TIME SPECTRA OF POSITRON ANNIHILATION IN SOME LIQUIDS AND SOLIDS

by

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This Thesis is Submitted in Partial
Fulfillment of the Requirements for the Degree of
Master of Science

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JULY, 1963

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ACKNOWLEDGEMENTS

I should very much like to thank Dr. B. G. Hogg for his generous assistance in this experiment and for the interesting discussions which he encouraged.

Also I should like to express my appreciation to Mr. D. P. Kerr for his help in the initial stages of the experiment.

This work was supported by the National Research Council of Canada and the Petroleum Research Fund of the American Chemical Society.

ABSTRACT

The change in the long lifetime (τ_2) for positron annihilation has been measured as a function of the temperature for Diphenylamine, N-Phenylbenzylamine and P-Bromotoluene. In each case the temperature range covers a first order phase transition of the substance under investigation.

The annihilation time spectra of positrons were studied in LiH and NaH. In both cases a complex spectrum was obtained. NaH showed a long lifetime of 2.6 nanoseconds and a short one of 0.43 nanoseconds while LiH showed a long one of 2.1 nanoseconds and a short one of 0.41 nanoseconds. The long components had an intensity of 10% in LiH and 15% in NaH.

CHAPTER I

INTRODUCTION

Several authors (Ruark[1], Ore[2]) have delved into the theoretical considerations of the interactions of positrons and electrons. They have predicted that a system consisting of a positron and an electron can exist metastably in vacuum and in a suitable nonvacuous environment. The bound system was given the name positronium by Ruark. The quantum mechanical argument closely parallels that for the hydrogen atom and to first order theory the "Bohr" radius of the ground state is 1.06 Angstroms and the energy of formation is 6.8 ev. in vacuum.

The ground state configuration consists of the 1^1S (singlet positronium) and the 1^3S (triplet positronium) substates. From statistical considerations it follows that the triplet substate should be three times as probable as the singlet. Spontaneous transitions between the ground state substates are absolutely forbidden in the absence of an external magnetic field, however both states are unstable against self-annihilation. The conservation laws infer that in the absence of an external electromagnetic field the singlet state decays with the emission of two quanta of electromagnetic radiation correlated at 180 degrees in the center of mass coordinates, whereas the triplet state annihilates with the subsequent appearance of three coplanar quanta. Single quantum annihilation may occur in the presence of a strong magnetic field.

The mean lifetimes against annihilation have been calculated for both states in a vacuum environment.

$$\tau_1 \text{ (singlet)} = 1.25 \times 10^{-10} \text{ sec.}$$

$$\tau_2 \text{ (triplet)} = 1.39 \times 10^{-7} \text{ sec.}$$

(Ore[1] and Powell[3]; Ferrell[4]).

It was pointed out by A. Ore that the formation of positronium in a nonvacuous environment is energetically favored only in a rather restricted range of positron velocities. When a positron enters a nonvacuous medium it generally does so with an energy which is greater than some exitation potentials of the atoms in the attenuating material. At such energies the cross sections for inelastic collisions are considerably larger than the cross section for the formation of positronium. As a result the positron will most probably engage in a number of inelastic collisions. The probability for the binding process becomes important in a fairly narrow range of velocities roughly centered about the "orbital" velocity of the outer atomic electrons of the environment. At lower velocities elastic collisions predominate.

Ore[1] has given the lower bound of the energy for which binding is possible in the case of an atomic gas. If E_i is the ionization energy for gas atoms, and if E_p is the binding energy of positronium (approximately 6.8 ev.) then the lower bound for the positron energy E' is given by:

$$E' = E_i - E_p$$

R. A. Ferrell[4] has extended the theoretical treatment to a more general case.

Binding with free electrons is not possible in the absence of an external field.

Now experimentally one measures the mean lifetimes of positrons in matter. Annihilation from the bound state rather than direct annihilations of positrons was inferred by Deutsch and Shearer who noticed that the rate of annihilation was not proportional to the gas pressure for the short lifetime component. Backing for this argument was provided by the triple coincidence measurements by deBenedetti and Siegel[5] and by Deutsch and Dulit[6] who observed the quenching of the triplet state by an applied magnetic field.

However, in order that one may speak meaningfully about the absolute lifetimes of positronium in a given medium, one requires the assurance that the thermalisation time is small in comparison to the lifetime of the bound state. In other words, the inelastic collisions must be sufficiently effective in attenuating the initially fast positron to such velocities where the binding cross section is high. Calculations of thermalisation times have been carried out by deBenedetti et al.[5], Lee-Whiting[7] and Wallace[8]. They indicate a time of the order of 10^{-12} seconds for metals. Reliable estimates of thermalisation times of positrons in insulators and liquids do not exist at present. It will be assumed here that it is reasonable to discuss absolute lifetimes where fast positrons from Na^{22} are employed.

Experimental investigations have displayed a rather varied behavior of the time spectrum of the annihilation radiation in a wide range of materials.

In metals only one lifetime is observed and this is rather insensitive to the type of metal under study. The magnitude of the lifetime stays in the range from $1\text{-}3 \times 10^{-10}$ seconds even though the electron densities may vary considerably from one metal to another.

Recently Bell has found a long component of very low intensity in some metals. In the case of ionic and valence crystals the results were similar. Here again, very recent measurements by Bisi et al.[9] have displayed a long component of low intensity.

The situation is not at all analogous in many organic amorphous and molecular materials which have been studied. These substances reveal a complex time spectrum. One observes a short lifetime of the order of 10^{-10} seconds, and a longer one of about $1\text{-}3 \times 10^{-9}$ seconds. The fast rate is in reasonable agreement with the theoretically predicted value and is not sensitive to the environment. However in the case of the longer lifetime there is considerable deviation from the theoretical prognosis for triplet positronium in vacuum. Furthermore the magnitude of the long component is strongly dependent on temperature and pressure as well as on externally applied magnetic fields. At points of first order phase transitions a sharp change is found to occur in the long lifetime.

The fractional amount of positrons annihilating from the triplet state, the so called I_2, is also found to be sensitive to the environment. It decreases when a magnetic field is applied and when paramagnetic atoms or ions are added in small concentrations to the substance under study. An example of this latter effect is shown by Deutsch[10], who added small amounts of NO and O_2 to his samples.

Similarly, in angular correlation measurements, where momentum distributions of the two photon annihilation process are investigated, two distinct momentum groups are often found. One is a narrow component which corresponds to low values of angular momentum; the other is a wider distribution and is due to pairs annihilating with higher angular momenta. Page et al.[11] have shown that the appearance of the narrow component is probably associated with the formation of positronium. Ferrell[12] has pointed out that pick-off annihilation is unlikely to fall into the low momentum region and hence the narrow distribution is likely due to annihilations from the singlet state.

On this basis the angular correlation bears out the quenching effect which was observed in the studies of the time spectra.

It should be noted here that the self-annihilation rate of positronium in the triplet state is proportional to the amount of overlap of the wave functions of the positron and electron which form the bound pair. This overlap is not sufficiently sensitive to the temperature and pressure to account for the strong dependence on these parameters. Therefore, if we are to follow up the assumption that positronium is formed, we are forced to consider auxiliary mechanisms in order to account for the strong influence of electron density variations and magnetic fields.

One such mechanism is the "pick-off" process whereby the bound positron annihilates with an atomic electron. The probability of such a process will be directly proportional to the amount of overlap of the wave functions of the positron and the atomic electron and hence will be a function of the electron density in the immediate vicinity of the positronium atom. This will be sensitive to the pressure and temperature of the sample and therefore the pick-off process could be effective in shortening the long lifetime. The short lifetime should then also be affected but this effect will be lost in experimental uncertainty since the self-annihilation from the singlet state is itself highly probable.

Another possibility was examined by Ferrell[12]. It concerns the triplet to singlet state conversion with the aid of a magnetic field, either molecular or microscopic in nature.

It further appears feasible that a positron compound is formed in some cases. Several groups have attacked this problem from the theoretical standpoint. Wheeler et al.[13] have examined the halogen chlorides. Neamtan et al.[14] have considered the formation of H^-e^+ bonds. Bisi et al.[9] have very recently shown that LiCl, CsCl, and LiH exhibit a complex decay scheme. The case of LiH shows a long lifetime of 1.5×10^{-9} seconds with an intensity of 6-7% and a short lifetime of 2×10^{-10} seconds.

In the case of these materials the short lifetime is regarded as being due to annihilations of free positrons while the long component is thought to emanate from the annihilation of positrons from the bound system $Li(H^-e^+)$. Again a density dependence is observed and to first order this is a linear relationship according to Bisi et al. Further evidence for positron compound formation was indicated by Lundholm et al.[15]

Another form of binding was inferred by Ferrell[12] from the following evidence:

On addition of diphenyl-picryl hydrazal, a negative free radical, the τ_2 value was reduced whereas the I_2 stayed constant within experimental error. In angular correlation measurements DeZafra[16] noticed a broadening of the distribution without the formation of a narrow component. These observations could be reconciled by assuming that positronium atoms were bound to the D.P.H. molecule or radical.

CHAPTER II

APPARATUS

In these measurements Na^{22} sources were employed. When Na^{22} decays with the emission of a positron, Ne^{22} is formed in an excited state and undergoes a transition resulting in the emission of a 1.28 MeV gamma ray. The time lapse between the appearance of the 1.28 MeV gamma and the positron is too short to be measured with the apparatus of this experiment, hence the detection of the gamma ray is taken as the reference point in the lifetime measurements.

The annihilation of the positron with an electron is accompanied by two 0.511 MeV gamma rays correlated at 180 degrees. The lifetime of the positron is determined by measuring the time lapse between the advent of the 1.28 MeV gamma and that of the 0.511 MeV gamma.

The mechanics of the measurements are as follows: Na^{22} sources were made by evaporating aqueous sodium chloride solution of high specific activity (2 mc/mg) on thin mica (5 mg/cm^2) sheets. These sources were then immersed in glass test tubes containing a few cubic centimeters of the particular substance under investigation. This source assemblage was then placed into the temperature regulation equipment which consisted in some cases of a furnace and in others of a cryostat or simply a thermos bottle. The detectors were then brought as close as possible to the location of the source as conditions permitted.

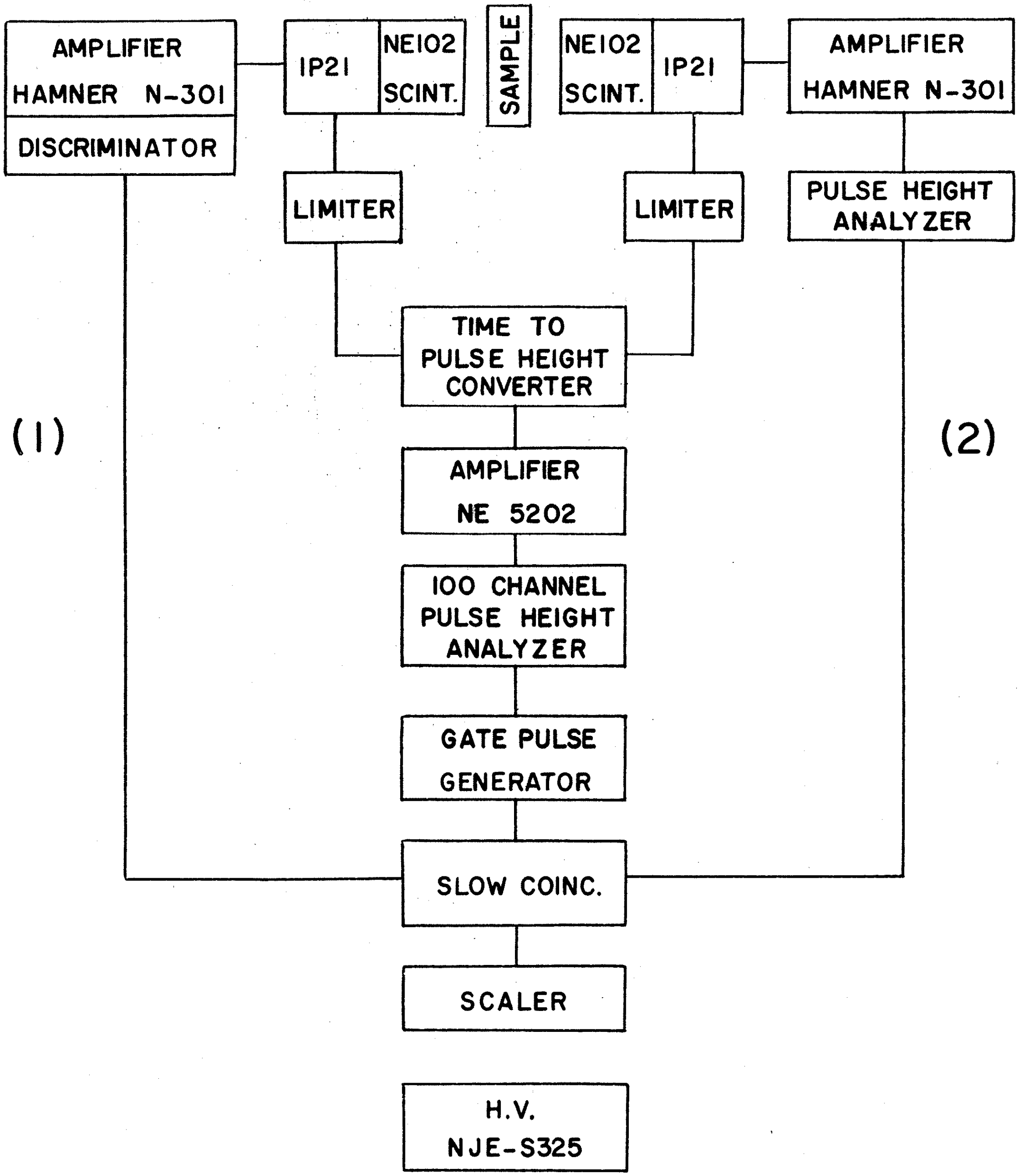
The schematic diagram of the electronic apparatus is shown in Figure 1.

Cylindrical plastic phosphours (NE102), optically coupled to IP21 RCA photomultipliers served as detectors. On detection of gamma rays these photomultipliers deliver negative pulses of the order of ten volts. These pulses are then employed to trigger limiter circuits which are designed to put out fast rising, flat topped pulses. The rise time of the limiter pulse is about one nanosecond and the length of the pulse approximately two microseconds. These flat pulses are chopped to a length of 32 nanoseconds by shorting stubs and are subsequently fed to a time-to-pulseheight converter. The latter is designed to produce an output pulse whose height is directly proportional to the time interval during which two input pulses overlap. This is accomplished by directing the pulses from the two limiters onto two separate control grids of a 6BN6 pentode which is biased so as to give an output pulse only when both grids are activated simultaneously. The resulting pulse then travels to the control grid of a 5842 triode via an RC network. The RC network has an exponential voltage versus time response, however, only the closely linear initial section of this is employed. Hence the output of the triode becomes directly proportional to the time during which the 6BN6 conducts, that is, to the time during which the limiter pulses overlap.

The triode output is then amplified and fed to a Victoreen

FIG. I

BLOCK DIAGRAM OF APPARATUS



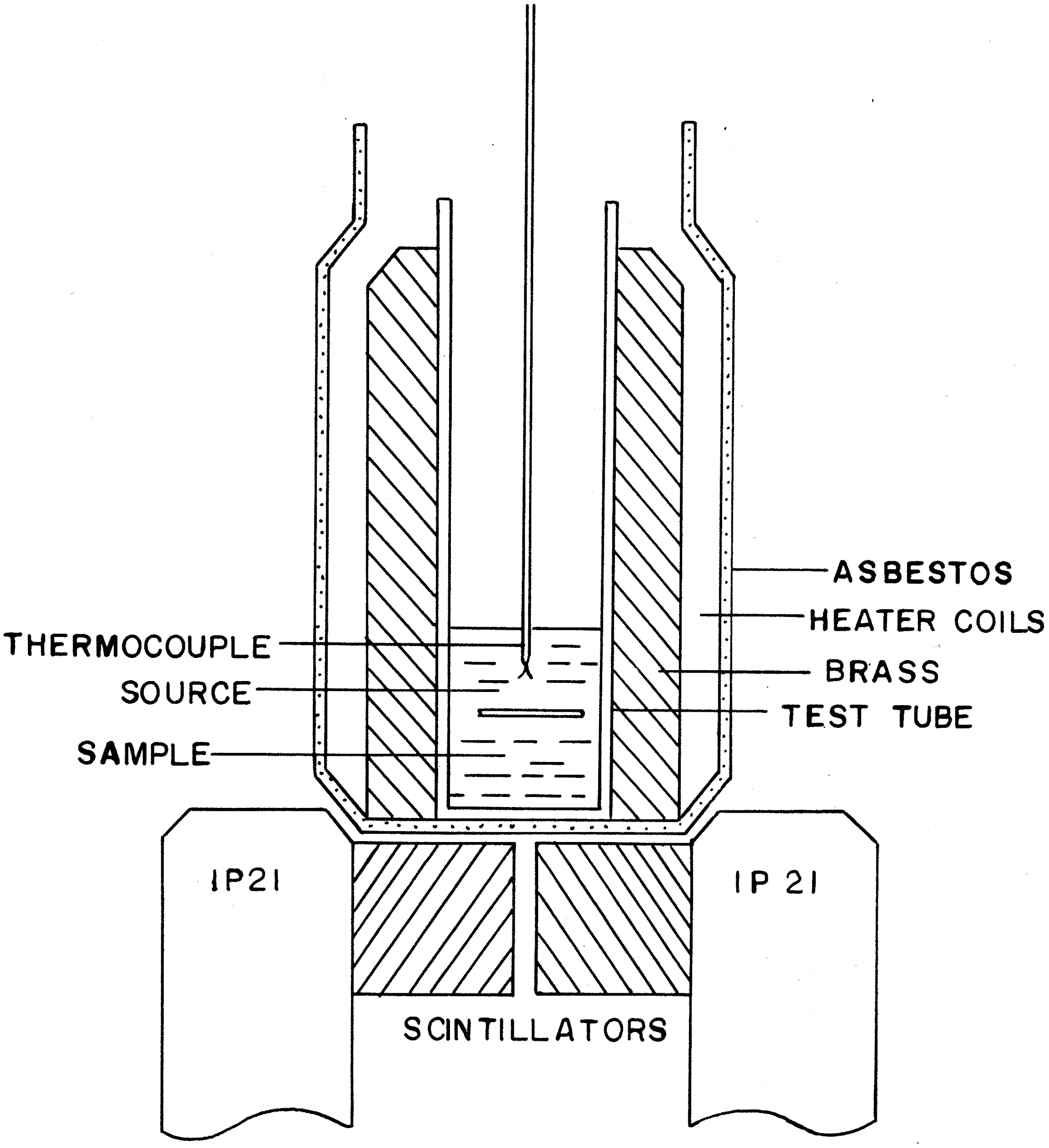
pulseheight analyser. The latter is gated so as to accept only those pulses which correspond to a combination of a 1.28 MeV gamma and a 0.511 MeV gamma.

Appropriate gating is achieved in the following manner: The side channels shown in the schematic diagram of the apparatus are calibrated so that one will pass pulses emanating from the detection of a 1.28 MeV gamma ray, while the other passes those from a 0.511 MeV ray. A differential discriminator tags those photomultiplier pulses which correspond to energy dissipations of between 0.2 and 0.5 MeV as annihilation gammas, while an integral discriminator passes as 1.28 MeV events those pulses which imply an energy loss of more than 0.5 MeV in the phosphors.

FURNACE

For measurements above 24 degrees centigrade the temperature was controlled by an arrangement shown in Figure 2. The sample tube was held inside an electrically heated brass sleeve which was encased by an asbestos jacket. The entire set-up was then again surrounded by an insulating jacket to counteract the influence of drafts due to the air conditioning unit. Temperature measurements were made with a copper-constantan thermocouple or by a mercury thermometer mounted inside the brass sleeve. The variation in the temperature was kept within one degree centigrade.

FIG·2


ASBESTOS
HEATER COILS
THERMOCOUPLE
BRASS
SOURCE
TEST TUBE
SAMPLE
1P21
1P 21
SCINTILLATORS

CHAPTER III

CALIBRATION OF THE APPARATUS AND INTERPRETATION OF RESULTS

1. Calibration

The side channels were calibrated employing a Cs^{137} source which decays with the emission of a β and is followed by a 0.662 MeV gamma ray. The differential discriminator level was adjusted until the counting rate was reduced to one count per second with the gate opened fully. Then assuming a linear energy response for the discriminator the level settings were calculated for 0.2 and 0.5 MeV pulses.

In a similar manner the level for the discriminator in the other side channel was set to allow 1.28 MeV pulses to pass.

The time response of the apparatus was then determined utilizing a Co^{60} source. Co^{60} has two gammas in cascade in its decay scheme which can be used to simulate prompt coincidences since the time lapse between the two cannot be resolved by the apparatus of this experiment.

The pulses from one of the limiters are artificially delayed by inserting a measured length of RG7U cable between the limiter and the time to pulse height converter. The pulse transmission speed of RG7U cable is 0.84c; a measured length of cable will therefore introduce a known time delay between the pulses coming from the two limiters.

A prompt resolution curve was obtained for a set of delay values ranging from 3 to 14 nanoseconds in steps of one nanosecond. The channel number of the centroid of each resolution curve was plotted as a function of the delay. The slope of the resulting straight line was then taken as a measure of the sensitivity of the apparatus. Its value was typically of the order of 0.200 nanoseconds per channel.

The resolution of the apparatus was obtained by multiplying the full width at half height by the sensitivity.

Figure 3 shows a typical prompt Co^{60} curve and Figure 4 a calibration graph.

2. Interpretation of the Data

The duration of the runs varied between twelve and thirty-six hours depending on the geometry of the setup. Generally there were upward of ten thousand counts in the centroid channel.

The time spectrum at all times occupied less than the first sixty channels of the 100 channel analyser section. The background counting rate was approximated by averaging the number of counts per channel in the first 20 channels following the end of the time spectrum.

After subtracting the background the data was analysed according to the method of Green and Bell[18]. The spectrum was plotted on semi logarithmic graphpaper and a straight line was fitted to the tail of the data either by eye or by applying least squares analysis.

FIG. 3

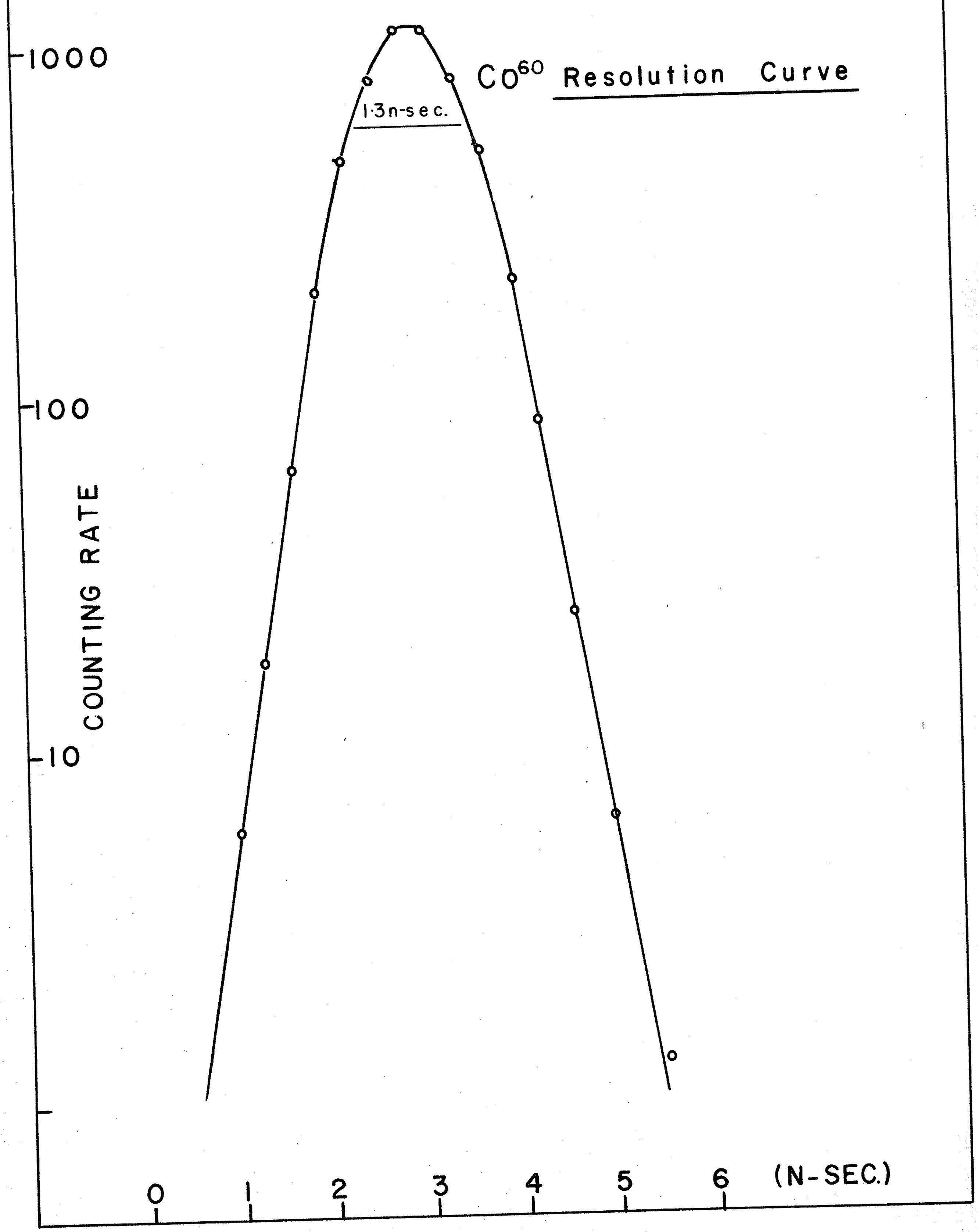
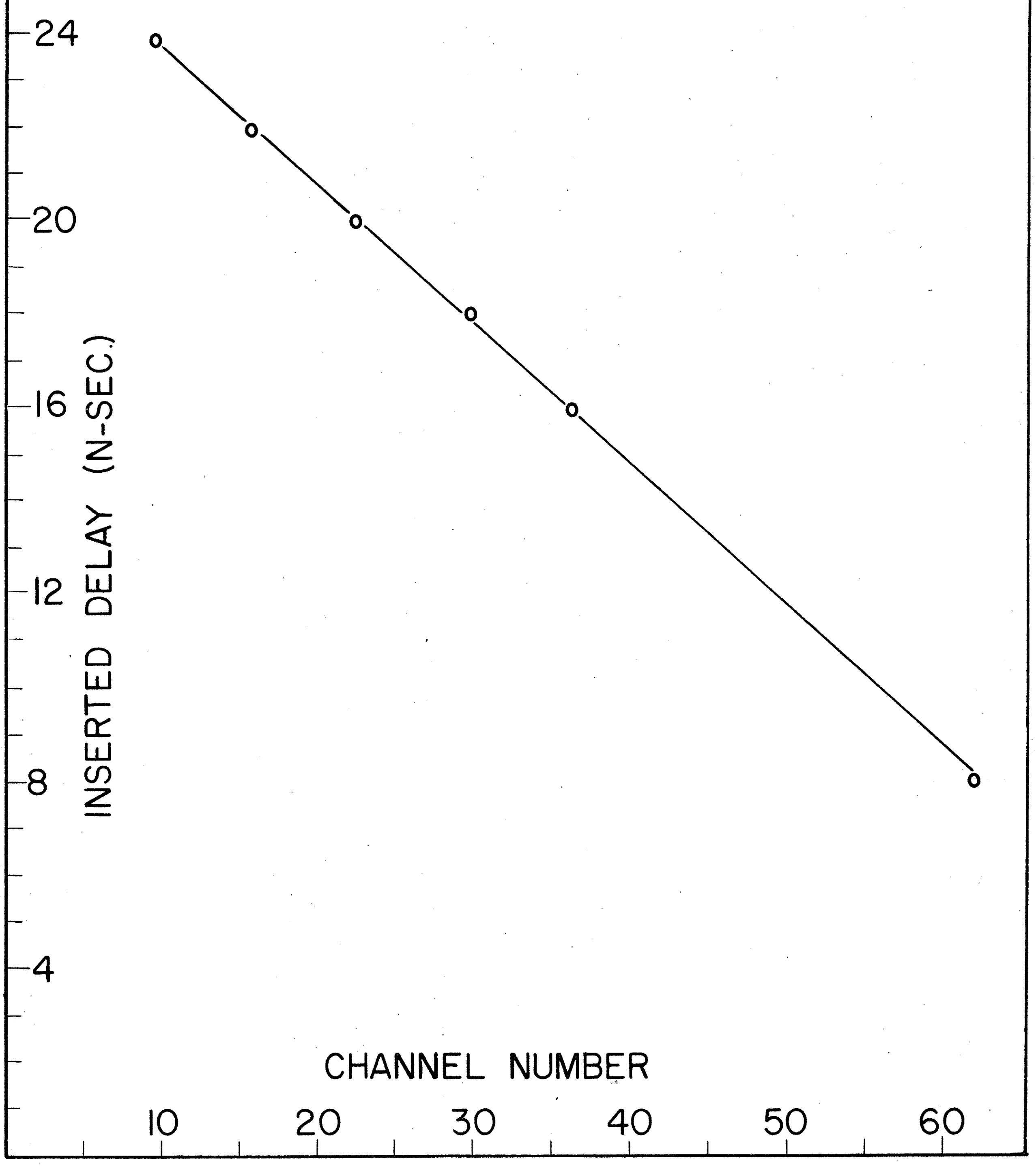

1000
100
10
COUNTING RATE
Co^{60} Resolution Curve
1·3n-sec.
0
1
2
3
4
5
6
(N-SEC.)

FIG. 4

TIME TO PULSE HEIGHT CONVERSION



The τ_2 value is then given by:

$$\tau_2 = \frac{0.434}{\text{slope}}$$

where in the case of least squares fits, the slope is given by:

$$\frac{\frac{1}{n}\sum x_i \sum y_i - \sum x_i\, y_i}{\left(\sum x_i^2 - \frac{1}{n}\left(\sum x_i\right)^2\right)}$$

with

x_i = time delay of the i^{th} point.

y_i = log (base 10) of the number of counts at the i^{th} point.

n = the number of points used in the analysis.

The intensity of the long lived component, I_2, was calculated by taking the ratio of the area under the tail (extended to the position of the centroid) to the area under the total curve.

CHAPTER IV

DISCUSSION OF RESULTS

I. Diphenylamine

This substance has the chemical formula $C_{12}H_{11}N$. Its melting point is at 54 degrees centigrade and it has a specific volume change of 99 cc/kg at the liquid solid transition.

Typical time spectra of the annihilation radiation of positrons in this substance are given on page 21 and the behavior of the τ_2 value as a function of the temperature is displayed on page 24. The latter graph indicates that the τ_2 value changes only very slowly with variations in the temperature if the phase remains the same. The values of τ_2 on either side of the transition temperature differ considerably. This difference is estimated as 1.1 ± .2 nanoseconds.

The point at 54 degrees centigrade corresponds to a run with the diphenylamine sample in a mixture of both the solid and the liquid phase.

II. P-Bromotoluene

The chemical composition of this substance is described by the formula C_7H_7Br. It undergoes a first order phase transition at 24.8 degrees centigrade with an accompanying specific volume change of 88 cc/kg.

Again, within experimental uncertainty the value of τ_2 remains almost constant in a given phase, but as in the case of diphenylamine there is a sharp jump in the immediate vicinity of the transitions temperature region. The magnitude of the change is here estimated to be $0.6 \pm .1$ nanoseconds.

III. N-Phenylbenzylamine

The chemical composition is $C_{13}H_{13}N$; the liquid-solid transition is accompanied by a specific volume change of 76 cc/kg.

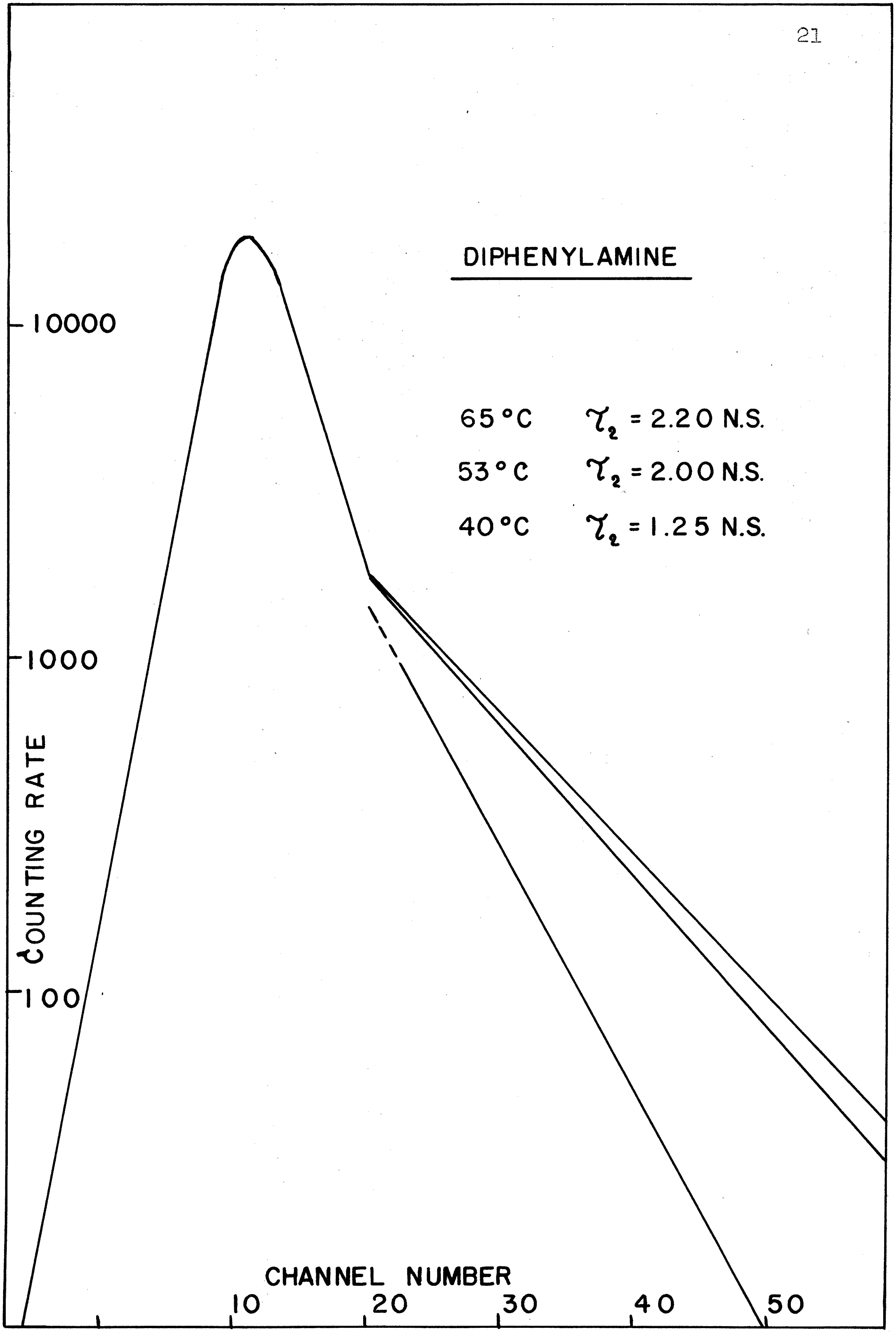
As can be seen in the graph on page 26 the τ_2 behaves in a manner analogous to the earlier cases. Again only a slow variation in τ_2 is observed over a range of temperatures at constant phase. The phase was observed to change at 23 ± 2 degrees centigrade and a jump of $0.75 \pm .2$ nanoseconds is noted in this region.

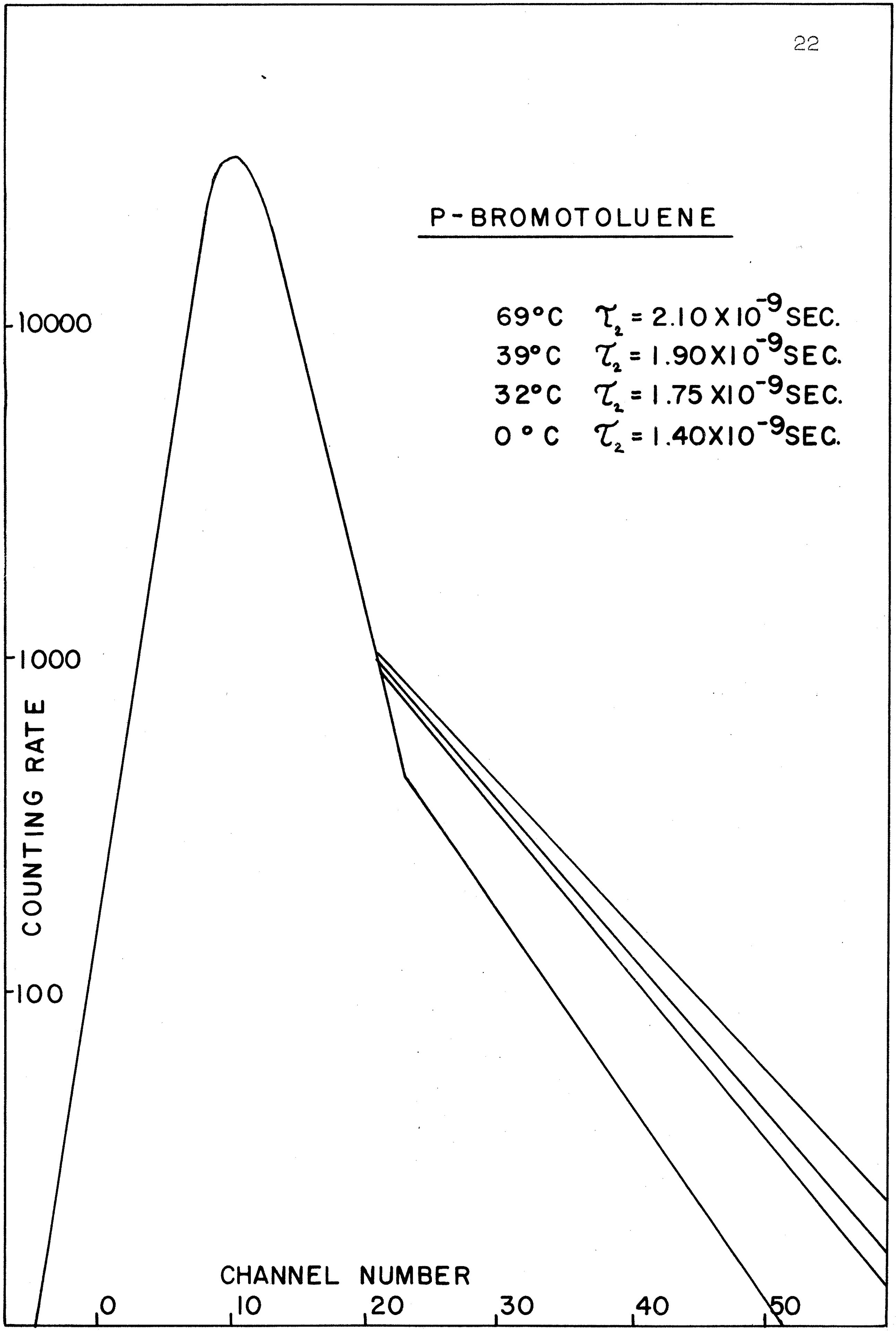
This behavior of τ_2 at first order phase transition points has been observed by others, among them H. Clarke[18], although for the cases of cyclohexane and phenol no definite change in τ_2 could be associated with phase changes. Both these latter two substances have specific volume changes of less than 60 cc/kg.

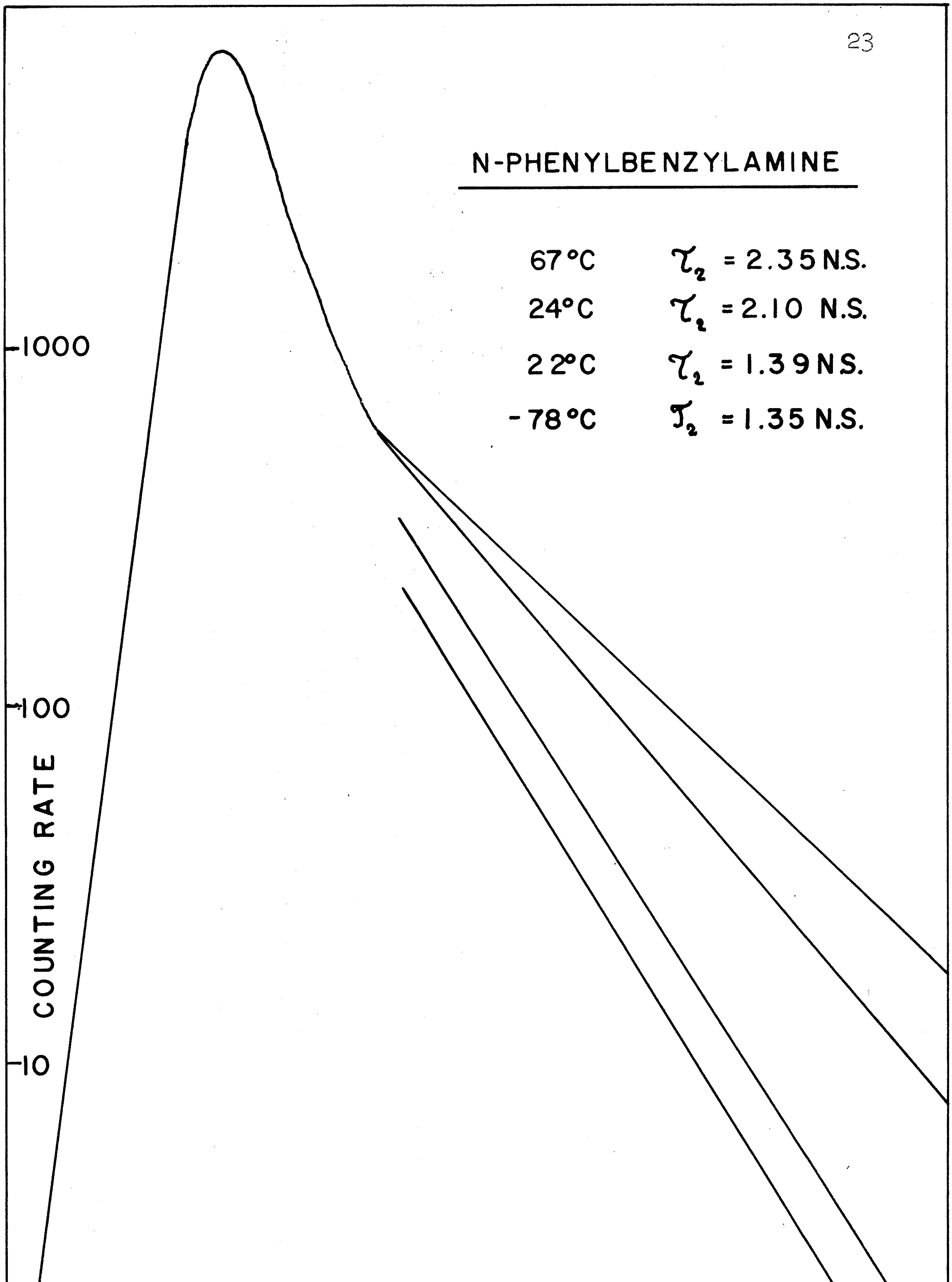
In general this type of behavior of the long lifetime component can be well reconciled with the arguments advanced on behalf of positronium. As was mentioned in Chapter I, the pick-off process is expected to be of major importance in shortening the long lifetime of positronium. Furthermore, the effectiveness of the pick-off process is dependent on the density of the atomic electrons in the vicinity of the positronium atom; this density will most certainly be determined by the proximity of the neighbouring molecules. Hence the change in specific volume at phase transitions will be related to the effectiveness of the pick-off process.

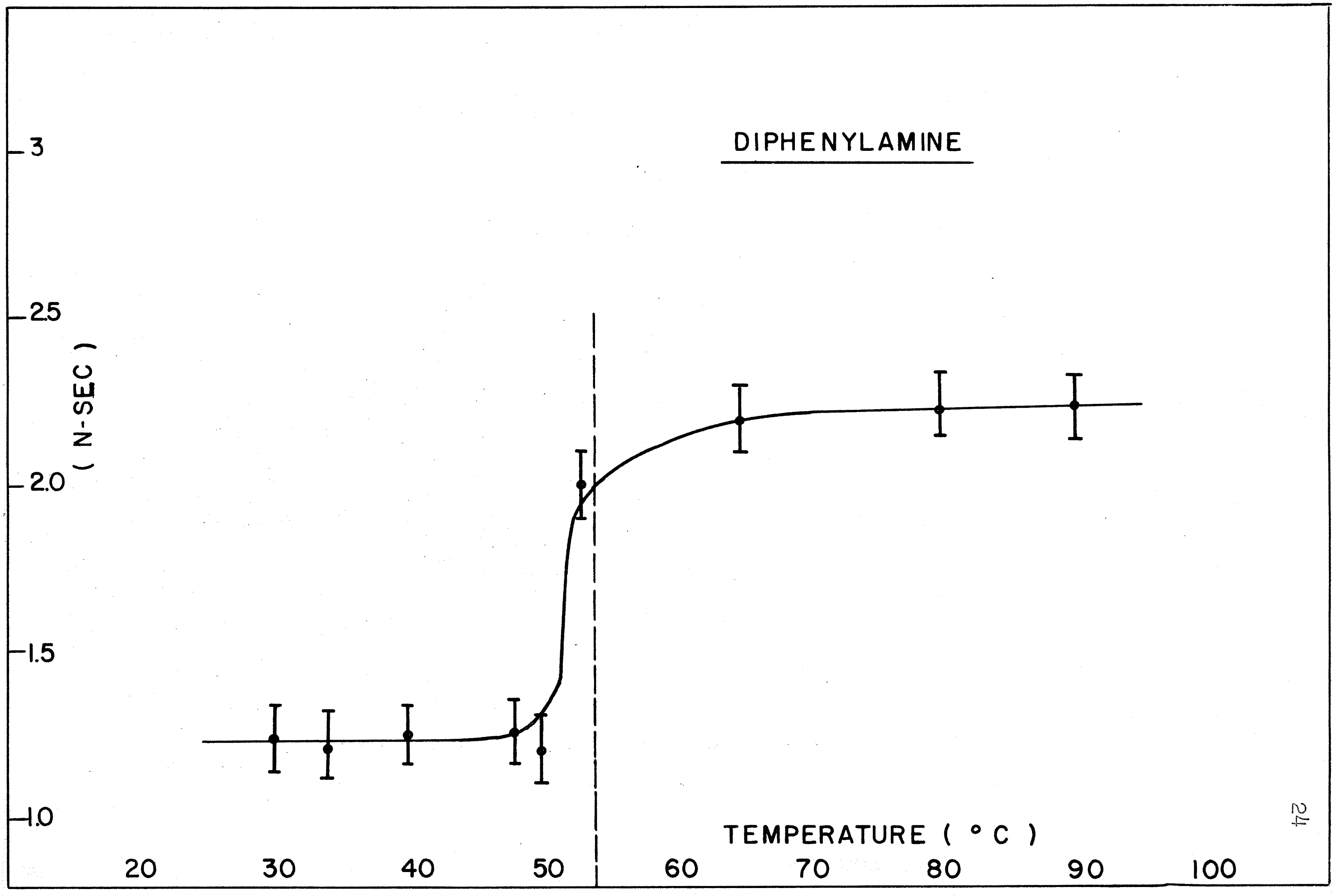
If the pick-off process were the sole mechanism active in defining the long lifetime, one would expect a smooth functional relationship to exist between the change in τ_2 at phase transition points and the associated change in specific volume. The jump in τ_2 at first order phase changes would be expected to increase with increasing change in the specific volume. The curve should settle to a value of approximately 10^{-7} seconds for $\Delta\tau_2$ at very large values of Δv since this would simulate the transition of a positronium atom in a dense medium to positronium in vacuum.

The data on page 32 bears out this behavior only very roughly. This would appear to be a good indication that the pick-off process is challenged in importance by other factors, such as molecular structure for example.


DIPHENYLAMINE
65 °C τ_2 = 2.20 N.S.
53 °C τ_2 = 2.00 N.S.
40 °C τ_2 = 1.25 N.S.
10000
1000
100
COUNTING RATE
CHANNEL NUMBER
10
20
30
40
50


P-BROMOTOLUENE
69°C $\tau_2 = 2.10 \times 10^{-9}$ SEC.
39°C $\tau_2 = 1.90 \times 10^{-9}$ SEC.
32°C $\tau_2 = 1.75 \times 10^{-9}$ SEC.
0 °C $\tau_2 = 1.40 \times 10^{-9}$ SEC.
10000
1000
100
COUNTING RATE
CHANNEL NUMBER
0
10
20
30
40
50


N-PHENYLBENZYLAMINE
67°C τ_2 = 2.35 N.S.
24°C τ_2 = 2.10 N.S.
22°C τ_2 = 1.39 N.S.
-78°C τ_2 = 1.35 N.S.
1000
100
10
COUNTING RATE
CHANNEL NUMBER
1
10
20
30
40
50


DIPHENYLAMINE
(N-SEC)
3
2.5
2.0
1.5
1.0
TEMPERATURE (° C)
20
30
40
50
60
70
80
90
100

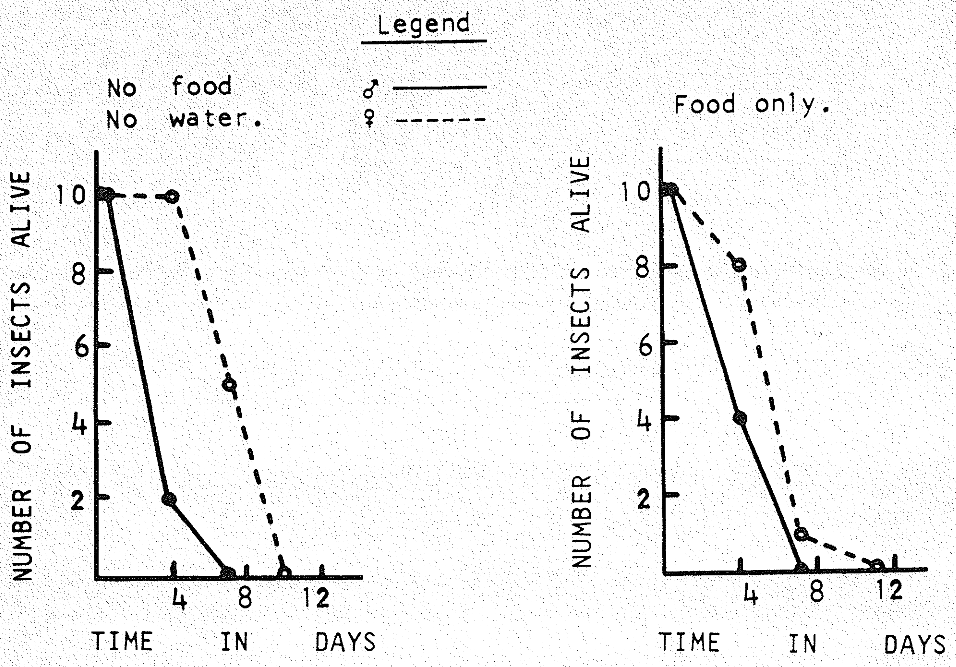


FIGURE 9 a.

FIGURE 9 b.

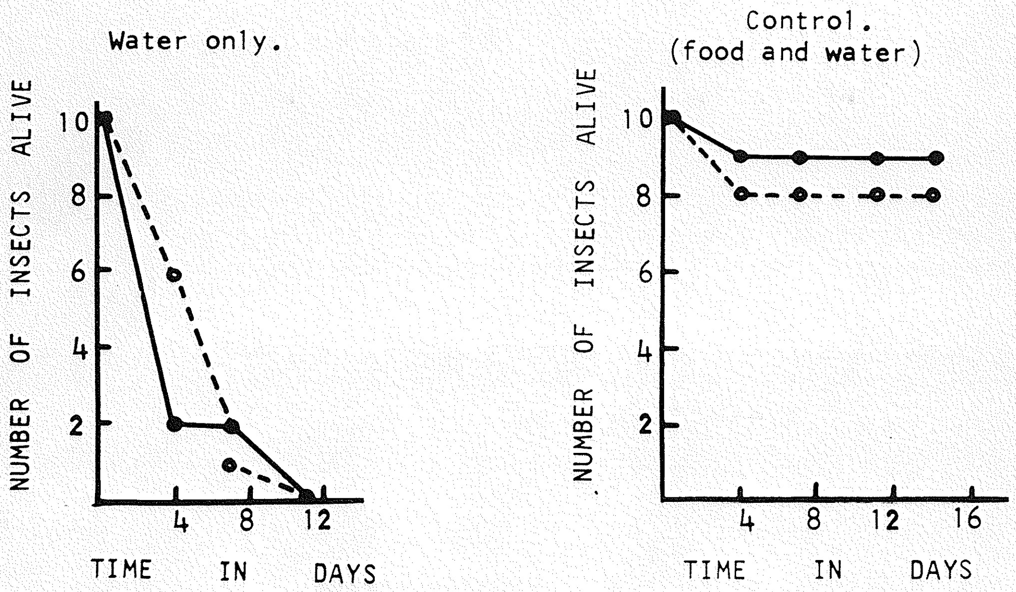
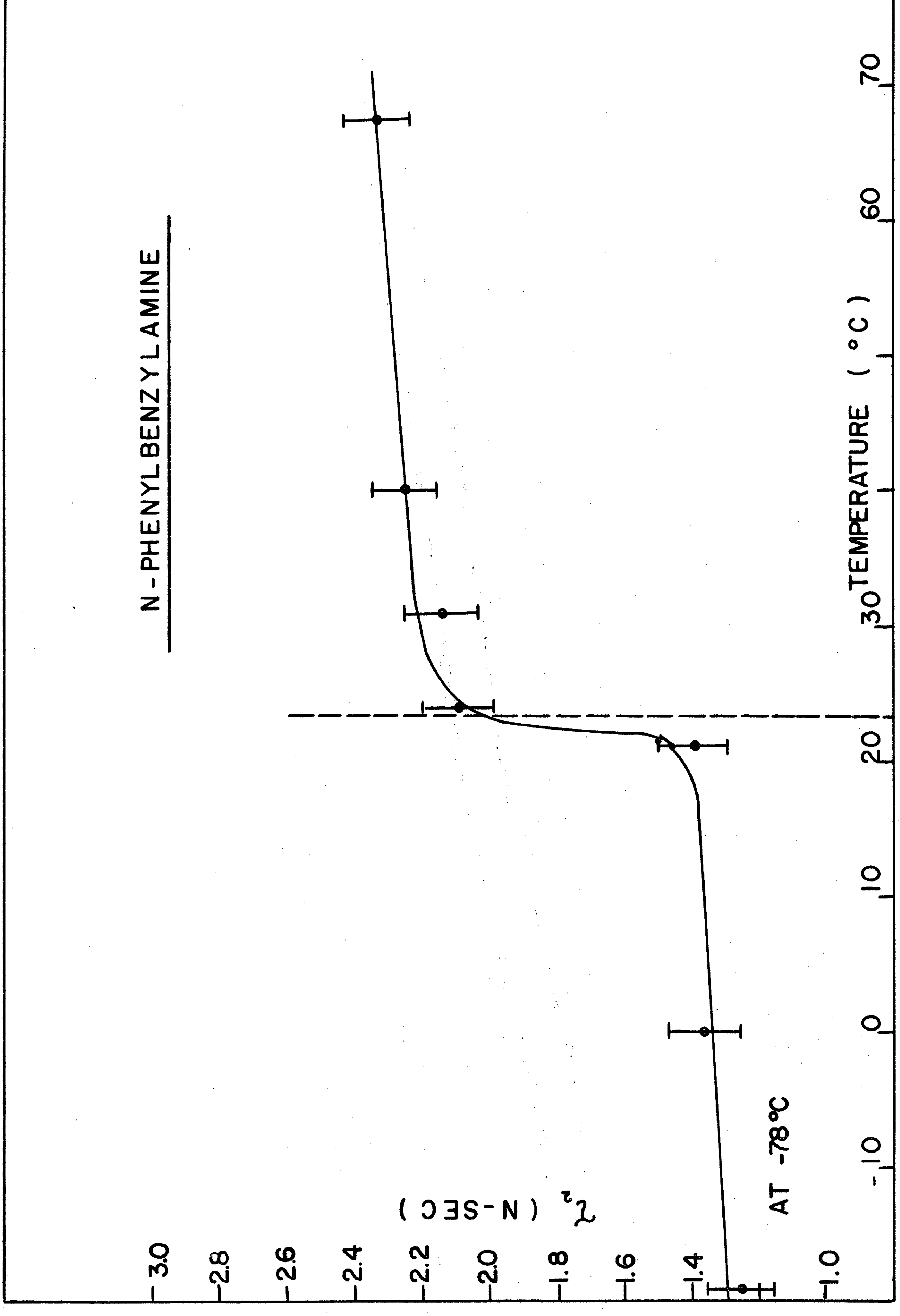


FIGURE 9 c.

FIGURE 9 d.

FIGURE 9

THE SURVIVAL OF SECOND INSTAR NYMPHS OF _Blattella germanica_ (Linn.) STARVED AT 26.7°C AND 30% R.H.


N-PHENYLBENZYLAMINE
τ_2 (N-SEC)
3.0
2.8
2.6
2.4
2.2
2.0
1.8
1.6
1.4
1.0
AT -78°C
-10
0
10
20
30
TEMPERATURE (°C)
60
70

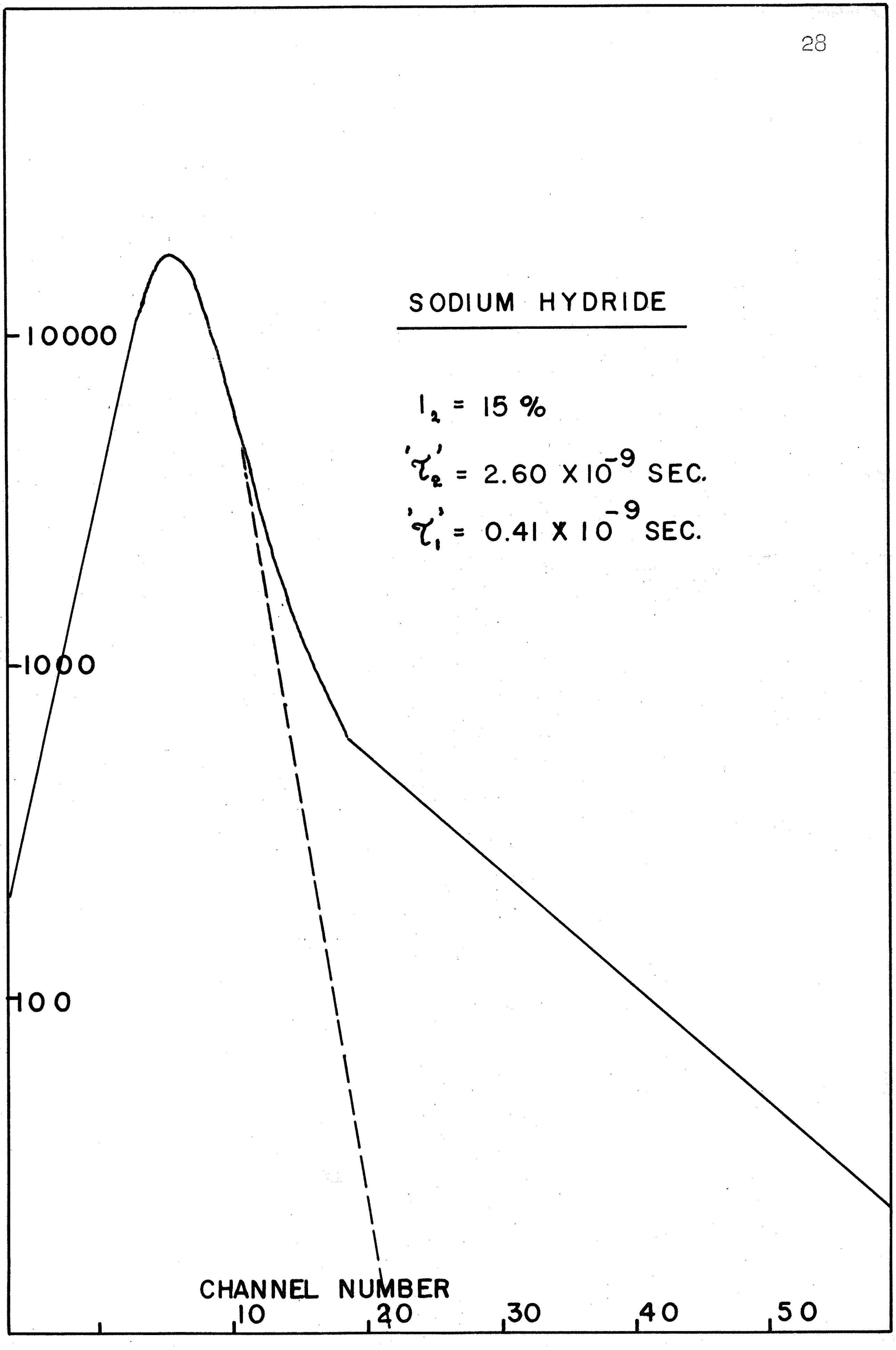
NaH AND LiH

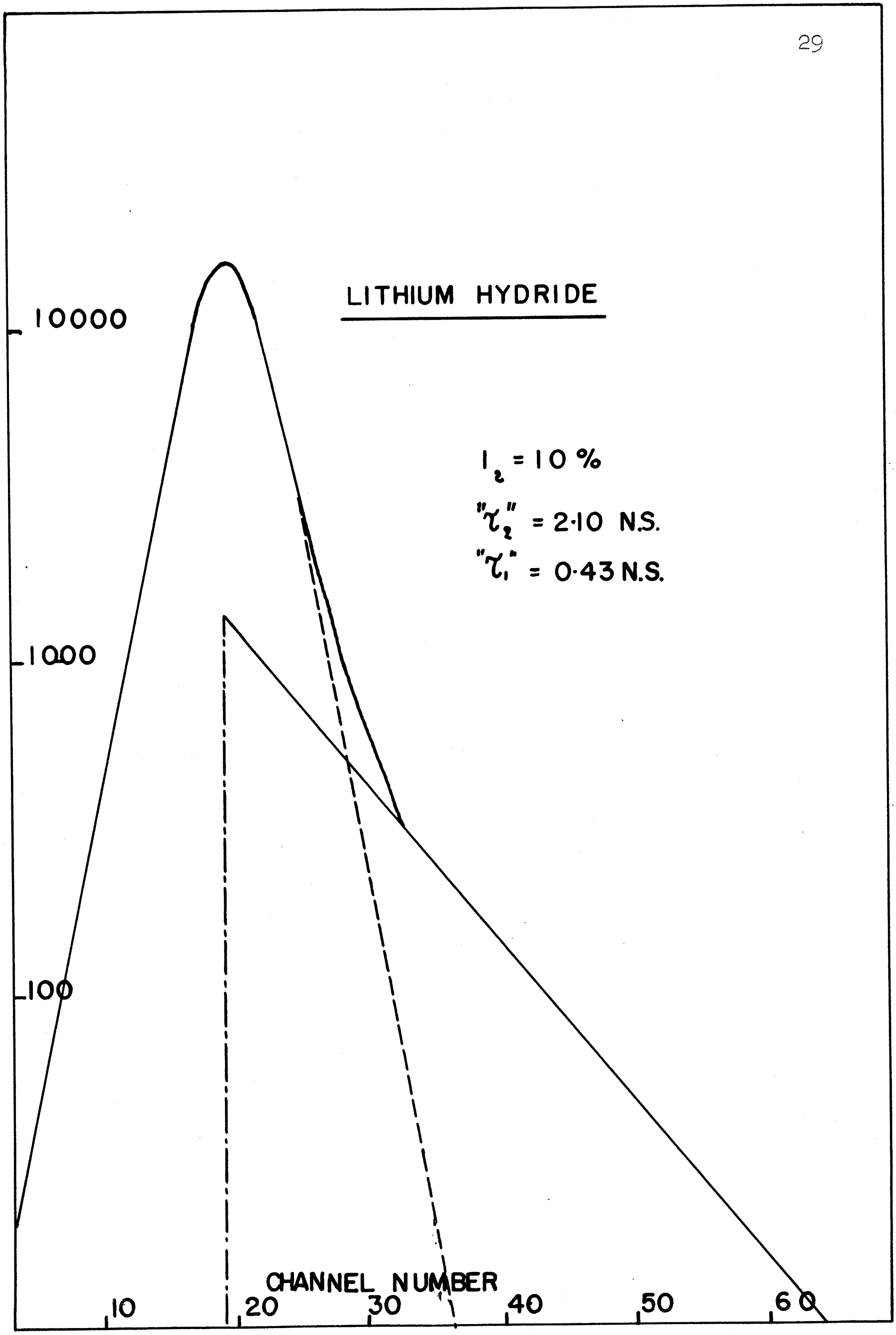
Both substances form ionic crystals and represent the unusual situation where the hydrogen ion is negative. The samples employed in this experiment were of a finely powdered form with a density of .75 gm/cc and .45 gm/cc for NaH and LiH respectively. The single crystal form displays densities of 1.3 gm/cc and 0.8 gm/cc respectively.

In each case the time spectrum was observed to be complex. A long lifetime component was found to be 2.6 ± .3 nanoseconds for NaH and 2.1 ± .3 nanoseconds for LiH, with intensities of 15% and 10% respectively. The short lifetime component was determined in each case by subtracting the tail due to the long component and then measuring the slope of the remaining tail. The values obtained in this manner were 0.41 ± .1 nanoseconds for NaH and 0.43 ± .1 nanoseconds for LiH. The errors here are relatively large since the slope of the tail approaches the natural slope of the prompt coincidence curve.

Some difficulty appears in the interpretation of these results. According to R. A. Ferrell[4] it is unlikely that positronium is formed in ionic crystals since the existence of the Ore gap in these substances is not probable.

If this argument is valid then one is forced to consider other processes in the case of LiH and NaH in order to account for the existence of the complex time spectrum.


SODIUM HYDRIDE
I_2 = 15 %
$'\tau_2' = 2.60 \times 10^{-9}$ SEC.
$'\tau_1' = 0.41 \times 10^{-9}$ SEC.
10000
1000
100
CHANNEL NUMBER
10
20
30
40
50


LITHIUM HYDRIDE
$I_2 = 10\%$
"τ_2" = 2·10 N.S.
"τ_1" = 0·43 N.S.
10000
1000
100
CHANNEL NUMBER
10
20
30
40
50
60

It is noteworthy that the short lifetime of 0.41 nanoseconds in LiH agrees within the experimental uncertainty with the value of the lifetime of the bound state Li (H^-e^+) which was calculated by Neamtan et al.[14] If this represents a valid correlation then one is left with a long lifetime component which requires an interpretation. Again it is possible that triplet positronium is formed in small amounts. This would not refute the argument for the formation of a positron compound as discussed by Neamtan et al. since the singlet lifetime might well be of the same order of magnitude as that of the bound state H^-e^+, and the two may not be separable.

Bisi et al.[9] (prepublication) have chosen to interpret the short lifetime which in their experiment with single crystal LiH was found to be 0.2 nanoseconds as relating to positrons annihilating from the free state. They then associate the long component with the annihilation from the bound state H^-e^+. They point out the difficulty in reconciling the experimental and theoretical values of the lifetime.

It can be argued that if the short lifetime is due to annihilations from the free state then the disagreement between the values of Bisi et al. and this work may be caused by differences in mean lattice point densities in the two samples. The solid crystal density is 0.8 gm/cc while the density of the powdered sample was .45 gm/cc. Hence the ratio of the lifetimes should be ~ 2. Within experimental error this is borne out.

We are hence not in a position to decide on the type of process(es) involved in the annihilation of positrons which enter these ionic crystals.

TABLE I

SUMMARY OF RESULTS

Sample	Temp. (°C)	τ_2 (nanoseconds)
Diphenylamine	29	1.20 ± .1
"	33	1.18 ± .1
"	40	1.25 ± .1
"	48	1.27 ± .1
"	50	1.17 ± .1
"	53	2.00 ± .1
"	65	2.20 ± .1
"	80	2.25 ± .1
"	92	2.30 ± .1
P-Bromotoluene	-78	1.25 ± .1
"	0	1.40 ± .1
"	32	1.75 ± .1
"	39	1.90 ± .1
"	52	1.94 ± .1
"	69	2.10 ± .1
N-Phenylbenzylamine	-78	1.25 ± .1
"	0	1.35 ± .1
"	22	1.39 ± .1
"	24	2.10 ± .1
"	32	2.15 ± .1
"	40	2.25 ± .1
"	67	2.35 ± .1

TABLE I (Cont'd)

Sample	Temp. (°C)	τ_1 (ns)	τ_2 (ns)	I_2
NaH	24	$.4 \pm .1$	$2.6 \pm .5$	15%
LiH	24	$.4 \pm .1$	$2.1 \pm .7$	10%

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