to that of the electron.

The Annihilation Process

Just as the process for pair production creates an electron β^- and a positron β^+ when a gamma ray of energy greater than 1.02 ($2m_0c^2$) Mev passes through the intense electric field of a nucleus,

1) $$\gamma \rightarrow \beta^+ + \beta^-$$

so also may the reverse reaction occur when an electron and a positron annihilate with one another.

$$\beta^{+} + \beta^{-} \rightarrow n\gamma \qquad (2)$$

Here "n" is an integer which theoretically can have values 1, 2, or 3 depending upon the environment and state of the electron relative to the positron at the moment of annihilation. The theory for positron annihilation will be presented in Chapter II.

Scope of the Present Research

Due to the apparent environmental effect upon the annihilation process, equation (2), it seemed reasonable that a further study of positron annihilation in matter would yield useful information as to the properties of matter. Such has been the case in experiments on the angular correlation of the annihilation gammas for the case where $n = 2$. The results of these experiments show that the momentum distribution of the conduction electrons in metals is in agreement with that predicted by theory (15, 42, 21, 9).

Another approach to the study of positron annihilation in matter which appears to be most promising is that of its mean lives, especially the τ_2 component which will be discussed in Chapter III along with a review of what has been done to the present with regard to lifetime measurements. Although experiments indicate that there is a definite dependence of the τ_2 component upon the parameters of the surrounding material, at present there is no satisfactory theory as to how these parameters influence the lifetime. The most promising explanation offered thus far has been that by Bell and Graham (5) who have proposed the order-disorder effect.

The order-disorder effect refers to the rearrangement of the molecular or crystalline structure of a material as it passes from say the solid phase to the liquid phase when the temperature is raised. In general the structure of the material continues to become more orderly as the temperature is further decreased for a given phase. However, it is the transitions of gas to liquid or liquid to solid at which the most radical order-disorder changes occur.

In pursuance of this suggestion, the author and his colleagues have performed more extensive experiments on a wide variety of materials in order to determine more accurately the behavior of the τ_2 component as a function of temperature and phase state. A thorough discussion of this work is given in Chapter IV. Finally, in Chapter V the results of our investigations, further remarks and suggestions are given.

CHAPTER II

POSITRONIUM THEORY

There are essentially two ways in which the positron-electron annihilation may occur. One is by direct collision and the other is by the formation of a quasi-stable system prior to annihilation. The possibility of the formation of such a quasi-stable system was first proposed by Mohorovicic (34) in 1934 and later Ruark (39) introduced the name positronium.

The structure of positronium is very much like that of the hydrogen atom. So much so in fact, a simple substitution of the reduced mass of positronium for that of hydrogen is all that is necessary in order to determine the positronium spectral series and wave function from that of hydrogen. The reduced mass of a two particle system is given as

3) $$\mu = \frac{Mm}{M+m}$$

Since the mass of the proton M in hydrogen is much larger than that of the electron m, (3) may be approximated by

4) $$\mu \simeq m$$

For positronium M = m so that the reduced mass becomes

5) $$\mu = \frac{m}{2}$$

which is seen to be one half as large as that for hydrogen. The Bohr radius a_0 for hydrogen is given as

6) $$a_0 = \frac{\hbar^2}{\mu e^2}$$

$2\pi\hbar$ -- Planck's constant
e -- electronic charge

so that it follows from equation (5) that the Bohr radius for positronium is twice that of hydrogen.

With this we may now transform the hydrogen atom wave function

7a) $$\Psi_{k,l,m} = Y_{l,m}(\theta,\phi) R_{k,l}(r)$$

into that for positronium. Here $Y_{l,m}(\theta,\phi)$ are normalized spherical harmonics and $R_{k,l}(r)$ are normalized Laguerre functions, k is the principal quantum number, l is the orbital angular momentum quantum number and m is the Z-component of the angular momentum quantum number l and r is the radial coordinate. For the ground state k = 1, l = 0, m = 0. This is the singlet state $^1S_0$. For this case equation (7) becomes

7b) $$\Psi_{1,0,0} = \frac{1}{2\sqrt{\pi}}\left(\frac{1}{a_0}\right)^{3/2} \cdot 2\exp\left(-\frac{r}{a_0}\right)$$

Substitution of the positronium Bohr radius $2a_0$ in place of the hydrogen atom Bohr radius gives the wave function for positronium.

7c) $$\Psi_{1,0,0} = \frac{1}{2\sqrt{\pi}}\left(\frac{1}{2a_0}\right) \cdot 2\exp\left(-\frac{r}{2a_0}\right)$$

The wave lengths for hydrogen spectral terms are given as

7d) $$\lambda = \frac{c\hbar^3}{2\pi^2 e^4}\, \mu^{-1}\left(\frac{1}{n_f^2} - \frac{1}{n_i^2}\right)^{-1}$$

so that it is obvious that the corresponding spectral terms for positronium will be twice as long as those of hydrogen. The fine structure of positronium involves the spin of both particles and has been worked out by Pierenne (38) and Berestetski (7). The splitting of the singlet and triplet levels is 8.45×10^{-4} ev. and the binding energy for the ground state is 6.77 ev.

Annihilation of the Positron in Flight

Obviously a determination of state of the positron-electron pair prior to annihilation is of prime importance in developing a theory that will adequately explain the annihilation process. If the majority of the positrons annihilate before coming to rest, the collision cross section for the positron will be the important parameter to be considered. However if the positron slows to thermal velocities in a time several orders of magnitude smaller than the observed life time, then the time of flight may be ignored and other factors such as electron density and molecular structure will have to be considered in the mechanisms explaining positron annihilation.

Lee-Whiting (30), as well as Beg and Stehle (4), has treated this problem for the case of metals in the following manner. He assumed the conduction electrons to be a free electron gas at 0^{o}K and took the interaction between the electron and positron the screened Coulomb potential $e^2/[r_{1,2} \exp(q r_{1,2})]$ where q is the screening parameter, e is the electronic charge, and $r_{1,2}$ is the distance between the pair.

The number of collisions experienced per second by the positron is $t^{-1}P(t)$ where P(t) is the probability that the positron has changed its state from $\bar{K}_1$ to $\bar{K}_1'$ due to a collision with an electron in state $\bar{K}_2$ prior to collision and state $\bar{K}_2'$ after collision. A time dependent perturbation calculation gives the probability of a change of state as

$$8) \quad P(t) = \frac{1}{\pi^4} \frac{e^2 t^2}{\hbar^2} \int \frac{d(\bar{K}_1 - \bar{K}_1')}{(|\bar{K}_1 - \bar{K}_1'|^2 + q^2)^2} \int \frac{1 - \cos x}{x^2} \, dK_2'$$

where

$$9) \quad x = \frac{t\hbar}{2m} \left[(K_1'^2 + K_2'^2) - (K_1^2 + K_2^2) \right]$$

and $\bar{K}_1$ and $\bar{K}_2$ are wave vectors for positron and electron respectively. The rate of energy loss R(E) is found by multiplying the integrand of $t^{-1}P(t)$ by

$$10) \quad E(K_1) = \frac{\hbar^2}{2m} (K_1^2 - K_1'^2)$$

which is the energy loss per collision.

Therefore

$$11)\quad R(E) = \frac{e^2 t}{2\pi^4 m} \int \frac{(K_1^2 - K_1'^2)\, d(\bar{K}_1 - \bar{K}_1')}{(|\bar{K}_1 - \bar{K}_1'|^2 + q^2)^2} \,.$$

Upon integration

$$12)\quad R(E) = \frac{m e^4}{3\pi \hbar^3} E F\left(\frac{K_1}{q}\right)$$

where

$$13)\quad F\left(\frac{K_1}{q}\right) = \frac{32}{35}\left(\frac{K_1}{q}\right)^4 \left[1 - \frac{20}{9}\left(\frac{K_1}{q}\right)^2 + \cdots\right] .$$

Equation (13) is valid for the energy of the positron $E \leqslant E_f$, E_f being the Fermi energy of the free electron gas. By approximate integration of equation 12 it will be found that the positron loses its energy exponentially with time and will thermalize on the order of 10^{-12} sec. Since the observed lifetime is about 10^{-10} sec., most of the positrons will be expected to thermalize before annihilation.

Elsewhere, Bethe (10) has shown that the total probability of two quantum annihilation while in flight is

$$14)\quad S(\epsilon_0) = F(\epsilon_0) / [4 \log(mc^2/RZ)]$$

where (15) $$F(\epsilon_0) \doteq \int_0^{E_0} \frac{\sqrt{\epsilon^2 - 1}}{\epsilon^2(\epsilon + 1)} \left[\frac{\epsilon^2 + 4\epsilon + 1}{\sqrt{\epsilon^2 - 1}} \log(\epsilon + \sqrt{\epsilon^2 - 1}) - (\epsilon + 3)\right] d\epsilon$$

and 16) $$\epsilon_{(0)} = \frac{E_{(0)}}{E_{(0)} + m_0 c^2}$$

The terms in the last two equations are defined as

R– ionization potential of hydrogen

Z– atomic number of the stopping material

m – positron mass

C – velocity of light

E – positron kinetic energy

E_0 – initial kinetic energy of positron

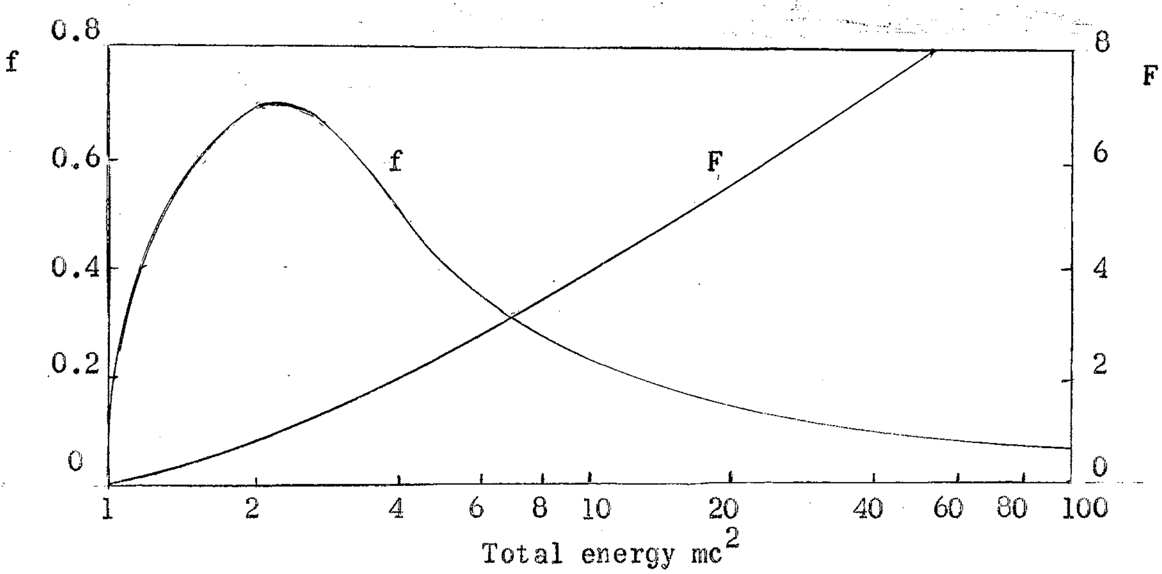


Figure 1. $F(\epsilon)$ and $f(\epsilon)$, the integrand of (15), are plotted as a function of the positron energy. The $F(\epsilon)$ curve gives the total two gamma annihilation probability and the $f(\epsilon)$ curve gives the differential annihilation probabilities. From Bethe (10)

The value of the log term in in (14) for lead, air, and hydrogen is 6.1, 8.6, and 10.5 respectively. For a positron of energy 0.54 Mev, we find from Figure 1 that $F(\epsilon_0) = 0.8$. From these values the total probability of a positron in flight annihilating into two quanta is given in Table I.

TABLE I

THE TOTAL PROBABILITY $S(\epsilon_0)$ OF A POSITRON ANNIHILATING IN FLIGHT FOR LEAD, AIR, AND HYDROGEN

Probability in %	Material
3.3%	lead
2.0%	air
2.8%	hydrogen

In order for a one gamma annihilation to occur, the positron must annihilate with an inner shell electron. Bethe (10) has calculated the ratio of the probability of one gamma to two gamma annihilations to be $\left(\frac{Z}{137}\right)^4 \chi(\epsilon)$ for annihilation with K shell electrons.

where

$$17)\quad \chi(\epsilon) = 4\sqrt{\frac{\epsilon-1}{\epsilon+1}}\ \frac{(\epsilon^2+2/3\epsilon+4/3) - \frac{\epsilon^2+2}{\sqrt{\epsilon^2-1}}\log(\epsilon+\sqrt{\epsilon^2-1})}{(\epsilon^2+4\epsilon+1)\log(\epsilon+\sqrt{\epsilon^2-1}) - (\epsilon+3)\sqrt{\epsilon^2-1}}$$

In Figure 2, $\chi(\epsilon)$ is plotted as a function of ϵ.

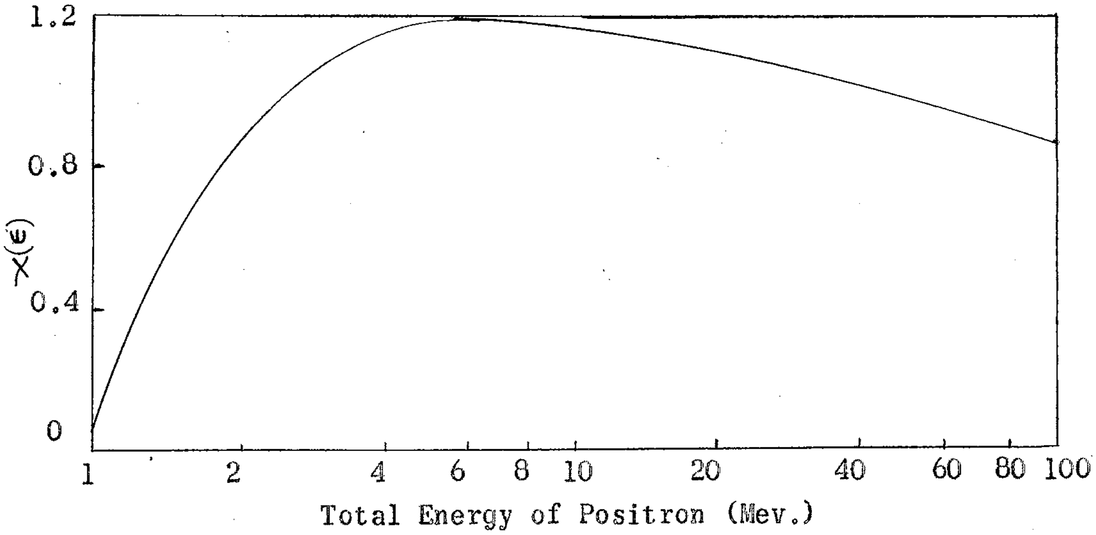


Figure 2. The ratio of one to two quantum annihilation as given by (17). From Bethe (10)

From Figure 2 it is seen that only at high energies does the one gamma process become significant, relative to the 2 γ process. At the other extreme, thermalized positrons have nearly a zero probability for one quantum annihilation as compared with the two quantum process. As an example, consider the case of a 0.54 Mev positron passing through lead, Z = 82. From Figure 2 it is found that $\chi(\epsilon)$ = 0.7 and we have for the ratio

$$18)\quad R = \left(\frac{82}{137}\right)^4 \cdot 0.7 \approx 0.09$$

For L, M, etc, shells the probability is even less being proportional to k^{-3}, where k is the principal quantum number. Screening will tend to decrease the probability still further.

For the case of three quantum annihilation it will be shown later that the probability of three quantum decay is smaller than the two quantum process by a factor of about 370.

It is now evident that for the energy range of the positrons less than 0.54 Mev, annihilation in flight will be of minor importance. This now leaves only the other possibility that there exists a quasi-stable state prior to annihilation. Unless this were true certain phenomena such as the relative intensity of annihilation radiation could not be adequately explained as it is now assuming the formation of positronium.

Selection Rules for Positronium Decay

With the fact established that many of the positrons decay from a quasi-stable system, we now proceed to investigate the selection rules governing positronium decay. If positronium forms in an excited $l \neq 0$ state, it will make an immediate transition to the ground state and eventually annihilate. Since the electron-positron wave function does not overlap sufficiently for annihilation to occur when in excited states, only the ground state of positronium will be considered. In the ground state there exists two substates: 1) the singlet state $^1S_0$ known as parapositronium wherein the spins of the two particles are antiparallel and 2) the triplet state $^3S_1$ known as orthopositronium and it has parallel electron- positron spins.

The selection rules for $^1S_0$ and $^3S_1$ decay may be developed from the conservation principles of parity and angular momentum, or the principle of invariance under charge conjugation (43), (31). In this section the selection rules will be worked out in detail using the method of charge conjugation.

Invariance under charge conjugation requires that there be a complete symmetry between the two charge conjugate states of particles whose states are described by complex wave functions. The act of reversing the signs of the charges within a system and all else remaining the same is defined as charge conjugation and will be represented by the operator C. This operator commutes with the energy H, momentum L, total angular momentum J, and parity P. This means that C may be diagonalized with any of these operators. Consequentially, if a state of positronium is specified with respect to J, P, L, and H, the eigenvalues of C may be determined for that state.

Expanding the state function of any state of positronium in terms of free particle state functions and operating upon them with C we have

$$19) \quad C\Psi_{P_s} = C\Big(\sum_{p,s_1,s_2} c[\bar{p}, s_1, s_2]\, a^*_{s_1}(\bar{p})\, b^*_{s_2}(-\bar{p}) + \sum_{r} \cdots + a^*_{r_1} a^*_{r_2} b^*_{r_3} b^*_{r_4} + \cdots \Big)\, \Psi_{vac}$$

where the second summation represents the effect of virtual pair production, $\bar{p}$ is the relative momentum vector, s_1 and s_2 are the spins of the electron and positron respectively, a^*_{r1} a^*_{s1} b^*_{r2} b^*_{s1} a^*_{r2}, etc. are creation and annihilation operators, and Ψvac is the state function for the vacuum. Since C commutes with the number of free particles, it is sufficient to determine the behavior of the first term with respect to C. Performing the operation we have

$$20a) \quad C\Psi_{P_s} = \sum_{p,s_1,s_2} c(\bar{p}, s_1, s_2)\, C a^*_{s_1}(\bar{p})\, C^{-1} C\, b^*_{s_2}(-\bar{p})\, C^{-1} C\, \Psi_{vac}$$

20b) $C\Psi_{P_s} = -\sum_{p,s_1,s_2} c(-\bar{p}, s_2, s_1)\, a^*_{s_1}(\bar{p})\, b^*_{s_2}(-\bar{p})\, \Psi_{vac}$

For the ground state the function $c(\bar{p}, s_1\ s_2)$ is an even function in $\bar{p}$. Thus (20) may be written

21) $C\Psi_{P_s} = -\sum_{p,s_1,s_2} c(\bar{p}, s_2, s_1)\, a^*_{s_1}(\bar{p})\, b^*_{s_2}(-\bar{p})\, \Psi_{vac}$

As for the spin terms s_1 and s_2, in the triplet state $s_1 = s_2$ and for the singlet state $s_1 = -s_2$. Consequently the first quantity under the summation sign behaves as $c(\bar{p}, s_1, s_2) = c(\bar{p}, s_2, s_1)$ for the triplet state and as $c(\bar{p}, s_1, s_2) = -c(\bar{p}, s_2, s_1)$ for the singlet state. Making these substitutions into (21) we find the eigenvalues of C for the ${}^1S_0$ and ${}^3S_1$ states to be + 1 and -1 respectively.

Next the behavior of a system of photons under charge conjugation must be established. The state function for a single photon is A^{μ} where A^{μ} is the electromagnetic field function. The total Hamiltonian for the photon is $A^{\mu} j_{\mu}$ where j_{μ} is the current. Since C commutes with the Hamiltonian, we may write

22)
$$C A^{\mu} j_{\mu} = A^{\mu} j_{\mu} C$$
$$C A^{\mu} j_{\mu} C^{-1} = A^{\mu} j_{\mu} C C^{-1}$$
$$C A^{\mu} j_{\mu} C^{-1} = A^{\mu} j_{\mu}$$
$$C A^{\mu} C C^{-1} j_{\mu} C^{-1} = A^{\mu} j_{\mu}$$

The transformation of j_{μ} under charge conjugation has the effect of changing the current since the field is reversed.

23) $C j_{\mu} C^{-1} = -j_{\mu}$

Substituting (23) into (22) yields

24) $-C A^{\mu} C^{-1} j_{\mu} = A^{\mu} j_{\mu}$

Dividing both sides by $-j_\mu$ we have

25) $C A^\mu C^{-1} = -A^\mu$

Operating upon the photon state function with C gives

26) $C A^\mu \Psi_{vac} = C A^\mu C^{-1} C \Psi_{vac}$

By definition

27) $C \Psi_{vac} = \Psi_{vac}$

so that a substitution of (25) and (27) into (26) gives

28) $C A^\mu \Psi_{vac} = -A^\mu \Psi_{vac}$

This shows the eigenvalue of C for a single photon to be -1. In general the eigenvalue for a group of n photons is $(-1)^n$.

Now the principle of invariance under charge conjugation may be expressed as follows: The eigenvalue of a system of annihilation photons under charge conjugation operation must be equal to the eigenvalue of the positronium system under charge conjugation which gave rise to the annihilation photons. Since the eigenvalue of parapositronium is + 1, and the eigenvalue of only an even number of photons is + 1, it follows that the singlet state must decay by 2n photons where n = 1, 2, 3, etc. The probability of n being greater than 1 is negligible. Likewise for orthopositronium it is seen that the consistency of eigenvalues requires that $^3S_1$ decay by three photons.

Two Photon and Three Photon Annihilation Rates

In the lowest approximation the annihilation of a free electron-positron pair is a second order process. The rate of positronium annihilation is given as

29) $R = |\Psi(r=0)|^2 \sigma v_+$.

where $|\Psi(r=0)|^2$ is the electron density at the positron, σ is the cross section for annihilation and v_+ is the velocity of the positron. Dirac (23) has calculated the cross section for the two photon case as

30) $$\sigma_{2\gamma} = \frac{\pi e^4}{m^2 c^3} \cdot f(\epsilon)$$

where $f(\epsilon)$ is the integrand of (15) and is plotted as a function of ϵ in Figure 1. In the limit as $E << m_0c^2$ (30) becomes

31) $$\sigma_{2\gamma} = \frac{\pi e^4}{m^2 c^3 v_+}$$

The electron density at the positron for the ground state is readily found by evaluating the square of the wave function, equation ~~(19)~~ (25), at $r = 0$.

32) $$|\Psi(r=0)|^2 = \frac{1}{\pi}\left(\frac{m e^2}{2\hbar^2}\right)^3$$

Substituting (31) and (32) into (29) we have for the rate of two gamma annihilation

33) $$R(^1S_0) = \frac{4\pi e^4}{m^2 c^3 v_+} \cdot \frac{m^3 e^6 v_+}{8\pi \hbar^6} \ \mathrm{sec}^{-1} \approx 8 \times 10^{-9}\ \mathrm{sec}^{-1}.$$

Here the factor four is necessary since the cross section given in (31) is averaged over the four possible spin states of the two particle system. The rate of three gamma annihilation is not quite so simple as for the two gamma case. Here annihilation takes place via two intermediate states. Ore and Powell (37) used the time-dependent perturbation theory to determine the rate of positronium annihilation into three photons. For this they get

34) $$R(^3S_1) = \left(\frac{16}{9\pi}\right)(\pi^2 - 9)\left(\frac{e^6}{2 a_0 m^2}\right) \ \mathrm{sec}^{-1} \approx 7.1 \times 10^6\ \mathrm{sec}^{-1}.$$

The reciprocal of the rates of decay gives the lifetimes τ_1 and τ_3 for the singlet and triplet states respectively.

35) $$\tau_1 \simeq 1.25 \times 10^{-10} \text{ sec.}$$
$$\tau_3 \simeq 1.4 \times 10^{-7} \text{ sec.}$$

The relative cross section for the triplet state annihilation may now be shown to be smaller than that for the singlet by a factor of approximately 370. Multiplying the ratio of τ_1 to τ_3 by the statistical factor 3 gives

36) $$\frac{\sigma_{3\gamma}}{\sigma_{2\gamma}} = 3\left(\frac{\tau_1}{\tau_3}\right) \simeq \frac{1}{370} .$$

The factor of 3 is necessary in order to compensate for the three substates of $^3S_1$ from which it might decay. This means that the two gamma annihilation is 1115 times faster than the three gamma mode of decay. The theoretical value of τ_1 is in excellent agreement with the experimental values. Although the experimental values vary from material to material, they are of the same order of magnitude as predicted by theory. For example, Bell and Graham (5) have found the mean lifetime for the τ_1 component in metals to be 1.5 x 10^{-10} sec. As for the ratio $\frac{\sigma_{2\gamma}}{\sigma_{3\gamma}}$, Basson (2) has reported the value 406 ± 50 for annihilations in metal.

As mentioned earlier there also exists the τ_2 lifetime which is due to an environmental effect known as "pick-off." This occurs when the positron in orthopositronium annihilates with an bound electron of opposite spin. The effect of this is to shorten the τ_3 lifetime of the trip-state by a factor of about 10^{-2}. More will be said about pick-off in the following chapter where the environmental effects will be considered.

CHAPTER III

POSITRONIUM ANNIHILATION IN MATTER

Much experimental work has been done with regard to studying the mean life of positronium in condensed materials and gases. Thus far the effects of temperature, pressure, electric fields, magnetic fields and impurities in aqueous solutions have been examined in order to determine the parameters that affect the mean life. Several surveys of these experiments have been made from time to time. Ferrell (25) has written a quite comprehensive review of the theory of positronium annihilation in solids. Berko and Hereford (8) in an experimental review have also included liquids. Earlier reviews by Deutsch (20), and Corben and DeBenedetti (13) establish the existence and properties of positronium in gases. It is the purpose of this chapter to present a current review which places emphasis upon the temperature dependence of positronium annihilation in matter.

The Environmental Effects Upon Positronium Annihilation

The environmental effects upon τ_2 and τ_3 lifetimes are much more pronounced than they are upon the τ_1 lifetime. It appears as if the τ_1 lifetime is completely insensitive to the lattice characteristics. Some materials such as metals, ionic crystals, and other crystalline materials appear not to give rise to the τ_2 lifetime. In general it is the amorphous and molecular materials which possess the τ_2 component. It was first found by Bell and Graham (5) (see Table V) that in those materials which did possess the τ_2 component, the τ_2 lifetime was proportional to the absolute temperature. Since the τ_2 lifetime is attributed to the pick-off of the positron in the orthopositronium by a bound outer shell electron of the opposite spin, it follows that for those materials exhibiting a τ_2 component, the τ_3 component will be directly affected. In Table II it is seen that for those materials having the τ_2 lifetime there is a decided increase in the rate of τ_3 decay. This may be understood in the following manner. Since the total annihilations must remain constant, a decrease in the two gamma decay rate will result in an increase in the three gamma decay rate and vice versa. The ratio of two to three gamma annihilations for direct collisions with free electrons has been theoretically determined to be 372. This is because the $^1S_0$ state decays 1115

TABLE II

OBSERVED THREE QUANTUM DECAY RATES IN CONDENSED MATERIALS FROM BERKO AND HEREFORD (8)

Material	Three Quantum at 20°C (counts/min.)	Positron mean lives (10^{-9} sec)	
		τ_1	τ_2
Beryllium	0.14 ± 0.05	0.17 ± 0.5	---
Aluminum	0.20 ± 0.05	0.15 ± 0.3	---
Crystalline quartz	0.22 ± 0.05	0.20 ± 0.3	---
Fused quartz	0.41 ± 0.10	0.35 ± 0.5	1.8 ± 0.2
Polystyrene	0.47 ± 0.12	0.37 ± 0.5	2.3 ± 0.2
Teflon	0.68 ± 0.10	Not quoted	3.5 ± 0.4

times faster than the $^3S_1$ state but the probability of formation of the $^3S_1$ state is three times greater. Dividing the rate of $^1S_0$ decay by the relative probability of $^3S_1$ formation yields the ratio of approximately 372/1. Basson (2) has experimentally determined a value of 406 ± 50 for the $2\gamma/3\gamma$ ratio in the case of metals. This agrees within the limits of experimental error with the theoretical value of 372, and is to be expected because of the small amount of positronium formation in metals.

On the other hand if a large amount of positronium is formed as is the case of positron annihilation in gases, the $2\gamma/3\gamma$ ratio reduces to approximately 3 (17). This requires that there be a strong decrease in the 2γ rate of decay and a correspondingly strong increase in the 3γ rate of decay. For materials which form positronium best this is just the case for once positronium is formed, the $^3S_1$ state must decay by three photons, except for a fraction which, due to pick-off, annililates by two photons. Since there is three times as much orthopositronium formed as parapositronium and the fast decay time of parapositronium is not relevant in this case, a theoretical minimum ratio is 1/3. Since all positrons do not annihilate from positronium states and not all orthopositronium decays by three photons, it is expected that the ratio $2\gamma/3\gamma$ will be larger than 1/3. The relation between the τ_2 lifetime and the increase of the τ_3 rate is that materials having a τ_2 lifetime are more effective in forming positronium than those which do not have a τ_2 lifetime.

The method for determining the mean lives of positronium from experimental data are varied (3, 35). The general techniques are discussed in the following paragraph where it is assumed that Na^{22} is the source of positrons. The same procedure will apply to any other acceptable positron source. An acceptable positron source is one that decays by positron emission, predominately to a single excited state of the daughter element which emits a gamma ray detectably different from 0.51 Mev within 10^{-11} sec. after the positron emission.

When Na^{22} decays by positron emission, a gamma ray of approximately 1.28 Mev is emitted, which in our case may be considered simultaneous. This is called the prompt gamma and is used to indicate the time of positron formation. When the positronium decays, the annihilation gammas give the signal for the end of the positron's life time. Since the time of positron flight is negligible as compared to the positronium mean life, as was shown in Chapter II, the lapse of time (referred to as the "delay time") between the occurrence of the prompt gamma and the annihilation gammas may in effect be considered to be a measure of the positronium lifetime. By plotting the delay time vs. the count rate, a curve such as that of Figure 3 will result for materials having a τ_2 lifetime. The P(x) curve in Figure 3 is the prompt curve determined for the case where the sample is removed from around the positron source or it is determined from a material which does not possess a τ_1 lifetime. The centroid of the P(x) curve minus 0.19 nanoseconds is the reference time and the shift of the centroid of the τ_1 part of the coincidence curve gives the τ_1 lifetime and the slope of the tail on the right of the curve gives the τ_2 mean life. The areas under the curves are proportional to the intensity of the corresponding decay modes.

Review of Positronium Annihilation

The first reported work performed on the lifetime measurements of positronium was by Shearer and Deutsch (17, 18, 19, 40). As a result of their experiments on positronium annihilation in gases, the formation of positronium was established. The mean life of orthopositronium was found

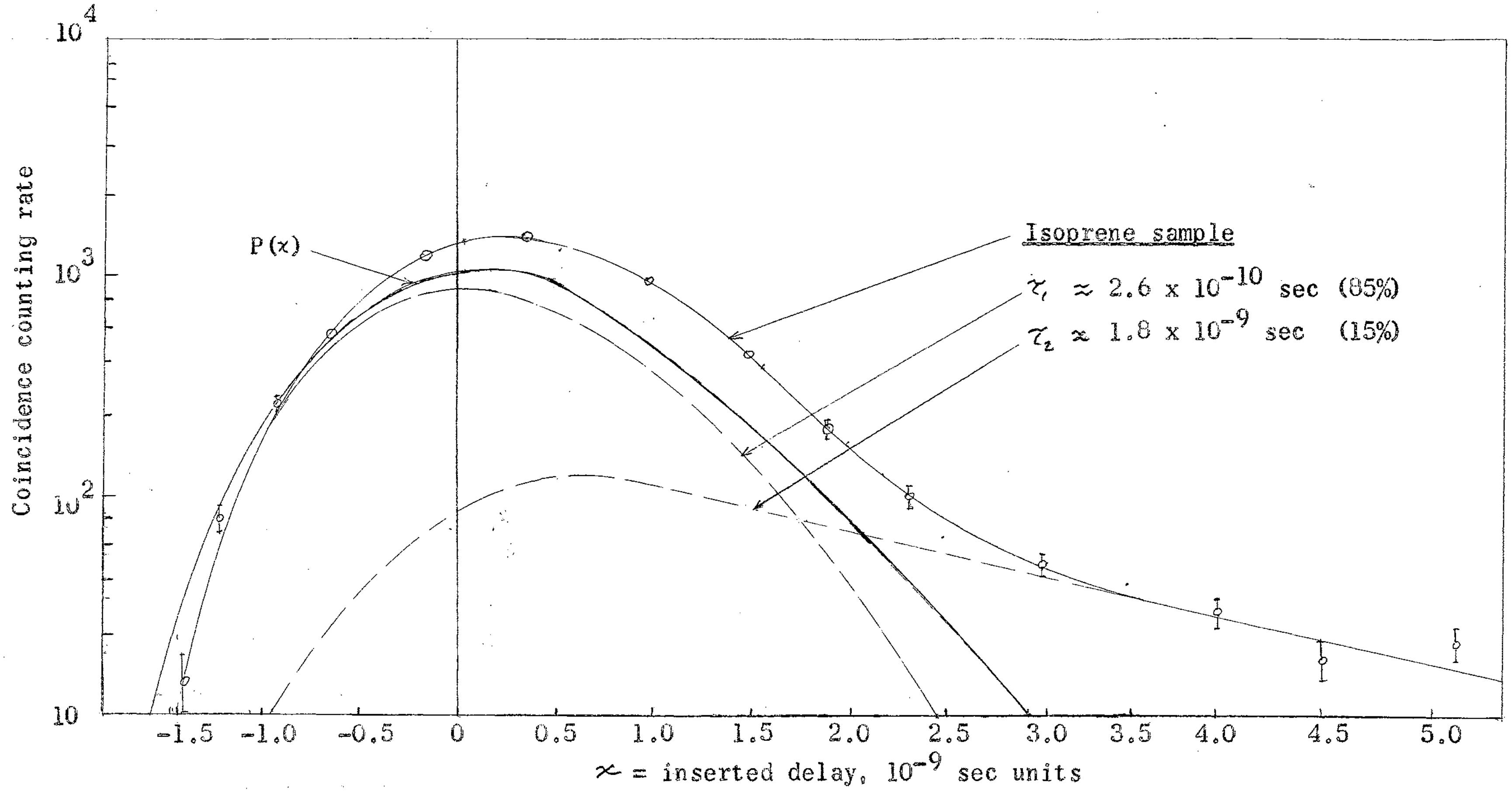


Figure 3. Coincidence resolution curve obtained with a sample of isoprene with an aluminum curve P (x) for comparison. The standard deviations are shown by vertical bars.

to be inversely proportional to the gas pressure and was of the orderof 10^{-7} sec. With the addition of 5% NO to N_2, it was found that the rate of three gamma annihilation was reduced by a factor of three. This effect was interpreted as due to electron exchange. See Table III for their results of orthopositronium lifetime as a function of pressure.

TABLE III

THE EFFECT OF PRESSURE UPON τ_3 AS DETERMINED BY SHEARER AND DEUTSCH (40)

Gas	Pressure (atm.)	τ_3 (10^{-7} sec.)
Nitrogen	0.5	2.6 ± 0.3
	1.0	1.5 ± 0.15
	2.0	1.0 ± 0.15
Argon	0.5	2.9 ± 0.6
	1.0	1.6 ± 0.3
	1.3	1.3 ± 0.2

The work of Shearer and Deutsch was soon followed by a number of experiments by others in order to determine the lifetimes of positronium in solids. Moore (33) found the value of $(1.0 \pm 0.5) \times 10^{-9}$ sec. for the τ_2 lifetime in stilbene and Millett (32) found a value of 1.4×10^{-10} sec. for plexiglass. Experiments performed by DeBenedetti and Richings (14) using metal samples gave much added support to the theory of positronium formation in matter. In their experiments with a group of metals, it was found that the lifetime was constant within the limits of experimental error. See Table IV for their measurements of τ_2 in metals.

TABLE IV

DIFFERENCE IN THE POSITRON HALF-LIFE IN THE METAL INDICATED AND IN ALUMINUM AS DETERMINED BY DeBENEDETTI AND RICHINGS (14)

Material	Valence	Difference in τ_2 (10^{-10} sec.)
Li	1	0 ± 0.5
Na	1	0 ± 0.5
K	1	0 ± 0.5
Ag	1	0 ± 0.5
Pb	4	0 ± 0.5
Cu	2	0 ± 0.5

These results are different from what one would expect if positrons annihilated in free collisions with valence electrons. If free collisions with valence electrons were the case, then the rate of positron annihilation would vary widely for the metals listed in Table IV. Since the lifetimes are measurable the same, the theory of poistronium formation is substantiated.

The first extensive lifetime measurements were made by Bell and Graham (5). The values for τ_1 and τ_2 for the materials observed are given in Table V. The most apparent conclusion of their measurements is that crystalline materials do not have a detectable τ_2 lifetime and amorphous materials do have the τ_2 lifetime. Another obvious factor is the

TABLE V

τ_1 AND τ_2 MEAN LIVES AND RELATIVE INTENSITIES OF POSITRONIUM DECAY IN CONDENSED MATERIALS AS DETERMINED BY BELL AND GRAHAM (5)

Material	°C	τ_1 (sec.)	%	τ_2 (sec.)	%
Li	20	$(1.5 \pm 0.6) \times 10^{-10}$	100		
Be	20	$(1.7 \pm 0.5) \times 10^{-10}$	100		
B "amorphous"	20	$(1.7 \pm 0.4) \times 10^{-10}$	100		
C "amorphous"	20	$(4.3 \pm 0.4) \times 10^{-10}$	100		
C graphite	20	$(3.0 \pm 0.3) \times 10^{-10}$	100		
C diamond	20	$(1.3 \pm 0.3) \times 10^{-10}$	100		
Na	20	$(1.5 \pm 0.6) \times 10^{-10}$	100		
Al	20	$(1.5 \pm 0.3) \times 10^{-10}$	100		
S monoclinic crystal	20	$(2.1 \pm 0.3) \times 10^{-10}$	100		
S "plastic"	20	$(1.5 \pm 0.4) \times 10^{-10}$	~70	$(4.5 \pm 0.6) \times 10^{-10}$	~30
K	20	$(1.5 \pm 0.6) \times 10^{-10}$	100		
Cu	20	$(1.2 \pm 0.5) \times 10^{-10}$	100		
Ag	20	$(1.5 \pm 0.6) \times 10^{-10}$	100		
Au	20	$(1.2 \pm 0.3) \times 10^{-10}$	100		
Hg	20	$(1.2 \pm 0.5) \times 10^{-10}$	100		

Table V (continued)

Material	°C	τ_1 (sec.)	%	τ_2 (sec.)	%
Pb	20	(1.5 ± 0.6) x 10^{-10}	100		
water	20			(1.7 ± 0.2) x 10^{-9}	~ 30
ice	-7			(1.2 ± 0.2) x 10^{-9}	~ 30
ice	-196			(0.85 ± 0.15 x 10^{-9}	~ 30
brine (NaCl)	20			(1.5 ± 0.2) x 10^{-9}	~ 30
NaCl crystal	20	(2.3 ± 0.3) x 10^{-10}	100		
quartz, α -crystal	20	(2.0 ± 0.3) x 10^{-10}	100		
quartz, fused	20	(3.5 ± 0.5) x 10^{-10}	71 ± 5	(1.8 ± 0.2) x 10^{-9}	29 ± 5
Polystyrene	20	(3.7 ± 0.5) x 10^{-10}	64 ± 5	(2.3 ± 0.2) x 10^{-9}	36 ± 5
Polystyrene	-196			(1.7 ± 0.3) x 10^{-9}	~ 30
cellulose nitrate	20			(2.3 ± 0.4) x 10^{-9}	~ 30
Teflon	20			(3.5 ± 0.4) x 10^{-9}	~ 30
Teflon	-78			(2.5 ± 0.4) x 10^{-9}	~ 30
teflon	-196			(1.6 ± 0.4) x 10^{-9}	~ 30

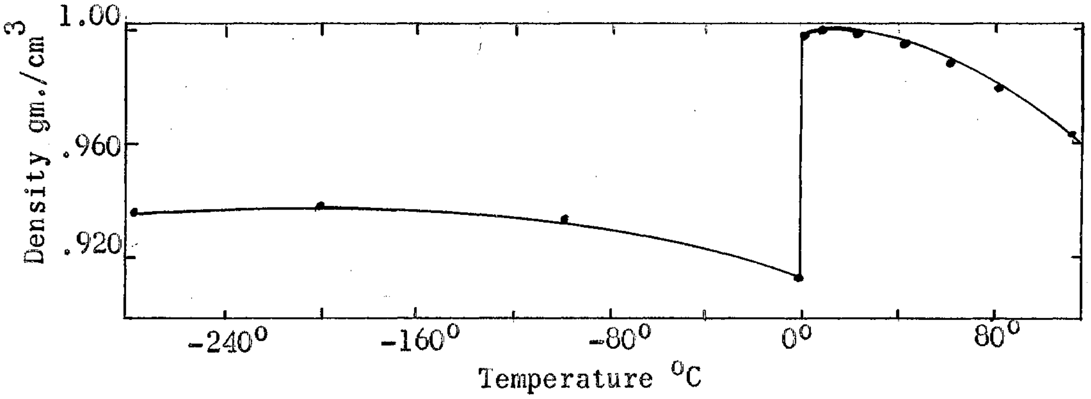
Table V (continued)

Material	^oC	τ_1 (sec.)	%	τ_2 (sec.)	%
polyethylene	20	$(3.0 \pm 0.5) \times 10^{-10}$	71 ± 5	$(2.4 \pm 0.3) \times 10^{-9}$	29 ± 5
borax crystals	20			$(9.0 \pm 2.0) \times 10^{-10}$	~ 30
fused borax	20	$(2.6 \pm 0.5) \times 10^{-10}$	68 ± 5	$(8.7 \pm 1.2) \times 10^{-10}$	32 ± 5
mica	20	$(2.2 \pm 0.5) \times 10^{-10}$	100		
isopropyl alcohol	20			$(2.3 \pm 0.4) \times 10^{-9}$	~ 30

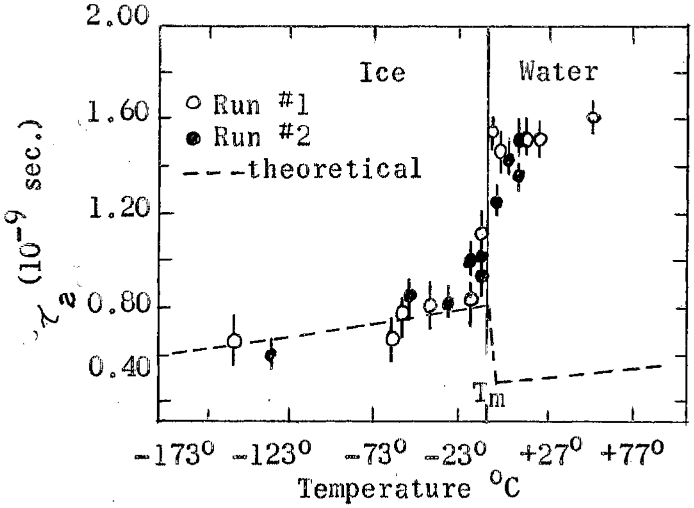
effect of temperature upon τ_2. In the case of polystyrene, water, and Teflon, τ_2 is seen to decrease directly with temperature. This effect is seen much clearer in Figures 4b, 5b, and 6 for the case of water, napthalene, and anthracene. Another interesting effect closely related to this is the effect of temperature upon intensity of the τ_2 component. This is illustrated in Figures 4c, 5c, and 6 again for the case of water, napthalene, and anthracene.

Bell and Graham (5) have proposed the order - disorder effect as an explanation for the change of τ_2 with temperature. Since it is the tendency of the atomic structure to become more orderly upon cooling and the τ_2 mean life decreases for all samples thus far examined, it is highly suggestive of some such relationship. Later DeZafra and Joyner (22) performed angular correlation experiments and as a result proposed the density effect in lieu of the order - disorder effect explanation. They found a direct correlation between density and the peak angular correlation counts. The correlation of τ_2 lifetimes and density is illustrated in Figures 4 and 5 for H_2O and Napthalene. In the case of H_2O the density vs. temperature curve, Figure 4a, is seen to give a very good fit to the inverse of the intensity vs. temperature curve, Figure 4c. For napthalene the density varies in a manner quite the opposite of water yet the intensity vs. temperature curve, Figure 5c, still gives a good fit to the density vs. temperature curve, Figure 5a.

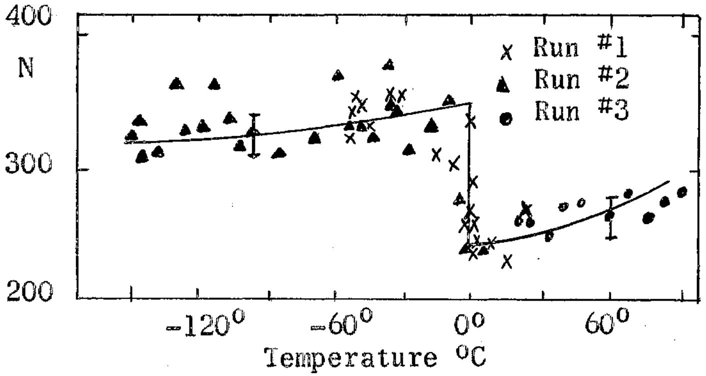
The density effect explanation is further supported by the following theoretical arguments. The "Ore gap" mechanism has been found to give the energetics of positronium formation in condensed matter (36). The width of the Ore gap is determined directly by the ionization potential of the surrounding atoms E_i, and the lowest excitation potential of



a. Density vs. temperature for water. DeZafra and Joyner (22).

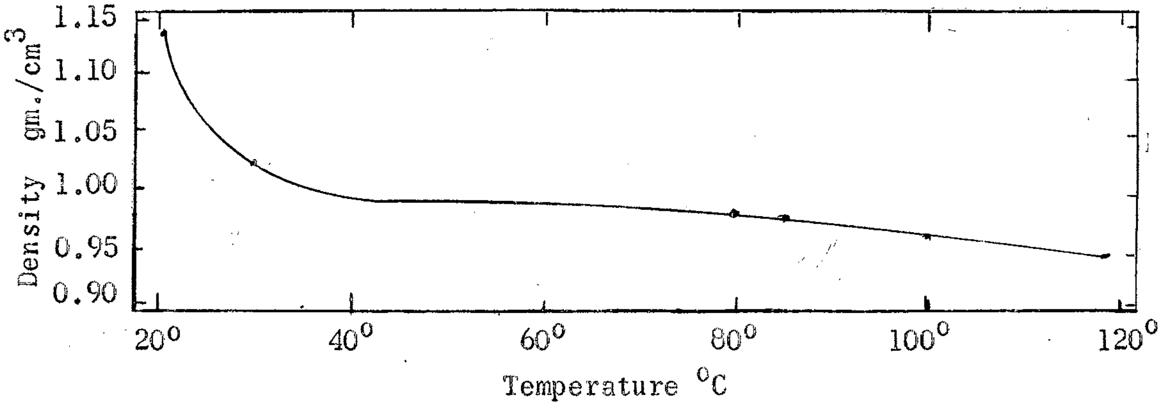


b. τ_2 lifetime vs. temperature for water. Brandt, Berko, and Walker (11).

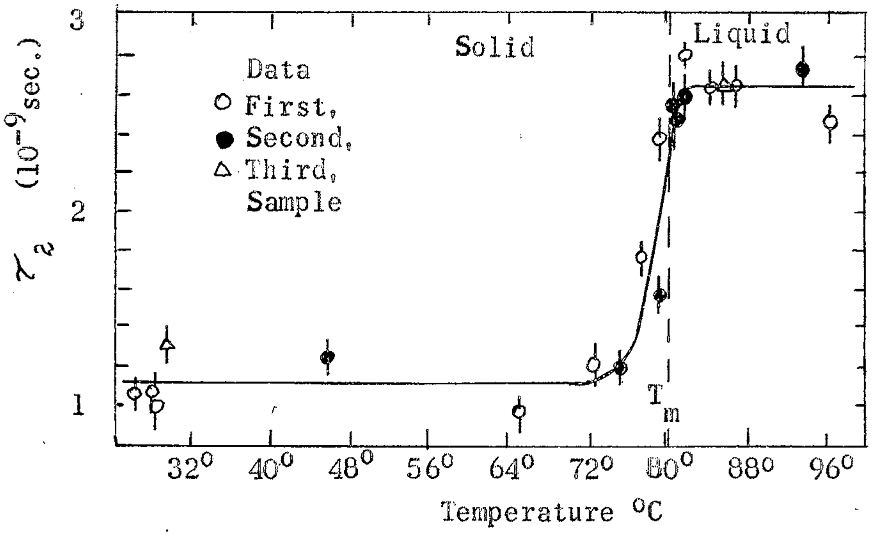


c. Peak rate vs. temperature for water. DeZafra and Joyner (22).

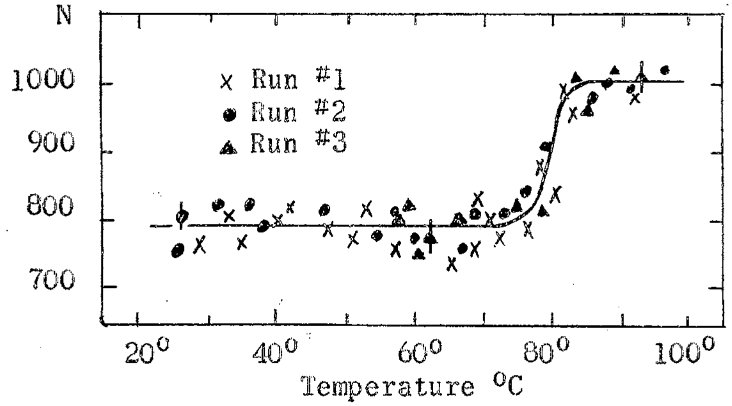
Figure 4. The dependence of density, τ_2 lifetime, and τ_2 peak rate upon temperature for water.



a. Density vs. temperature for napthalene



b. τ_2 lifetime vs. temperature for napthalene
Berko and Hereford (8)



c. Peak rate vs. temperature for napthalene
DeZafra and Joyner (22)

Figure 5. The dependence of density, τ_2 lifetime, and τ_2 peak rate upon temperature for napthalene.

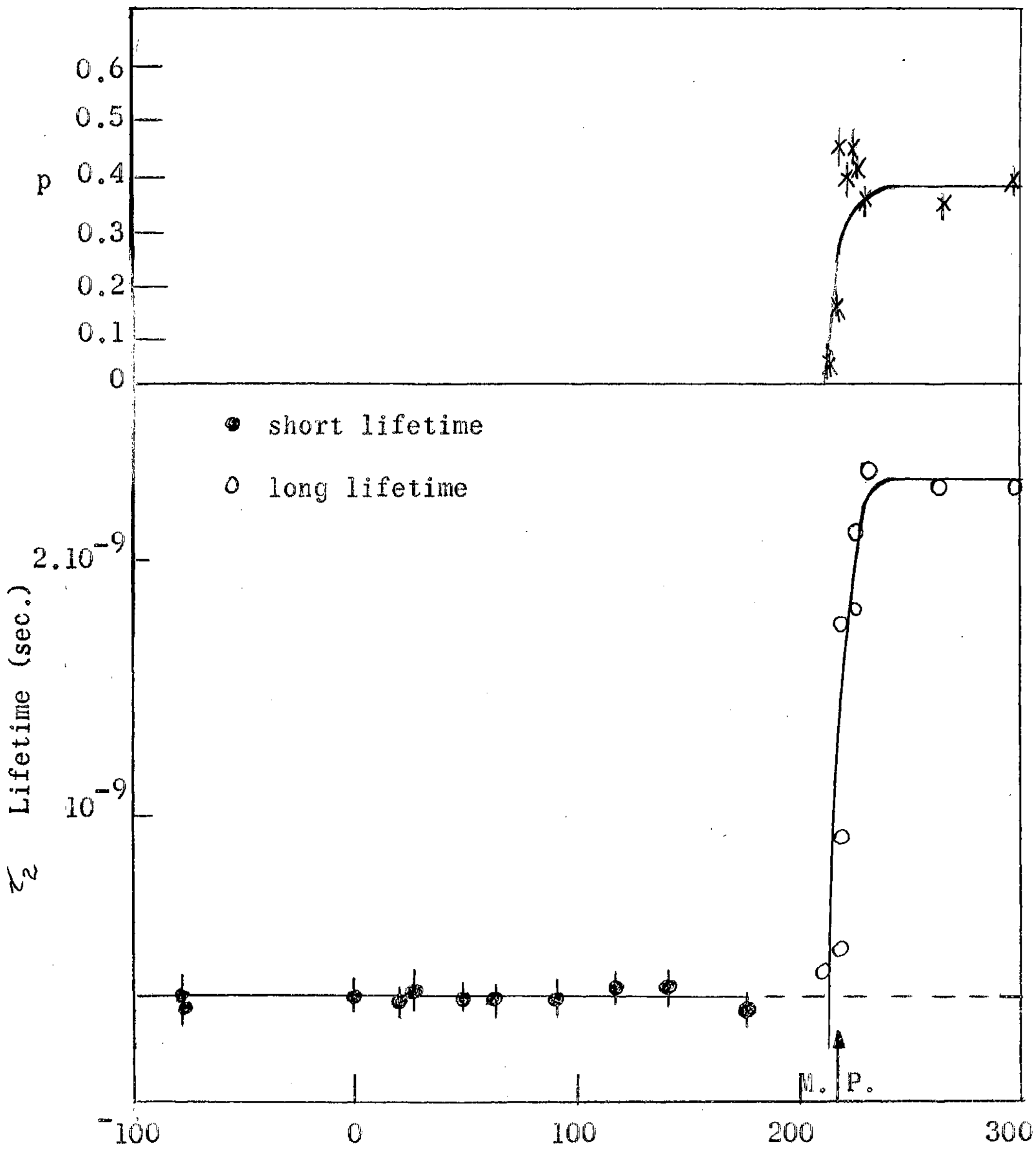


Figure 6. Positron lifetime and percentage "p" of positrons annihilating with a long lifetime as functions of temperature in the case of anthracene. From Conttini (12).

the surrounding atoms E_e, together with the positronium binding energy E_p. Figure 7 gives the relation of these energies and the formation of the Ore gap.

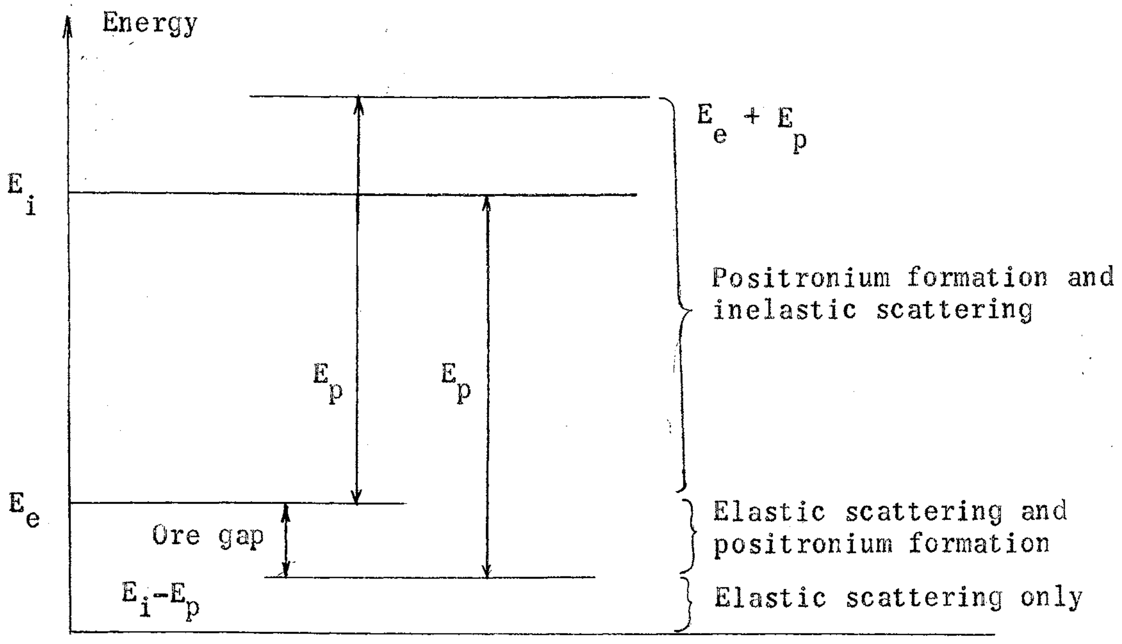


Figure 7. Formation of the Ore gap as determined from E_i, the ionization potential; E_e, the lowest excitation potential; and E_p, the positronium binding energy. The magnitude of the Ore gap is determined from the expression $E_p + E_e - E_i$.

Positronium formation by a positron of energy greater than $E_e + E_p$ is unlikely due to the preference for inelastic collisions, and positronium formation by a positron of energy less than $E_i - E_p$ is impossible due to the lack of sufficient energy to capture an electron. Now as the density of the material increases the binding energy of the positronium decreases due to the cramping of the wave function. It is seen from Figure 8 that the binding energy decreases noticeably when the positronium is compressed to the classical radius of its ground state (two atomic units). This in turn decreases the width of the Ore gap and in turn lowers

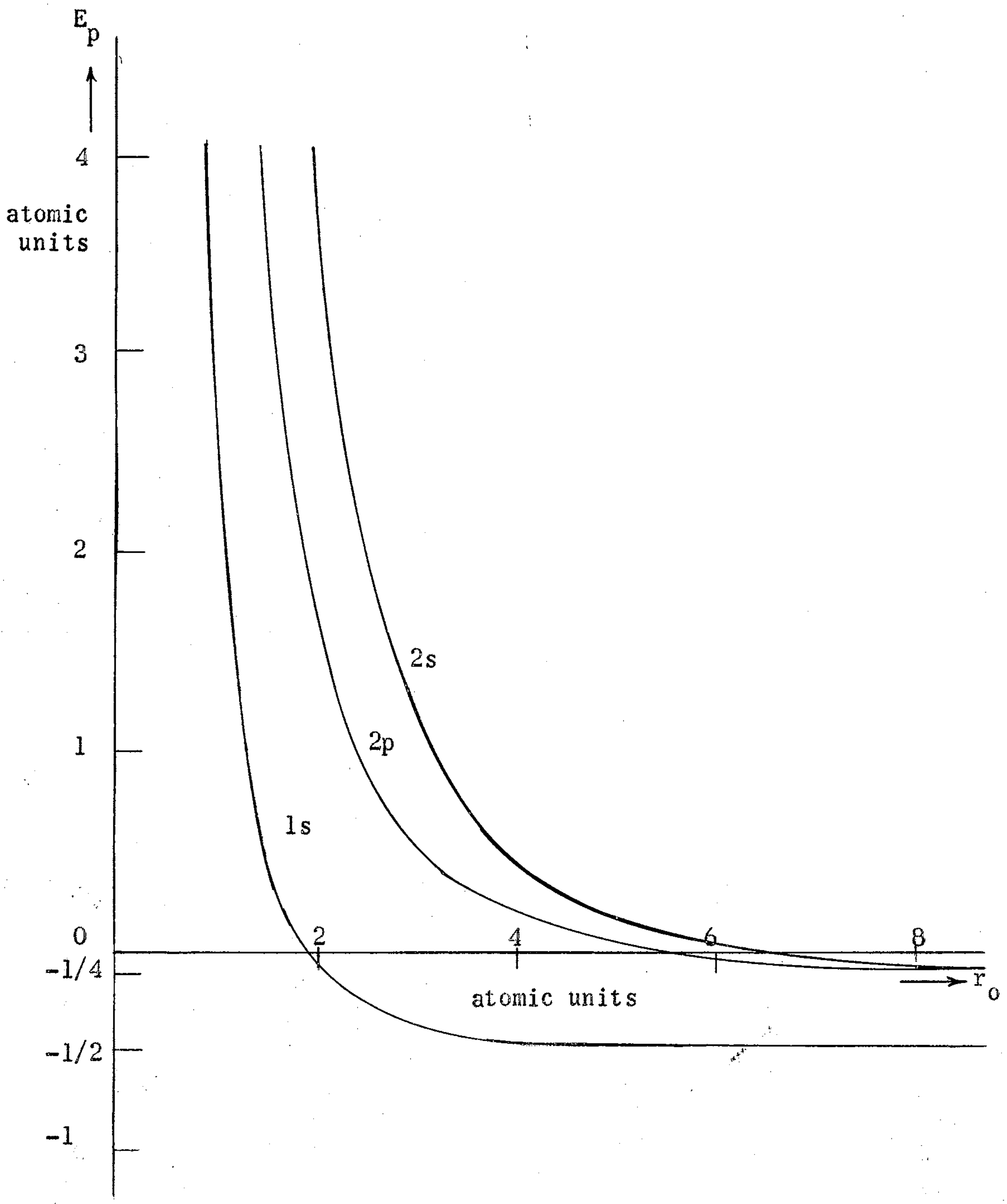


Figure 8. The binding energy of positronium for the 1s, 2s, and 2p energy levels E_p plotted as a function of the distance r_o at which the potential energy of the positronium is zero. From DeGroot and Seldam (16).

the probability of positronium formation.

Another explanation for the variation of τ_2 with temperature is that proposed by Brandt, Berko, and Walker (11), which is closely related to the density. They explain this as a quantum mechanical effect and show by their calculations that the free volume per molecule accessible to the positronium in a typical molecular lattice is 2 - 3 orders of magnitude smaller than the deBroglie wave length of the thermalized positronium atoms. Their work is directed mainly at determining the dependence of τ_2 upon temperature. Their experimental results for τ_2 dependence upon temperature are given in Figures 4b, 9, and 10. In the case of glycerol, polytetrafluoroethylene, and water, the experimental vs. empirical curves are compared with the same theoretical curves. It is seen that for polytetrafluoroethyelene and glycerol a very good correlation results. For water, however, there appears to be an anomaly. Several effects are suggested which may account for this incongruency but it was concluded that further experiments should be conducted in order to establish the changes which occur in the H_2O wave function upon melting and as a function of the dynamic properties of water. They further suggest that a τ_2 vs. a temperature curve as a function of pressure be obtained in order to separate the dynamic effects from the free volume effect.

It is very plausible that the temperature and the phase change dependence of τ_2 is primarily a free volume effect in the case of molecular substances. If this be true, then it would follow that the overlapping of the positronium wave function with the lattice wave functions would increase with the decrease in lattice spacing thus reducing τ_2. As indicated in Figures 4b, 9, and 10, this is essentially what does occur,

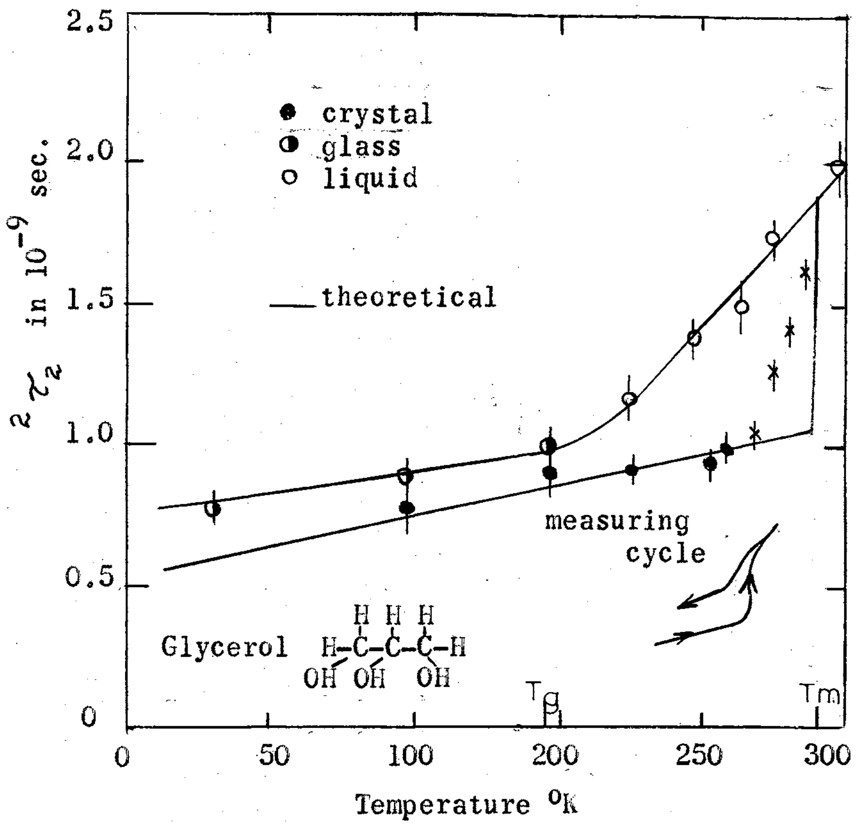


Figure 9. ${}^2\tau_2$ dependence upon v* and temperature in glycerol. The factor v* is proportional to the free volume within the molecular structure which is available for the formation of positronium. From Brandt, Berko, and Walker (11).

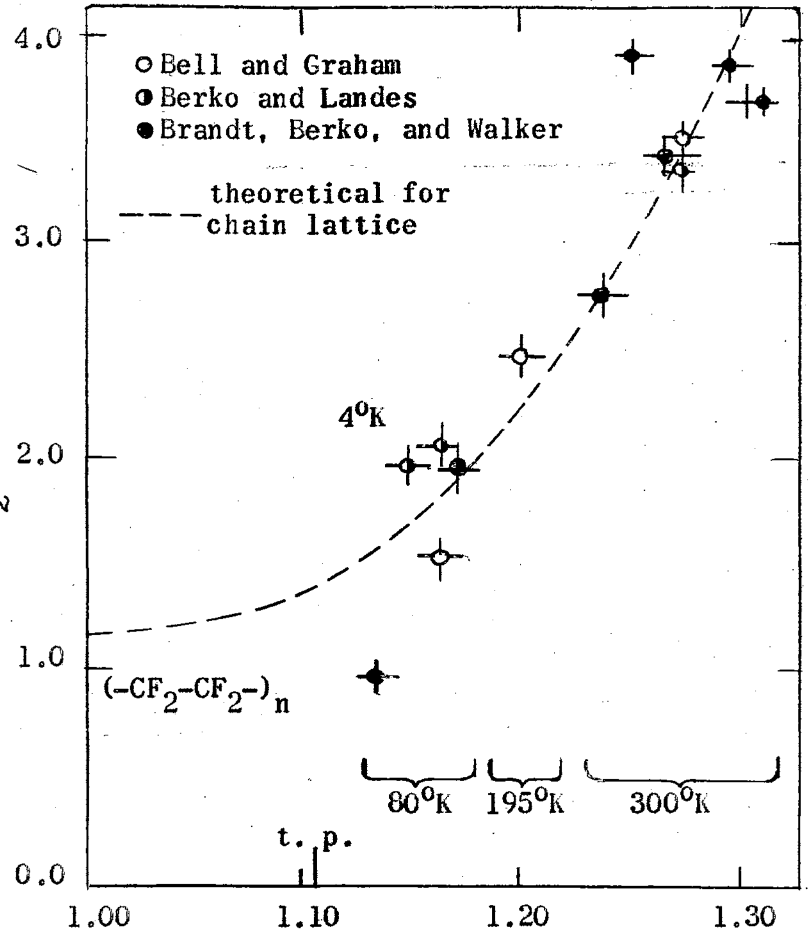


Figure 10. ${}^2\tau_2$ dependence upon v* in polytetrafluoroethylene. From Brandt, Berko, and Walker (11).

with water being the exception.

The application of an external magnetic field is found to decrease the rate of three quantum decay. This may be readily understood as a quantum mechanical effect (41). For the positronium ground state there are two m=0 spin states which become mixed in the presence of a magnetic field. This mixing of states will now lead to $^3S_1$ positronium decaying by two photons with the magnetic field absorbing the unbalanced angular momentum.

The application of a strong RF or static electric field will more than double the amount of positronium formed (21). This effect may be explained by the use of the Ore gap mechanism. Essentially what happens is that if a positron fails to form positronium before thermalizing to energies less than $(E_i - E_p)$ ev, the electric field will accelerate it to higher energies once again, thus giving it another chance to form positronium.

Finally one other factor which affects positronium is the chemical quenching of τ_2. It has been found that certain chemicals such as iodine will virtually eliminate the τ_2 lifetime (26). It is proposed that this could be due to either or both of the following: 1) the chemical binding of iodine and positronium, and 2) the high polarizability of iodine.

Summary: The two factors which seem to affect the τ_2 lifetime most directly and seem to be the most promising for further study are the density distribution of electrons available for positronium formation, and the structure of matter. In spite of all the experimental work already done, there is still a need for further investigation of the temperature and pressure dependence of τ_2 for a larger variety of materials.

CHAPTER IV

THE PRESENT EXPERIMENT

As mentioned in Chapter I, the present experiments were undertaken in order to accumulate further information on the temperature dependence of the τ_2 mean life. As has been shown from the results of earlier investigations (see Table II), it is the non-crystalline materials which display the longest τ_2 lifetimes. It is for this reason that the materials investigated herein are mainly amorphous and molecular in structure. For the specific materials investigated and the results for the τ_1 and τ_2 measurements see Table VI.

Preparation of Samples

The positron source used was Na^{22} which has a halflife of 2.6 years. As indicated by its decay scheme, Figure 11, it decays approximately 100% of the time by positron emission to an excited state of Ne^{22} which emits a gamma ray within 10^{-12} sec. to arrive at the ground state. Although Na^{22} can decay directly to the ground state of Ne^{22}, the probability is very small.

The source was prepared by evaporating $10\mu c$ of $Na^{22}CL$ solution between two 0.002" mylar films. Less than 2% of the positrons will be stopped within this thickness of mylar. Around the source container was placed a sample of sufficient thickness to stop the 0.54 Mev positrons. In the case of Teflon, lucite, and polystyrene, the source was deposited directly upon the sample. For experiments at -195.8°C, -78.5°C, and 0°C, a special dewar was used so that light pipes could be placed within ½" of the sample. The samples were placed within an airtight brass container with the source sandwiched in the middle. After each sample was run, an aluminum curve was obtained. This was necessary because of shifts that occur in the relative position of the curves within the multichannel analyzer. It was found that the channel in which the peak count occurred for aluminum varied as much as 4 channels over a period of 5 days. Each sample was run until the peak count was at least 1000.

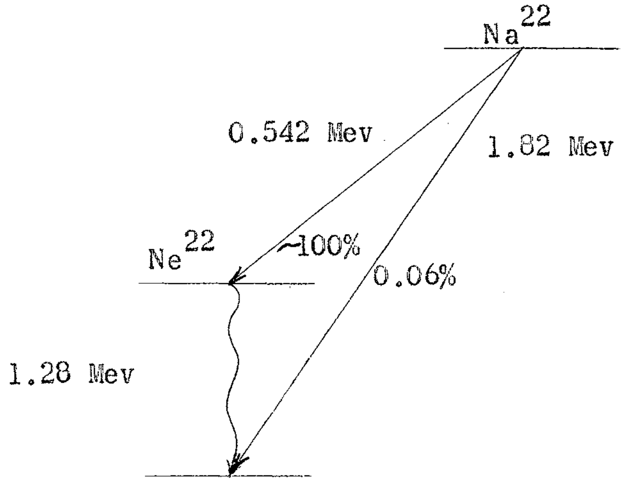


Figure 11. The decay scheme of Na^{22} into Ne^{22}.

Electronic Apparatus

The block diagram of the electronic apparatus used in the present experiments is given in Figure 12. The sample is placed between two light pipes, NE-160 scintillator crystals and photomultipliers A_1 and B_1. The photomultipliers A_1 and B_1 were run at as high a voltage as

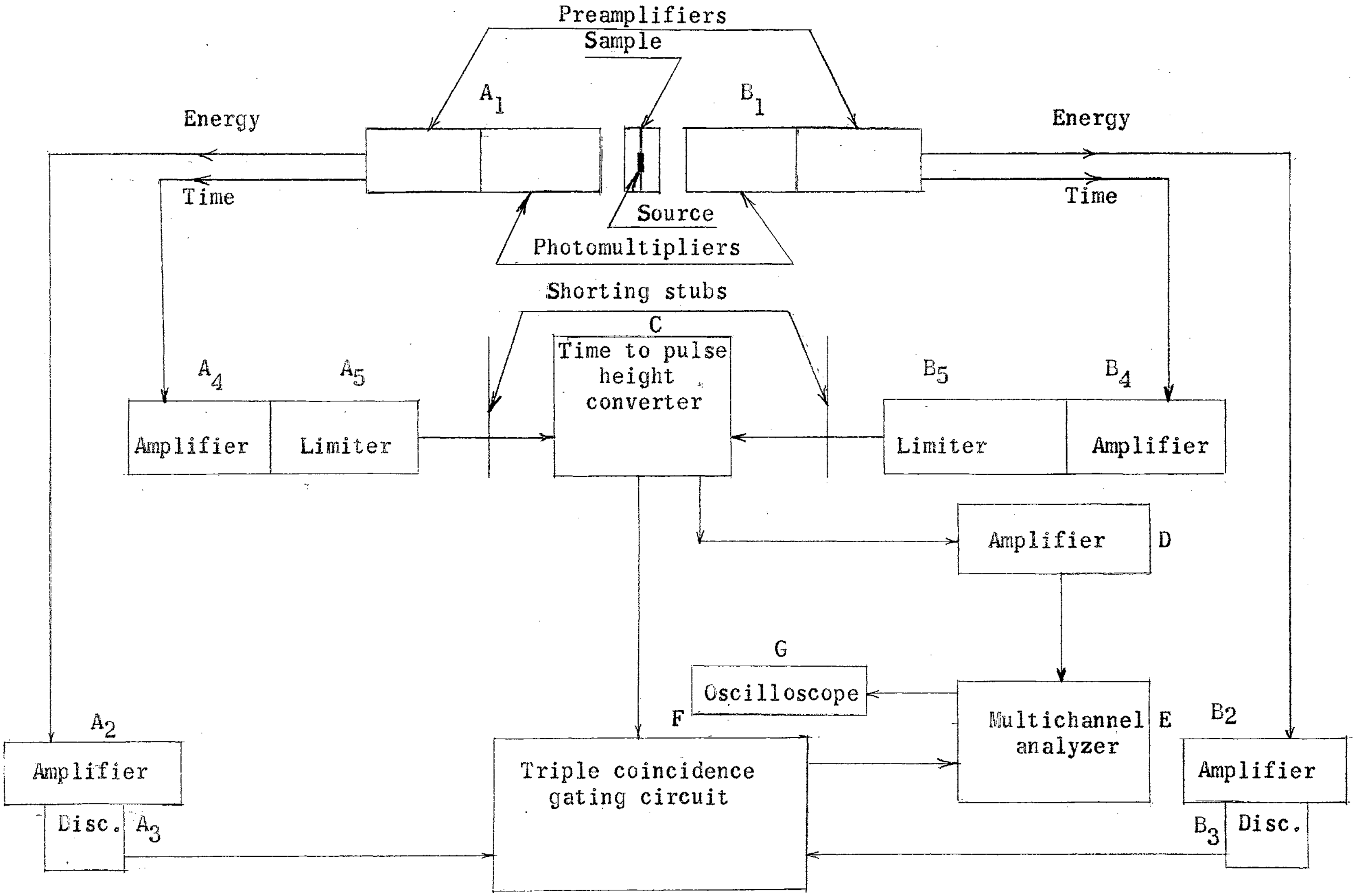


Figure 12. A block diagram of the electronics used in determining the lifetimes.

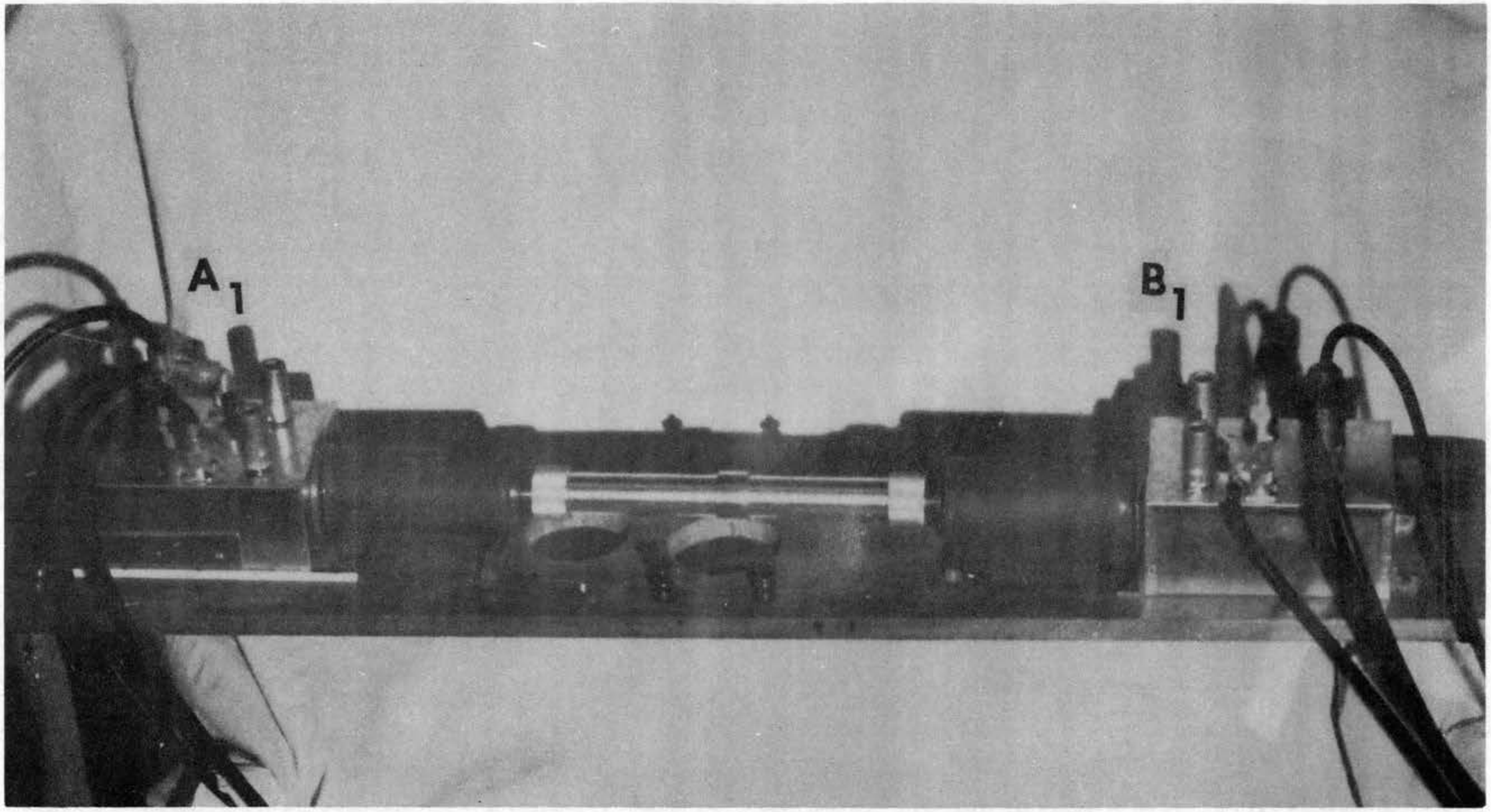


Figure 13. Photograph of the photomultipliers, preamplifers, and light pipes with a sample in place.

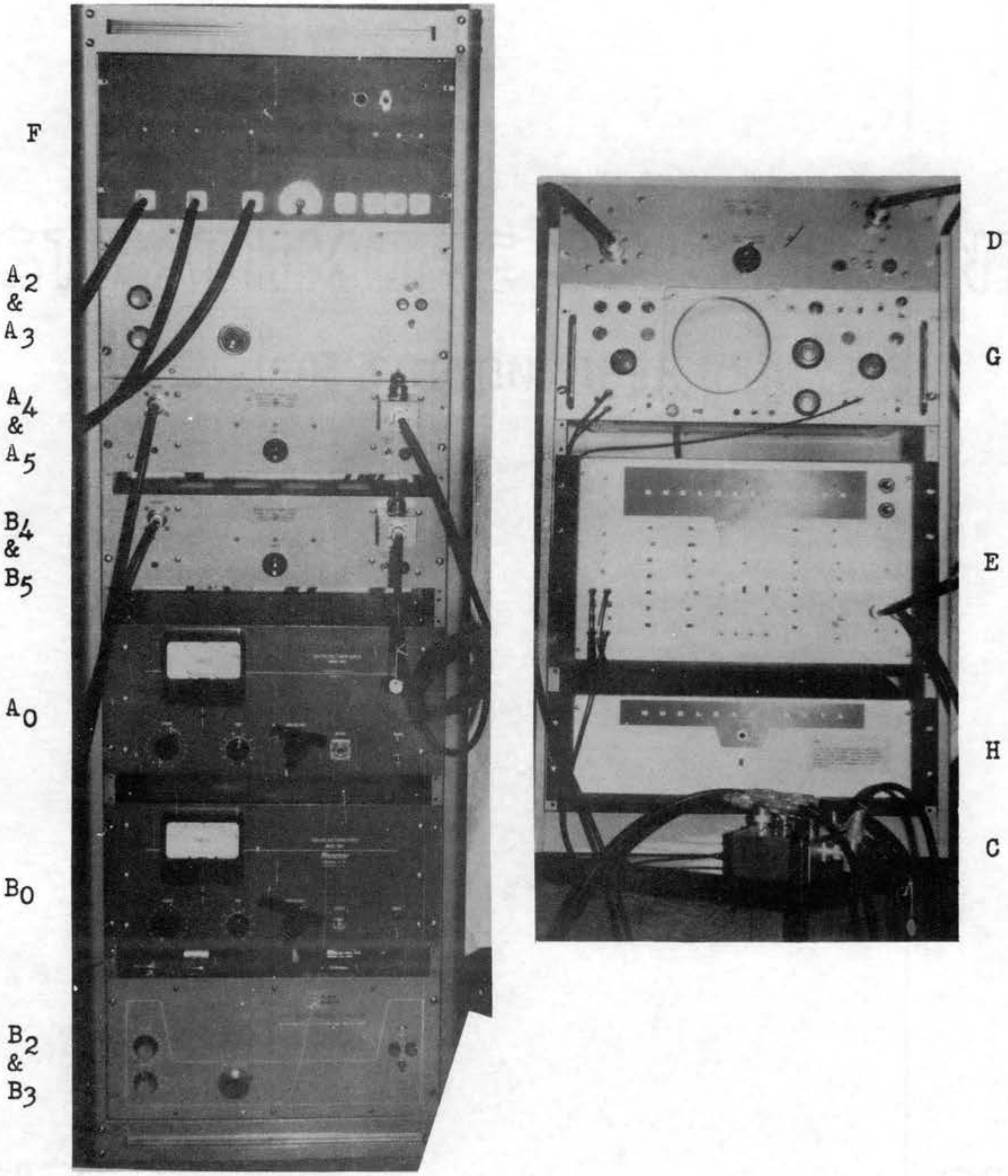


Figure 14. Photographs of the equipment used in determining the mean life of positronium. The block diagram for this equipment is given in Figure 12.

possible without generating excessive noise pulses. Ordinarily this was about 1700 volts. A time pulse and an energy pulse are taken from the preamplifier each time a gamma ray is detected by the scintillators. The energy pulses are amplified by Baird Atomic non-overloading amplifiers A_2 and B_2. The energy pulses from A_1 are then discriminated against at A_3 so that only energy pulses corresponding to the 1.28 Mev gammas will be passed to the triple coincidence gating circuit F. The energy pulses from B_1 are discriminated against at B_3 so that energy pulses corresponding to gammas of 0.51 Mev or greater will be passed to F. It is because B_2 passes both 0.51 Mev and 1.28 Mev pulses that it becomes necessary to limit the activity of the source to $\sim 10\ \mu c$. The time pulses from A_1 and B_1 are amplified by Hewlett-Packard 460AR wide band amplifiers A_4 and B_4. The pulses from the photomultipliers are cut off at 1.9 volts by the limiters A_5 and B_5 and then shorting stubs A_6 and B_6 cut the length of the pulse down to 14.5 nanosecond. They are essentially square wave pulses upon entering the time to pulse height converter C. At C the time difference between the detection of two pulses from A_1 and B_1 is converted into a pulse height where the height is inversely proportional to the time difference between pulses. The superposition of these two pulses is amplified by another Hewlett-Packard wide band amplifier D and is passed to a Nuclear Data 512 channel computer-analyzer E. A second pulse from C goes to the triple coincidence gating circuit F. This gating circuit, as its name implies, permits the multichannel analyzer E to accept pulses from the time to pulse height converter C only when the pulses from A_3, B_3, and C coincide at F. Excepting random spurious counts, such counts correspond to the creation of a positron and its annihilation some 10^{-9} seconds later. The information recorded in the multichannel analyzer may be displayed on an oscilloscope G. A more detailed description of the electronics is given in a thesis by J. Pigg (45).

The instruments were calibrated by using aluminum as the sample and plotting the channel in which the peak occurred as a function of the length of delay cable inserted between the limiters and the time to pulse height converter. The delay cable was RG7/u which has a delay time of 0.039×10^{-9} sec/cm. For the particular instrument settings used, a slope of this graph gave a calibration figure of 0.089×10^{-9} seconds per channel. The background count was determined by averaging the counts recorded in the upper 20 channels of each run where the lifetimes extended only to the upper 50 channels. These counts which correspond to a delay time of 10 - 13 nanoseconds, may be attributed to background only for the length of the runs used.

The data were typed out on an IBM typewriter and all analyses were made from the typewritten data. The curves were plotted in pairs, (the sample and aluminum curve), on four cycle 140 division semi-log paper.

Where the tails of the τ_2 curves were greater than 0.8 nanoseconds, a least squares calculation was used in order to fit a straight line to the tail of the curve. The reciprocal of the slope of this line multiplied by the calibration factor 0.089 nanoseconds per channel yielded the τ_2 lifetimes.

CHAPTER V

RESULTS AND CONCLUSIONS

In selecting the samples to be used, two factors were considered: 1) the amount of free volume, and 2) the Ore gap width. The free volume may be roughly determined from the atomic and intermolecular spacings. For several of the materials investigated, these values are listed in Table VI.

Since the classical radius of positronium is one angstrom, it is expected that the probability of positronium formation will be greater for materials having interatomic distances greater than 2 angstroms than for those having interatomic distances much less than two angstroms. It is for this reason that many inorganic crystals such as carbon and NaCl (see Table VI), do not possess a τ_2 component for their interatomic distances are of the order of 1 angstrom. For organic compounds these distances approach 5 angstroms and for amorphous materials such as polymerized isoprene this may be much greater, thus allowing for a greater probability of positronium formation, assuming the existence of an Ore gap.

In considering the presence of an Ore gap, one must take into consideration the rotational and vibrational energy levels as well as the atomic energy levels. Ordinarily the vibrational energy levels are 10 times smaller than the electronic levels and the rotational levels are 10 times lower than the vibrational levels. Since the lowest ionization energy levels are of the order of 10 ev. and the lowest rotational energy

TABLE VI

THE INTERATOMIC AND INTERMOLECULAR SPACING OF VARIOUS MATERIALS INVESTIGATED FOR τ_2 LIFETIMES

Material	Interatomic Distances	Intermolecular Distances
Aluminum	1.43	
Carbon		
diamond	1.42	3.4
graphite	1.0	2.0
Sulfur		
rhombic	2.1	3.3
Teflon	F - F 2.595	4.0
NaCl	1.4	
p-Dibromobenzene	0.9	1 - 3
Acetamide	CH_3- C 1.51 C - O 1.28 NH_2- C 1.38	2 - 7
Polyethylene	C - C 1.33 C - H 1.09	2.4 - 6.4
Napthalene	C - C 1.404 1.362	(Amorphous only) >5
Isoprene		>5

All units are in angstroms. The spacings between particular atoms is listed below the atoms indicated.

levels are of the order 0.1 ev., one would not expect an Ore gap for a very complex molecule. Similar considerations could be made with respect to the vibrational energy levels with the conclusion that no Ore gap would exist. However, the electronic, vibrational, and rotational energy states should be considered as competing processes for decreasing the positron's energy. This still leaves the electronic states as a possible means of absorbing the positron's energy, thus retaining the possibility of there being an Ore gap. One might conclude that in polyatomic molecules the probability of positronium formation is reduced due to the rotational and vibrational energy levels. The values of the Ore gap for several of the materials investigated for τ_2 lifetimes are given in Table VII. The values of E_e and E_i were obtained from various and sundry references listed in the Chemical Abstracts. These values were calculated from the expression $E_p + E_e - E_i$ and are given in electron volts.

TABLE VII

THE ORE GAP FOR SEVERAL OF THE MATERIALS EXAMINED FOR τ_2 LIFETIMES

Material	Ore Gap
Carbon	-2.93 ev.
Sulfur	-2.35 ev.
Polyethylene	+3.6 ev.
Isoprene	+0.3 ev.
Napthalene	+2.3 ev.
Sodium Chloride	+0.62 ev.
Aluminum	+3.98 ev.

Experimental Results

In analyzing the data for the materials investigated, the least squares method was used where sufficiently long lifetimes were observable, i.e., where $F(x) \gg P(x)$. Values determined by this method are indicated by the presence of the standard deviation. In cases where there were no appreciable tails, i.e., $F(x) \approx P(x)$, the centroid shift method was used for the determination of the lifetimes. The values determined by this method are indicated by the absence of the standard deviation. In Table VIII the values thus determined are listed along with the intensities of the τ_1 and τ_2 components. The graphs of the experimental curves from which the meanlives were determined by the author only are included in Appendix A. The $F(x)$ curve is found by the method outlined by Landes (44).

From an inspection of the τ_2 lifetimes plotted as a function of temperature in Figure 15, one sees that there is a gradual transition taking place in lucite between 23°C and -78°C. Below -78°C it appears as if the rate of change of τ_2 with temperature is very slight. This feature has also been noticed in water, Figure 4b; napthalene, Figure 5b; and anthracene, Figure 6. Actually such a radical change of τ_2 between 23°C is not to be expected for lucite since the melting temperature is approximately 150°C. It would be interesting to see if the τ_2 lifetime versus temperature curve levels off again before lucite reaches its melting point. If such did happen, this would be very indicative of some radical structural change occurring at a temperature somewhat below its melting point.

Also in Figure 15 are the results for polymerized isoprene τ_2 lifetimes versus temperature results. Here the τ_2 lifetime is seen to decrease much faster with temperature than does lucite. It can not be readily said

TABLE VIII

THE VALUES OF τ_1 AND τ_2 LIFETIMES AND THEIR RESPECTIVE INTENSITIES I_1 AND I_2 AS DETERMINED BY THE AUTHOR AND HIS COLLEAGUES

Material	τ_2 (10^{-9} sec.)	I_2 %	τ_1 (10^{-10} sec.)	I_1 %
Boric Acid	1.1 ± 0.2	26	1.6	74
Rhombic Sulfur			2.3	
Fused Borax			7.0	
Polystyrene				
23°C	2.32 ± 0.1			
-195.°C	1.75 ± 0.1			
Teflon	3.6 ± 0.4			
Apiezon	1.3 ± 0.1			
Polyethylene	2.5 ± 0.1			
Acetamide	0.89 ± 0.1			
Napthalene crystals	0.8*			
Amorphous Napthalene	1.33 ± 0.1	8	4.1	92
p - dibromobenzene			2.1	
Aniline Hydrochloride	1.1 ± 0.2	26	2.1	74

Table VIII (continued)

Material	τ_2 (10^{-9} sec.)	I_2 %	τ_1 (10^{-10} sec.)	I_1 %
Isoprene				
23°C	1.8 ± 0.2	16	2.6	84
-78°C	1.1 ± 0.2	13	1.0	87
-195°C			2.8	
Lucite				
23°C	2.39 ± 0.06			
0°C	2.10 ± 0.06			
-78°C	1.52 ± 0.06			
-195°C	1.24 ± 0.06			

*This was determined from the slope of a visually fitted curve.

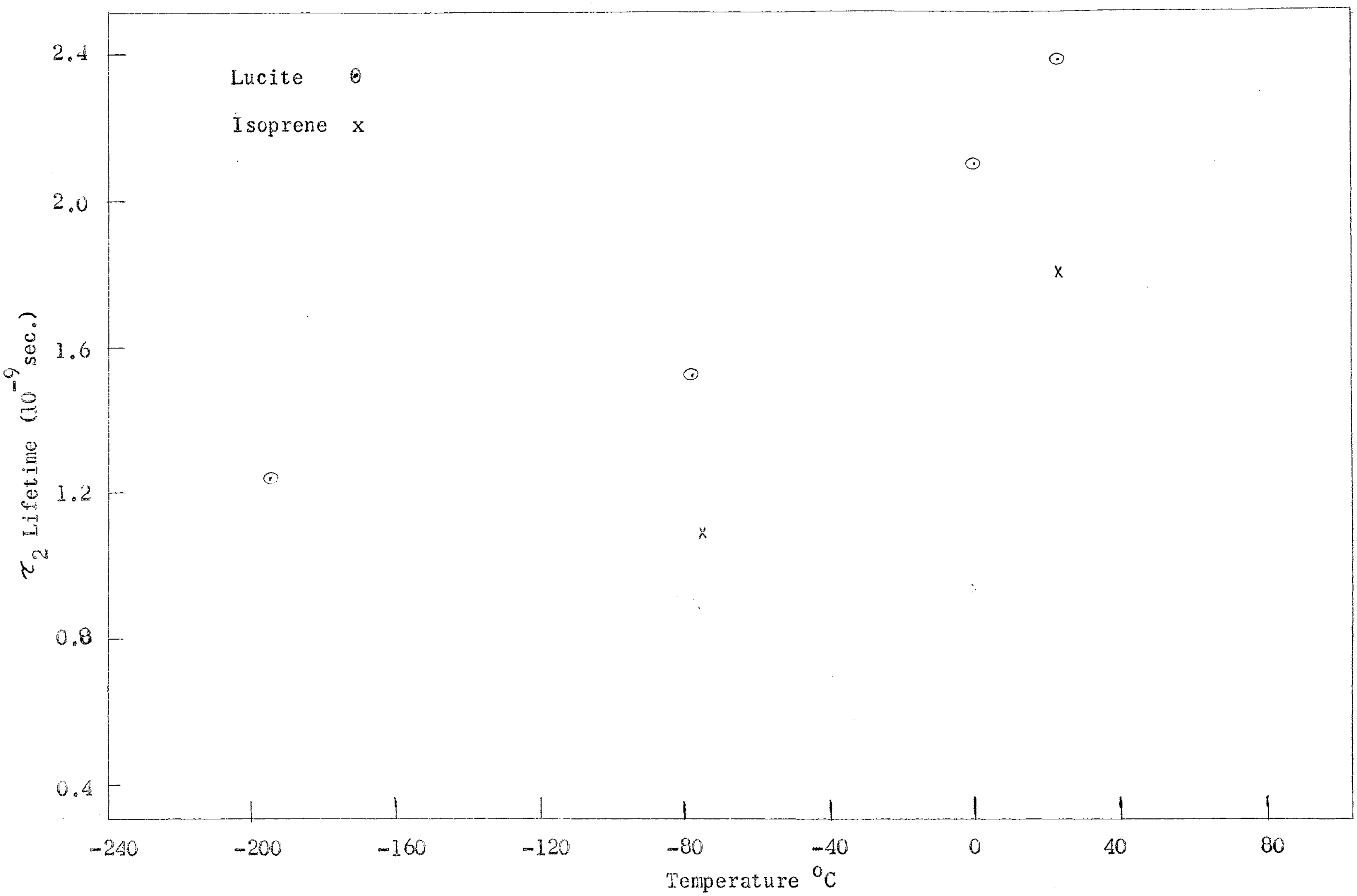


Figure 15. The τ_2 lifetimes for isoprene and lucite plotted as a function of temperature.

that there is no τ_2 component at -195°C since any lifetimes much smaller than 1 nanosecond were difficult to measure with the particular apparatus used. Isoprene melts at temperatures above 150°C, thus the occurrence of approximately 1×10^{-9} sec. change of τ_2 at 78°C is unusual. In order to adequately explain the rapid decrease in lifetimes for both lucite and isoprene as observed here, it will be necessary to investigate structure of these materials possibly by a study of infra-red spectrographs in order to determine by independent measurements what structural changes occur at these temperatures. Another interesting experiment which may be performed in this respect is to use several materials which go through structure changes at some point other than their melting or boiling points. By choosing these materials so that their degree of structure change became progressively larger from material to material one would be able to obtain some experimental information as to whether or not the τ_2 lifetime is directly proportional to the free volume. The reason that these transitions should not include the melting region is that as material melts, the electronic energy levels may change a great deal more than the changes brought about by the relative orientation of atoms in molecules such as the small contraction of the Teflon chain molecule at 20°C.

Also interesting to note is a comparison of the experimental results and the information listed in Tables VI and VII. This comparison is given in Table IX where either "Yes" or "No" is used to indicate if there is an Ore gap or sufficient free volume. It is readily observed that it is only where both sufficient free volume and an Ore gap occur that a lifetime has been observed. Conversely, in every case in which there exists sufficient free volume and an Ore gap, a τ_2 lifetime has also been observed.

TABLE IX

A COMPARISON OF THE EXPERIMENTAL RESULTS, TABLE VIII, AND THE INFORMATION LISTED IN TABLES VI AND VII

Material	Ore Gap	Free Volume	Lifetime
Sulfur Crystals	No	Yes	No
Polyethylene	Yes	Yes	Yes
Isoprene	Yes	Yes	Yes
Carbon	No	Yes	No
Napthalene Crystals	Yes	No	No
Napthalene Amorphous	Yes	Yes	Yes
Sodium Chloride	No	No	No
Aluminum	Yes	No	No

There are several interesting experiments which may yet be performed. From Table VI, it was seen that there did exist an Ore gap for aluminum yet there was not sufficient free volume for positronium formation. This suggests that an investigation of the τ_2 lifetimes as a function of the vacancy density would be in order. Particularly interesting in this respect would be a study of titanium dioxide since it is highly nonstoichiometric, and vacancies of the order of 10^{+19} per cubic centimeter may be generated.

Another aspect that has not been considered thus far is the positron affinity of materials. Such a positron affinity can be due to large dipole moments which affects the lifetimes sharply. Nitric oxide is such a material and produces a strong quenching of the lifetimes. Here it would be of interest to determine just how sensitive positron lifetimes are to the dipole moment.

It is apparent from the results thus far obtained from positronium annihilation experiments that it is quite sensitive to the structural and electronic characteristics of matter. In light of other effects, such as vacancy concentration, which are yet to be studied, it is yet too early to decide the exact dependence of positronium lifetimes upon the parameters of its surroundings.

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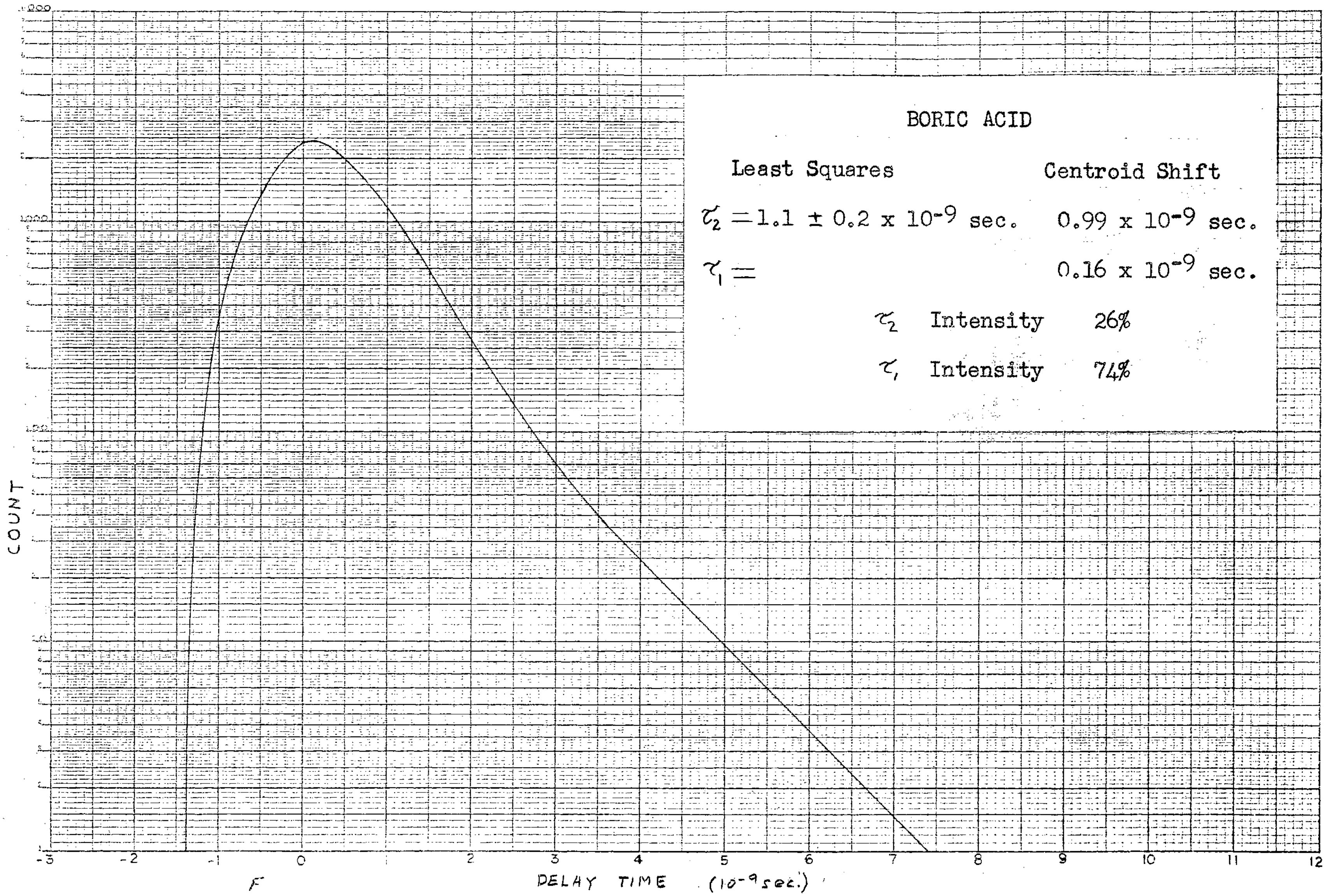
APPENDIX



BORIC ACID

	Least Squares	Centroid Shift
$\tau_2 =$	$1.1 \pm 0.2 \times 10^{-9}$ sec.	0.99×10^{-9} sec.
$\tau_1 =$		0.16×10^{-9} sec.

τ_2 Intensity 26%

τ_1 Intensity 74%

Figure 16. Boric Acid

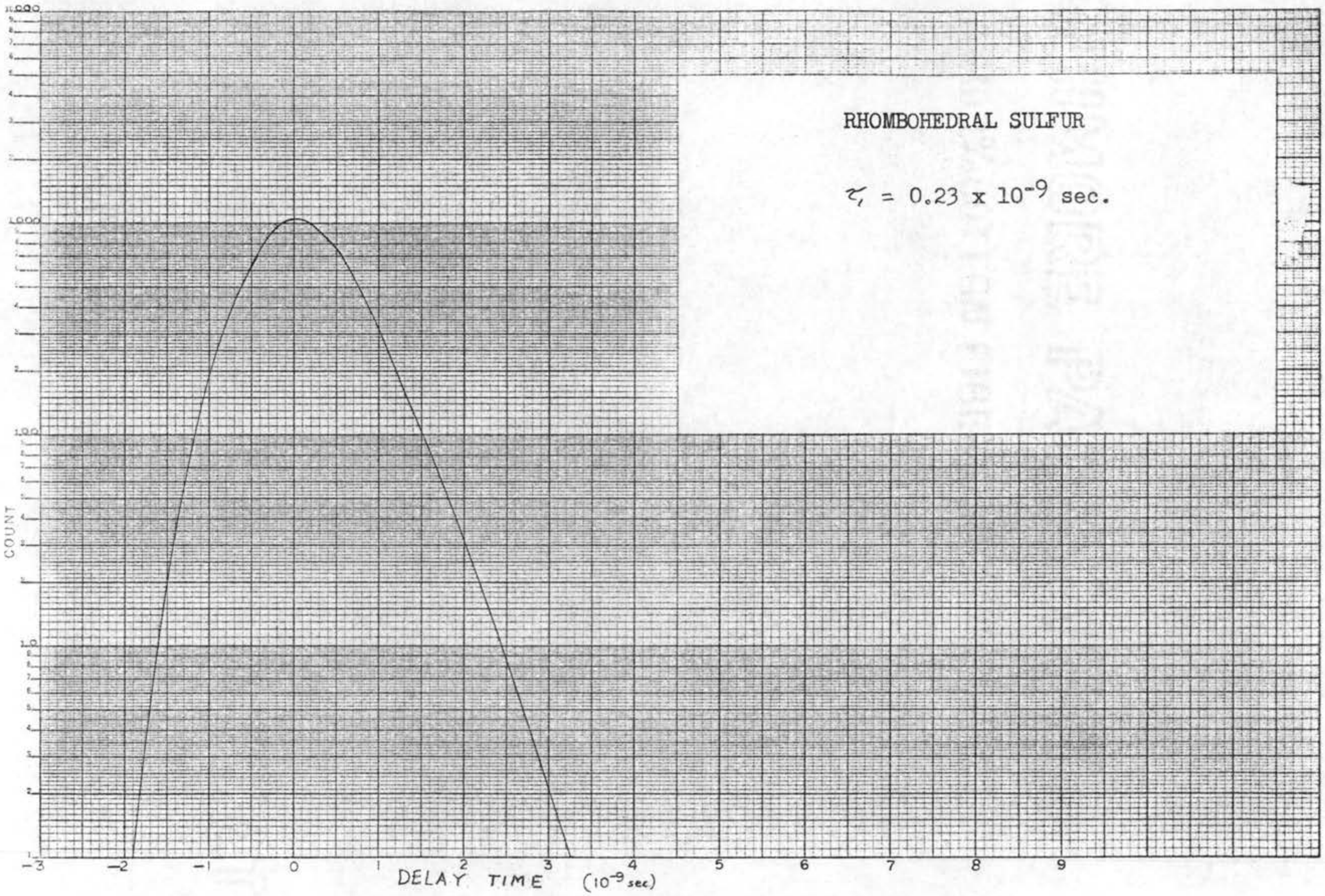


Figure 17. Rhombohedral Sulfur

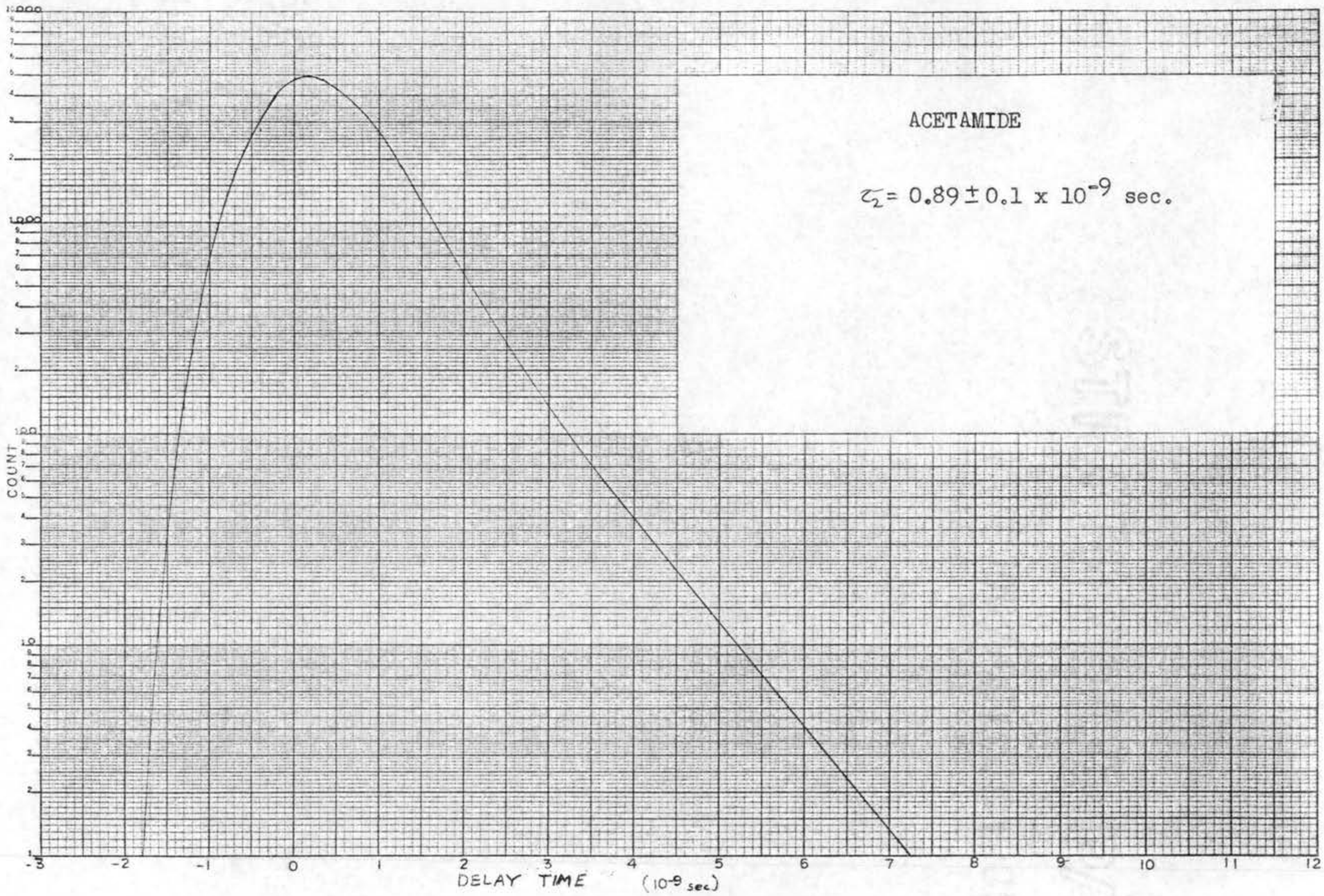


Figure 18. Acetamide

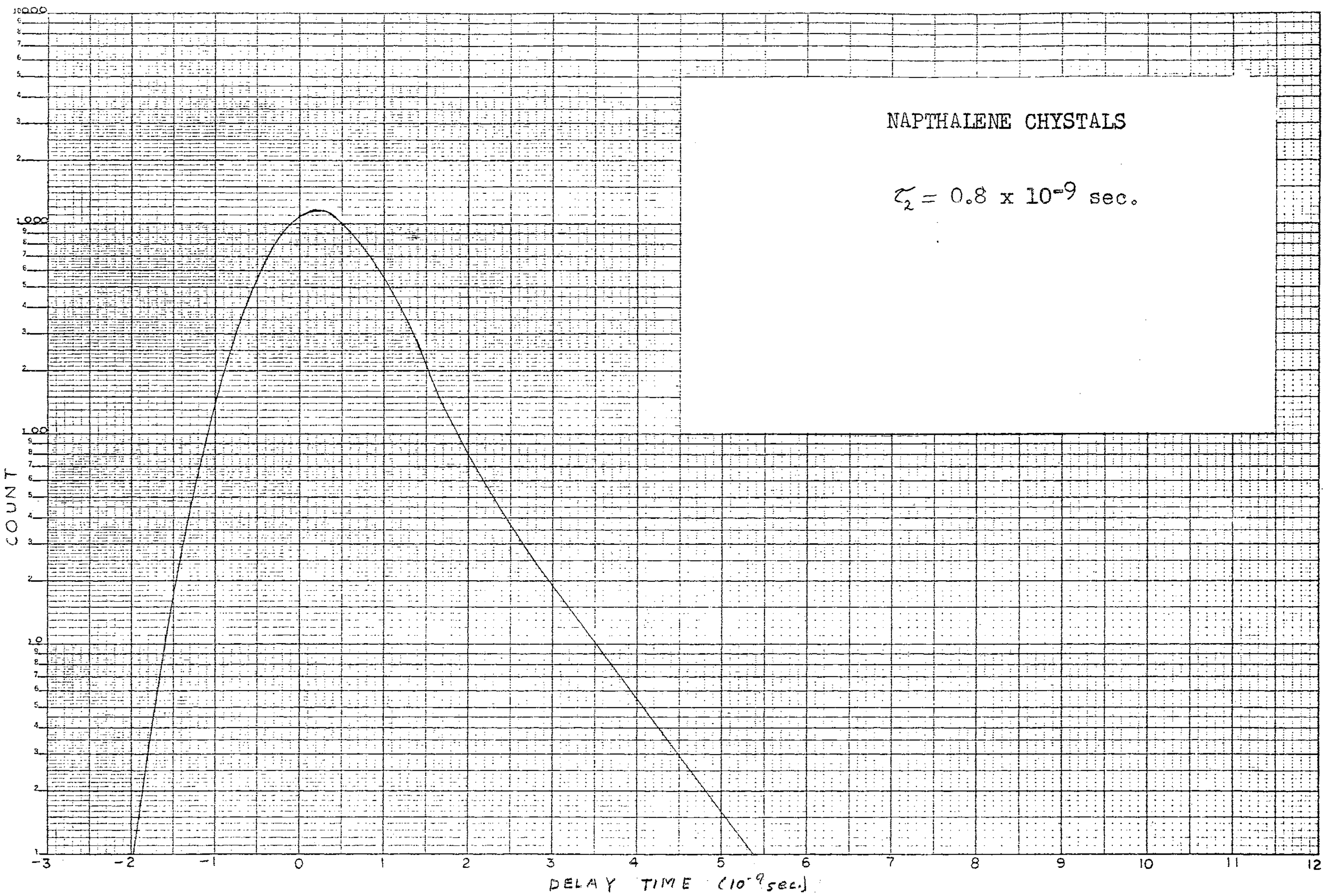


Figure 19. Napthalene Chystals

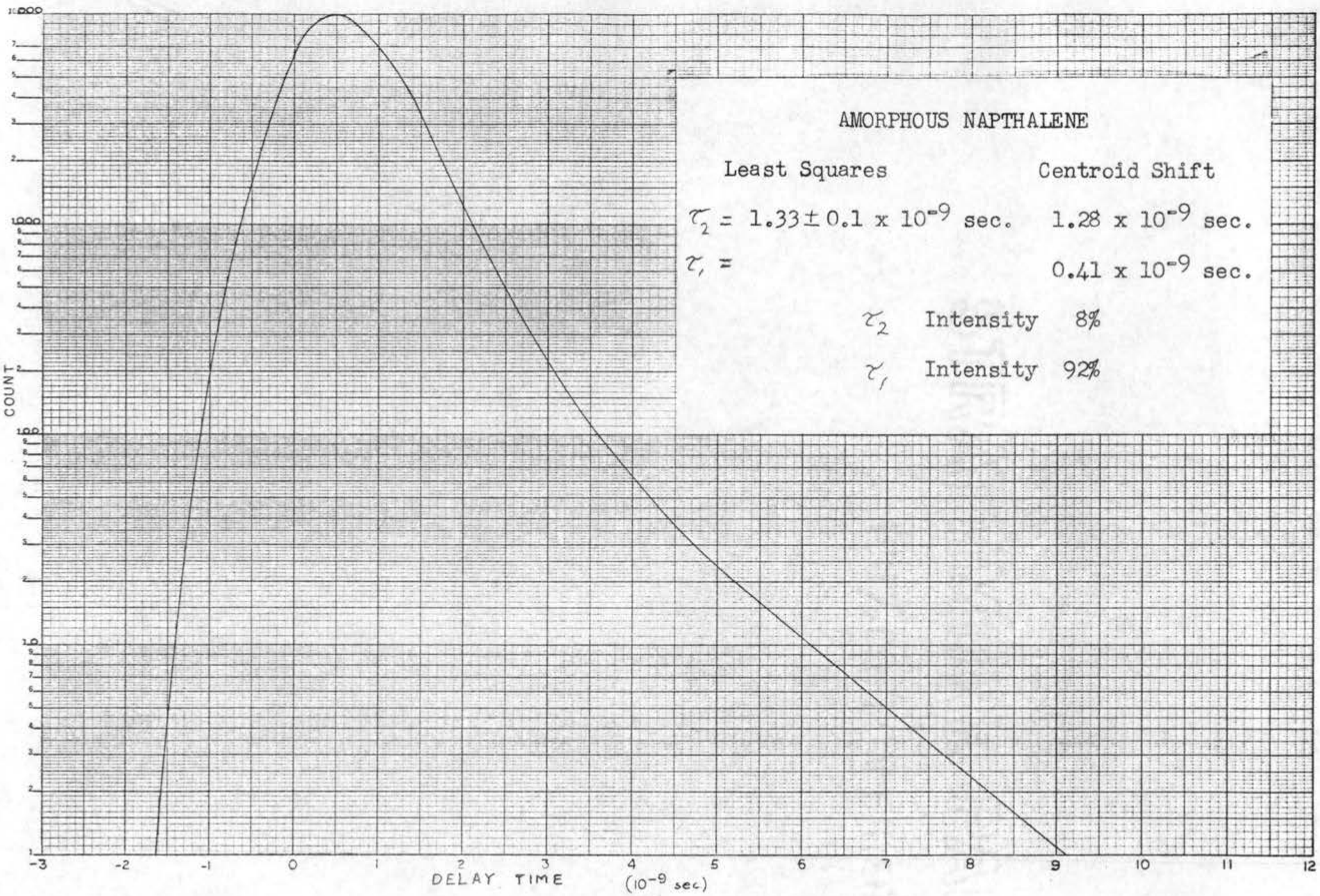


Figure 20. Amorphous Napthalene

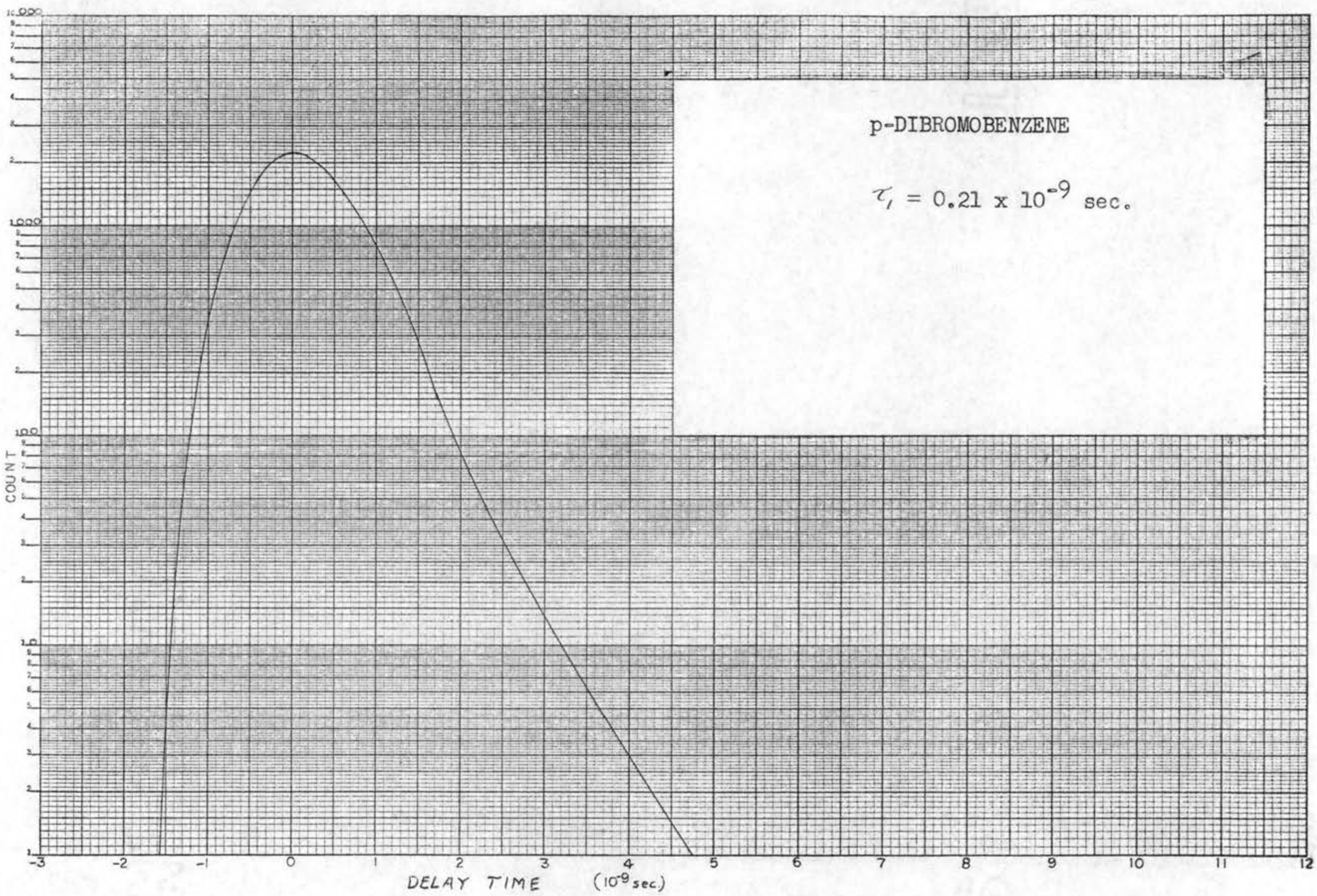
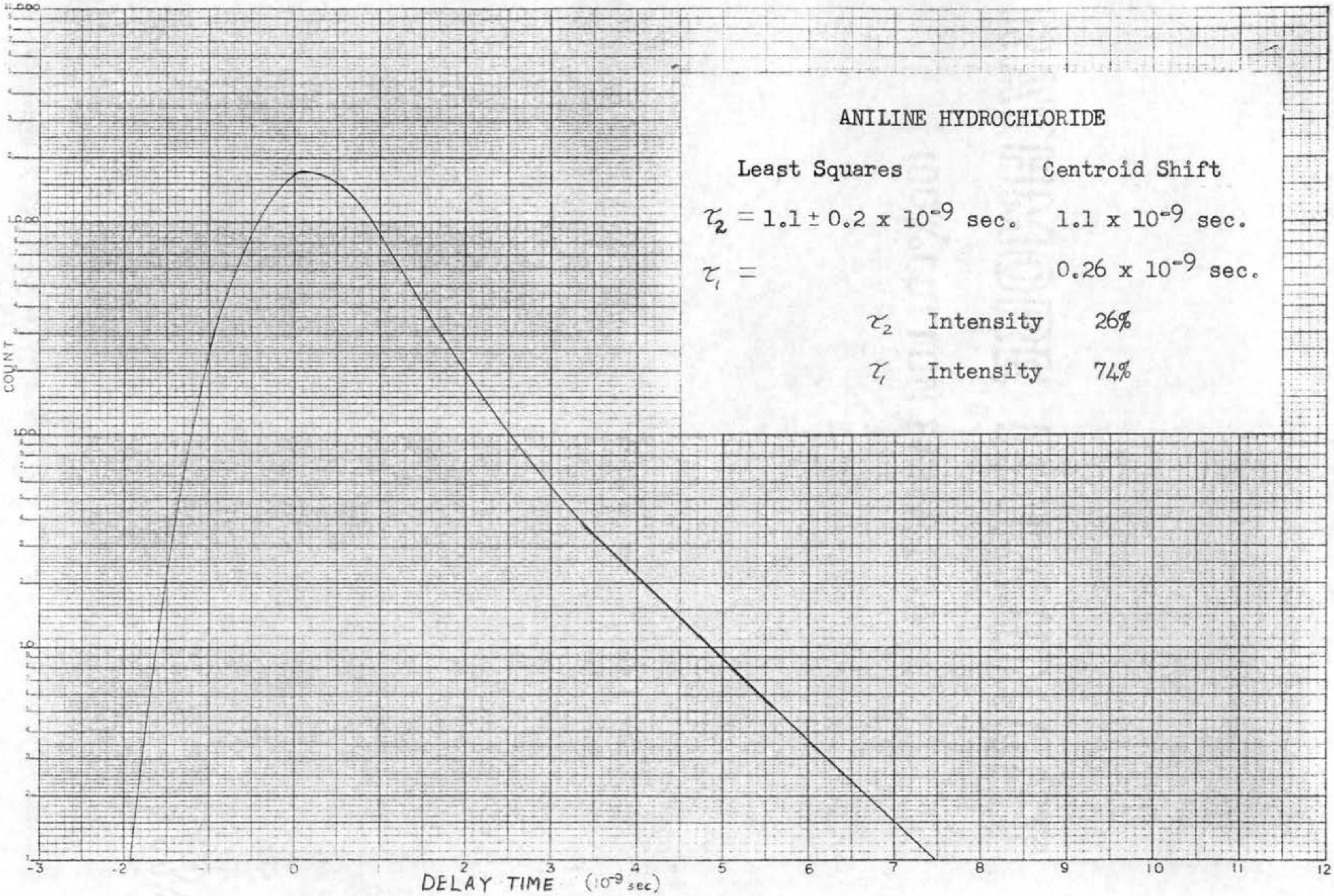


Figure 21. p-Dibromobenzene



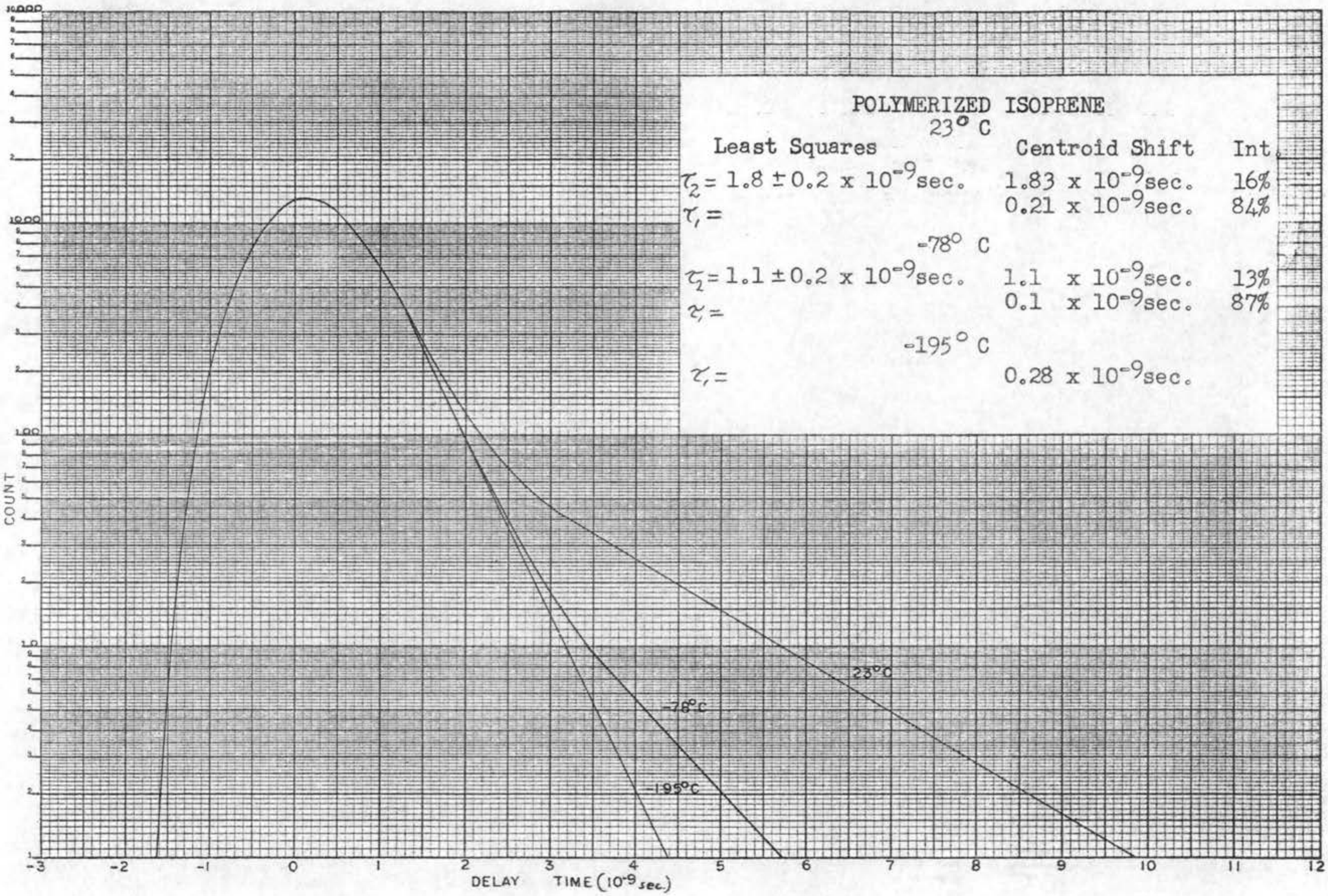
ANILINE HYDROCHLORIDE

	Least Squares	Centroid Shift
τ_2 =	$1.1 \pm 0.2 \times 10^{-9}$ sec.	1.1×10^{-9} sec.
τ_1 =		0.26×10^{-9} sec.

τ_2 Intensity 26%

τ_1 Intensity 74%

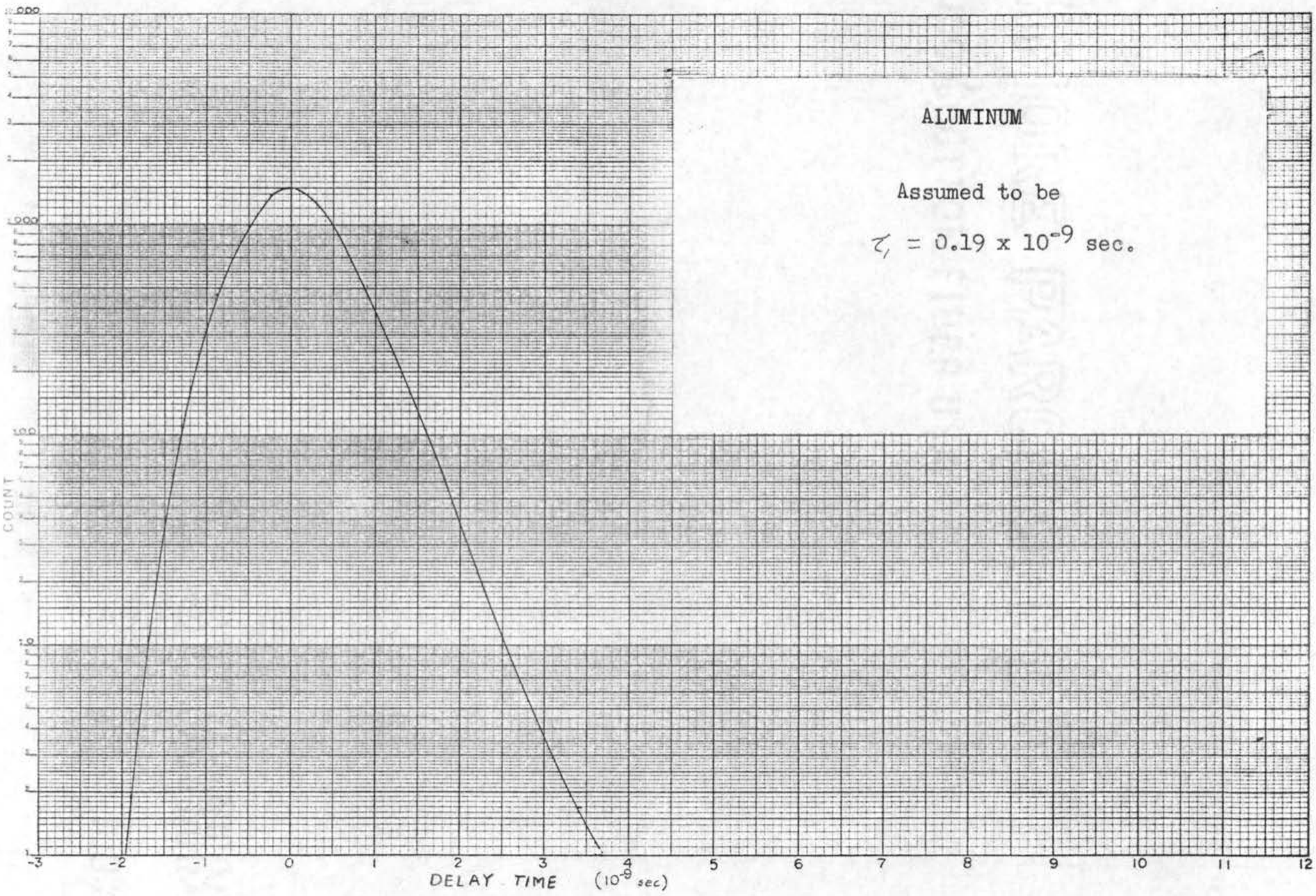
Figure 22. Aniline Hydrochloride



POLYMERIZED ISOPRENE

	Least Squares	Centroid Shift	Int.
	23° C		
$\tau_2 =$	$1.8 \pm 0.2 \times 10^{-9}$ sec.	1.83×10^{-9} sec.	16%
$\tau_1 =$		0.21×10^{-9} sec.	84%
	-78° C		
$\tau_2 =$	$1.1 \pm 0.2 \times 10^{-9}$ sec.	1.1×10^{-9} sec.	13%
$\tau_1 =$		0.1×10^{-9} sec.	87%
	-195° C		
$\tau_1 =$		0.28×10^{-9} sec.	

Figure 23. Polymerized Isoprene



ALUMINUM

Assumed to be

$\tau_1 = 0.19 \times 10^{-9}$ sec.

Figure 24. Aluminum

VITA

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