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FACULTY OF GRADUATE STUDIES

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DOCTOR OF PHILOSOPHY

of

WILLIE ROBERT FALK

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THE ANNIHILATION OF POSITRONS IN GASES

ABSTRACT

The lifetimes of 'free' positrons have been investigated (at room temperature) in the gases N₂, O₂, CO₂, He, Ar, and Kr by recording the time distributions of the annihilation events. Whereas all the polyatomic gases exhibited pure exponential decay spectra, the annihilation spectra for the noble gases were more complex exhibiting an initial 'shoulder' followed by an exponential decay. An analysis of the results suggests that in all cases the exponential decay corresponds to the annihilation of positrons in thermal equilibrium with the gas molecules. The application of a DC electric field was found to produce marked changes in the shape of the annihilation spectra in argon.

The annihilation spectrum in argon (at 10.5 atm) was investigated in detail. At zero electric field the initial portion of the spectrum is characterized by a relatively low non-constant annihilation rate, and is attributed to annihilation of positrons which are slowing down in the energy interval between the positronium formation threshold (8.9 eV in argon) and thermal energy. In this energy interval the positron can lose energy only by making elastic collisions with the atoms of the gas. A velocity-dependent annihilation rate over this energy range is suggested to account for this 'shoulder' in the annihilation spectrum. The time-width of this shoulder is found to be 340 nsec-atm. The rest of the spectrum exhibits an exponential decay as observed in the polyatomic gases.

Further investigations using applied DC electric fields (in the range between 0 and 122 V cm⁻¹ atm⁻¹) revealed an increase in the mean lifetime of the 'free' positrons with increasing electric field. For a field of 790 V/cm and a pressure of 10.5 atm this lifetime has increased by a factor of 2.

A diffusion equation governing the behavior of positrons in the noble gases has been derived and solved numerically for different cases of the elastic scattering cross section, the direct annihilation cross section, and the positronium formation cross section. A good fit to the experimental data for argon was obtained using the values $20(v_o/v)\tau a_o^2$.
$2.25 \times 10^{-4}(v_0/v)^{1.5} r a_0^2$ and $5.0 \times 10^{-5} r a_0^2$ respectively for these cross sections, where $v$ is the velocity of the positron and $v_0$ its velocity at thermal energies ($25^\circ C$). The form of the positronium formation cross section was taken as a step-function whose magnitude was adjusted to yield results consistent with the increase in positronium formation measured by Marder et al (1956). Further calculations are required in order to test the uniqueness of this set of cross sections.

Similar experiments in helium revealed only slight evidence for the presence of a shoulder and only a small dependence of the mean lifetime on the applied electric field. This suggests that the annihilation cross section is closely approximated by an inverse velocity dependence.

Preliminary investigations in krypton indicate a 'shoulder' width about one half as great as in argon.

A comparison of these results to those obtained from the Dirac cross section (assuming all the electrons of the atom participate) reveals that the observed rates in N$_2$, O$_2$, CO$_2$ (at low pressures), He, A, and Kr are larger than the Dirac values by factors of 2.14, 1.59, 2.64, 1.91, 1.63, and 1.67 respectively.

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Related Studies:

Functions of a Complex Variable W.H. Simons
PUBLICATIONS


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by

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A THESIS SUBMITTED IN PARTIAL FULFILMENT OF
THE REQUIREMENTS FOR THE DEGREE OF

DOCTOR OF PHILOSOPHY

in the Department
of

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We accept this thesis as conforming to the
required standard

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March 1965
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Date March 25, 1965.
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Preliminary investigations in krypton indicates a 'shoulder' width about $1/2$ as great as in argon.

The annihilation rate of the 'free' positrons (in the exponential region) was found to be directly proportional to the gas density for all the gases investigated except CO$_2$, where the annihilation rate can be represented by the equation

$$\lambda_a = (0.852 \varphi - 0.040 \varphi^2) \times 10^7/sec$$

where $\varphi$ is the density of the gas expressed in terms of the density at 1
atm and 25°C.

A comparison of these results to those obtained from the Dirac cross section (assuming all the electrons of the atom participate) reveals that the observed rates in N₂, O₂, CO₂ (at low pressures), He, Ar, and Kr are larger than the Dirac values by factors of 2.14, 1.59, 2.64, 1.91, 1.63, and 1.67 respectively.
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ERRATA

Page 128 Line 20: Read "experiments using time sorter", instead of "experiments using time sorter experiments".

Page 131 Line 5: Read "conventional" instead of "conventional".
Line 18: Read "line" instead of "time".

Page 132 Line 17: Read "nonconducting" instead of "nonconducting".

Page 134 Line 9: Read "Triple" instead of "Trippe".
Line 16: Read "that" instead of "than".

Page 153 Line 20: Read "azimuthal" instead of "azimuthal".

Page 156 Equation (18): Last term of this equation should read
\[ (r_a(v) + r_f(v))f_0. \]
CHAPTER I

INTRODUCTION

In a review article on positronium published in 1954 by DeBenedetti and Corben, the authors say, in part:

"In these days when the attention of the physicist is concentrated on the study of queer particles and of interactions which escape our comprehension, it is somewhat refreshing to turn our minds towards a subject which does not lack of fundamental importance and about which we can claim a certain amount of understanding. Such a subject is the behavior of electrons and positrons and their interaction with electromagnetic radiation. In this article we consider a particular example in this field: the annihilation of positron-electron pairs and the formation of the quasi-stable atom of positronium. Although such studies cannot be said to have brought basic changes in our thinking, they have resulted in a well organized body of information which can be regarded with a sense of aesthetic satisfaction."

The discovery of the positron by Anderson (1932) was quickly followed by a series of experiments and investigations which verified that this was indeed the particle predicted by Dirac's relativistic quantum theory of the electron. Since those early experiments the understanding of the behavior of the positron and its interaction with material media has increased rapidly to the point where the annihilation of positrons has proved to be a useful tool for the investigations of the structure of matter itself. Such an example is the case of the experimental investigations of the Fermi surface in metals (Wallace (1960)). Despite this considerable body of information which has been compiled, it is still impossible to predict the detailed nature of the interaction of a positron with the electrons of an atom. The solution of this fundamental problem requires an extensive effort in both the experimental and theoretical fields. The work reported in this thesis is directed at a partial elucidation of
Soon after Anderson's discovery, Mohorovicic (1934) made the exciting suggestion of the possibility of the existence of a bound positron-electron system, later given the name "positronium" by Ruark (1945). The existence of such an atom was established by Deutsch (1951a and 1951b) who investigated the annihilation rate of positrons in various gases. Using this atom, there soon followed, what can be considered, one of the major achievements of theoretical and experimental physics - the verification of the theory of quantum electrodynamics to terms of order $\alpha^3$. This experiment, performed by Weinstein, Deutsch, and Brown (1954), measured the hyperfine splitting between the $m = \pm 1$ level and the $m = 0$ level of orthopositronium.

Thus, while much progress has been made in the understanding of the bound positron-electron system, the problem of the behavior of "free" positrons - those which do not form positronium - has, however, received little attention to date. What information can be obtained from a study of these "free" positrons? Firstly, a measurement of the annihilation rate of positrons can yield important information regarding the annihilation cross section for a positron with the atomic electrons of material media. Secondly, it should be possible to obtain information about the scattering cross sections for positrons by suitably designed experiments. Moussa (1959) and others have stressed the need for information of this latter type, in particular, the elastic scattering cross sections of low energy positrons from the monatomic gases. Theoretical efforts in this area have been restricted almost exclusively to the simplest cases of scattering from such gases, and experimental results for comparison with
the theory are urgently needed before further progress can be made.

In addition, an investigation of the behavior of 'free' positrons is essential to a better understanding of the mechanism of positronium formation.

The present state of technology in experimental physics makes such investigations particularly feasible in low density materials such as gases. Furthermore, great simplifications should result using gases, since it is expected that the interaction of the positron should be restricted to a single atom (or molecule) of the gas at any one time.

This thesis reports an investigation of the behavior of 'free' positrons in the polyatomic gases nitrogen, oxygen, and carbon dioxide, and in the monatomic gases helium, argon, and krypton. The experimental technique involved the recording of the time distributions of the annihilation events of positrons in these gases. Of the latter group of gases, argon was studied in great detail because of the interest generated by the recent discovery of a velocity-dependent annihilation rate in this gas by Tao et al (1954), Falk and Jones (1964), and Paul (1964). For the first time, experimental effects were observed which depended on the detailed nature of the elastic scattering of positrons from these atoms and on the annihilation cross section of the positrons.

A detailed program of research was undertaken designed to illuminate in greater detail the specific velocity dependence of both the elastic scattering cross section and the annihilation cross section of positrons with the noble gas atoms. It was discovered that the application of DC electric fields to these gases modified to a marked extent the observed positron annihilation spectrum. This technique proved a useful
tool in exploring the velocity dependence of the cross sections.

The experimental investigation of these phenomena and a subsequent analysis of these results constitutes the primary contribution of this work.

In order to measure the time distributions of the positron annihilation events a system for recording lifetimes over the time interval of 0 - 600 nsec was constructed. The resolution of this instrument was about $3\frac{1}{2}$ nsec.
A. Introduction

The following discussion is confined to an examination of the behavior of positrons in gases in the low energy region of 0 - 20 eV. Although the positrons are emitted into the gas with much larger energies (up to a maximum of 542 keV) the phenomena that concern us here occur in the low energy region. In explanation of the above statement we consider the initial period after the positron has been emitted into the gas, during which it loses energy very rapidly by ionization and inelastic collisions with the gas atoms. The time required for a positron of initial energy 500 keV to reduce its energy to about 5 keV has been calculated in Appendix E and is found to be 0.7 nsec in argon at 10 atm pressure. During such a short time interval one expects, on theoretical grounds (Heitler (1954)), that very few positrons would annihilate, and the experiments of Gerhart et al (1954), and Kendall and Deutsch (1956) do lead to the conclusion that only a small fraction of the positrons annihilate while they still have an appreciable energy. In addition, experiments on the angular correlation of annihilation radiation in gases (Heinberg and Page (1957)), indicate that the momentum of the annihilating electron-positron pair corresponds to energies of the order of a few electron volts or less.

The observation made during the present experiments that the parapositronium annihilations (referred to in the following sections) occur
in the 'prompt' region of the annihilation spectrum, verify the rapid initial slowing down of the positron.

B. Annihilation Mechanisms and Scattering of Positrons at Low Velocities

The behavior of positrons in gases, once they have reached energies of about 100 eV or less, is best described by reference to the diagram of Fig.1. Three mechanisms are depicted, all of which lead to the eventual annihilation of the positron. Firstly, annihilations resulting from an encounter between a free positron and an atomic electron are called 'free' or 'direct' annihilations. Secondly, a probability exists that a positron will capture an electron from an atom forming the hydrogen-like atom, positronium. Evidence for the occurrence of these two mechanisms is well established experimentally (Green and Lee (1964)) and will be further discussed in later sections. Finally, a positron may combine with an atom to form a molecular complex, the evidence for which has recently been reported by Paul and Saint-Pierre (1963). All these mechanisms compete, the relative magnitude of each process depending on the particular nature of the gas in question.

C. Dirac Cross Section for Positron Annihilation

Dirac (1930) first calculated the annihilation cross section of a positron-electron pair for dematerialization into two photons. For non-relativistic velocities his result for the cross section reduces to

$$\sigma_{2\gamma} = \pi r_0^2 c/v \text{ per electron}$$

where \(r_0\) is the classical electron radius and \(v\) the relative positron-electron velocity. The above result is the spin-averaged cross section obtained by treating the positron and electron as free and noninteracting, an approximation equivalent to the 'plane wave' quantum mechanical treat-
FIG. 1. Annihilation Mechanisms of Positrons in Gases

- **Ionization and Inelastic Collisions**
- **Thermalization of Free Positrons**
- **Rapid Energy Loss**
- **Negligible Annihilation**
- **Direct Annihilation**
- **Positronium Formation and Annihilation**
- **Molecular Complex Formation**
- **Formation Threshold**
- **Annihilation**
- **Quenching**
- **E_{\text{ion}}**
- **E_{\text{max}} = 542\text{keV}**
- **\text{Na}^{22}**
ment. If the annihilating electron-positron pair is essentially at rest, the two quanta are emitted at 180°, each with an energy of 0.51 MeV. Details of the above calculation have been given by Jauch and Rohrlich (1955) and DeBenedetti (1964). While the cross section given by (1) approaches infinity as the velocity goes to zero, the annihilation rate in a medium is independent of v, being given by

$$\lambda = n_P \sigma_{2\gamma} v$$

(2)

where \(n_P\) is the effective electron density in the medium. The limited applicability of (1) in explaining quantitative results will become apparent in the discussion of direct annihilation rates of positrons.

A collision of a positron and an electron occurs in the \(3S\) (triplet) state 3/4 of the time and in the \(1S\) (singlet) state 1/4 of the time. Since selection rules prevent the decay of the triplet state via two photons, the cross section (1) represents one quarter of the annihilation cross section from the singlet state. The next higher order process for which the \(^3S\) state decay is allowed is three quantum annihilation with a relative probability compared to 2-photon annihilation (in a single collision) of

$$P_{3\gamma}/P_{2\gamma} = (4/9 \pi) (\pi^2 - 9) = 1/1115$$

(3)

For the ratio of the spin averaged cross sections one then obtains

$$\sigma_{2\gamma}/\sigma_{3\gamma} = 372$$

(4)

It is clear that for free positron annihilation, three quantum decays contribute a negligible amount and can be ignored. The situation is wholly different when we come to consider the \(3S\) bound state - orthopositronium. This atom can maintain a relatively isolated existence in a gas, and thus, once it is formed, has a high probability of decaying by the emission of
Arguments and calculations leading to the above selection rules have been presented by Yang (1950), Jauch and Rohrlich (1955), and De Benedetti (1964).

D. Positronium

1. Structure of Positronium

A comprehensive discussion of the early experimental and theoretical investigation of positronium has been presented in two excellent review articles by Deutsch (1953) and DeBenedetti and Corben (1954). References to more recent literature on this subject can be found in a recent publication by Green and Lee (1964). The present discussion is restricted to those features of positronium which are important in the interpretation of the experimental results presented in this thesis.

The positronium atom consists of the bound state of an electron and a positron, an atom whose gross structure is very similar to that of the hydrogen atom. The effect of the different reduced mass of the positronium system is to reduce all energies by a factor of two from those of the hydrogen atom. Thus the ionization potential, or equivalently the binding energy of the positronium atom is 6.76 eV, the first excited state has an energy of 5.1 eV, and the Lyman alpha-line has a wavelength of 2400 Å. Also, because of this different reduced mass, the mean electron-positron separation is twice the electron-proton separation in the hydrogen atom.

Positronium in the ground state is characterized by either of the spin configurations $^1S$ and $^3S$, referred to as para and orthopositronium respectively. In the former, the positron and electron spin couple to
produce a total spin of zero, whereas in the latter case a total spin of one is produced. Statistically, one would expect that the para state should be formed in $1/4$ of the cases and the ortho state in $3/4$ of the cases, since the number of magnetic substates present are one and three respectively. Measurements by Falk and Jones (1964) indicate that a fraction, $0.69 \pm 0.07$, of the positronium formed in argon is in the ortho-state. Formation of positronium in excited states has been sought in experiments designed to measure the Lyman alpha-line (Huges (1957), Brock and Streib (1958), and Duff and Heymann (1963)) but has not been observed to date. The only reported experimental evidence pointing to such excited states (Heinberg and Page (1957)) is, at best, inconclusive. In this regard, Deutsch (1953) points out that capture leading to an excited state is unlikely since the minimum energy required for even the $2S$ state is $(E_{\text{ion}} - 1.7) \text{ eV}$ and at these relatively high energies inelastic collisions leading to excitation of the atom likely predominate.

2. **Lifetime of the Positronium Atom**

The lifetime of the positronium atom can be obtained from equation (2) by replacing the effective electron density, $n_p$, by the properly normalized wave function of the atomic problem, to yield the effective density of electrons at the position of the positron (Deutsch (1953)). Thus, for singlet positronium (in the ground state) the mean life is given by (Jauch and Rohrlich (1955))

$$\tau_{2\gamma} = (\sigma_{2\gamma} v \left| \Psi(0) \right|^2)^{-1} = 1.25 \times 10^{-10} \text{ sec},$$

and for the triplet state

$$\tau_{3\gamma} = 1.39 \times 10^{-7} \text{ sec}.$$

Ore and Powell (1949) have calculated the gamma ray spectrum
resulting from three-photon annihilation and their results show that the spectrum varies approximately linearly with energy from zero to a maximum of 0.51 MeV. This is of considerable importance experimentally and will be referred to again in Chapter IV.

3. **Probability of Positronium Formation**

The fraction of the positrons which form positronium in a gas has been studied by numerous investigators and the results for various gases are summarized by Green and Lee (1964). In an attempt to better understand the mechanism of positronium formation, a measurement of this fraction affords the most direct experimental approach.

Consider first the energetics of positronium formation by referring to the energy level diagram of Fig. 2. If the ionization energy of the gas atom is $E_{\text{ion}}$, then a positron energy of at least $(E_{\text{ion}} - 6.8)$ eV is required to form positronium since the binding energy of positronium is 6.8 eV. Ore (1949) has presented a qualitative argument yielding the fraction of positrons forming positronium. He assumes that the last ionizing collision suffered by the positron is equally likely to leave the positron anywhere within the energy range from zero to $E_{\text{ion}}$. In this approximation, then, an upper limit to the fraction forming positronium is $f_{\text{max}} = 6.8/E_{\text{ion}}$. This is clearly an upper limit since the presence of an excitation level in the energy range of positronium formation will rapidly eliminate positrons with energies above $E_{\text{exc}}$ and reduce their energies below $E_{\text{exc}}$ by inelastic collisions. A lower limit for the fraction formed can be obtained by considering the number in the interval between $E_{\text{exc}}$ and $E_{\text{thr}}$ ($= E_{\text{ion}} - 6.8$). This yields

$$f_{\text{min}} = (E_{\text{exc}} - E_{\text{thr}})/E_{\text{exc}}.$$
FIG. 2. Energy Level Diagram for Positronium Formation. $E_{\text{ion}}$ and $E_{\text{exc}}$ are the Ionization and the Lowest Excitation Levels in the Atom Respectively and $E_{\text{thr}}$ is the Threshold for Positronium Formation.
For historical reasons the energy interval of permissible positronium formation between $E_{\text{ion}}$ and $E_{\text{thr}}$ is called the Ore gap and the quantities $f_{\text{min}}$ and $f_{\text{max}}$ referred to as the Ore limits. These latter quantities are given in Fig. 2 together with the energies of the various levels for argon and helium.

Experimental values for the fraction of positrons forming positronium in argon have been reported as 0.27, 0.30, 0.31 (Green and Lee (1964)) and 0.37 (Falk and Jones (1964)). While all these results are within the Ore limits it should not be misconstrued that this validates the qualitative picture of the Ore limits. The assumption of a uniform energy distribution after the last ionizing collision is, in particular, questionable. Possible, more realistic would be the assumption of a constant density in phase space.

Further information on the mechanism of positronium formation has been provided by the experiments of Marder et al. (1956) and their detailed interpretation by Teutsch and Hughes (1956). These experiments consisted of applying a strong electric field to the positrons in the gas, sufficient for the positrons to establish equilibrium at kinetic energies of the order of several electron volts.

The effect of the electric field on the formation of positronium can be understood as follows. Referring to Fig. 2, it is evident that once the energy of a positron has dropped below $E_{\text{thr}}$ it is no longer energetically able to form positronium. The presence of the electric field alters this by providing energy to the positron whereby it may again cross the threshold. An increase in positronium formation was observed by the change in the gamma ray spectrum resulting from the increased three-
quantum annihilations. For increasing electric fields a saturation point was eventually reached, above which no further increase in positronium formation occurred. These results indicated that at saturation the fraction of positrons forming positronium had increased by a factor 2.08, 1.41, and 1.53 for A, Ne, and He respectively. The general features of these results have been substantiated by the further work of Obenshain and Page (1962).

Perhaps the most important physical result obtained from these electric field experiments was the determination of the elastic scattering cross sections for positrons from the noble gas atoms. This is of paramount importance to the present work and will be considered further in section I.

4. Quenching of Orthopositronium

Earlier in this section the mean lives of singlet (para) and triplet (ortho) positronium were given as 0.125 nsec and 139 nsec respectively. In practice, neither of these components is observed to have the theoretical value; the singlet, because of its very short lifetime, is obscured both by the relatively large instrumental time resolution and the initial slowing down period; the triplet, because of its long lifetime suffers many collisions with the gas atoms, during which some quenching occurs converting the triplet state into the singlet state.

Several of the different possible quenching mechanisms are summarized.

(i) Pick-off Quenching

During any collision of the positronium atom with a molecule of the gas, the positron is temporarily in a region of much
higher electron density and as a result has a considerable probability of annihilating with an atomic electron of suitable spin orientation. Experiments by Heymann et al (1961) and Duff and Heymann (1962) suggest that this is the dominant quenching mechanism in the noble gases. The quenching cross section which has a value of about $10^{-21}$ cm$^2$ (in argon) increases with increasing atomic number as one would expect for such a process. Typically, the orthopositronium lifetime in argon at 10 atm pressure is about 100 nsec. Celitans and Green (1964) and Celitans et al (1964) have obtained good agreement with these results, provided, however, that the argon contain an impurity such as O$_2$ with a concentration between one part in $10^5$ and one part in $10^3$. For purer argon they report a considerably smaller quenching cross section which does not vary linearly with the pressure.

(ii) Exchange Quenching

Gases such as NO and O$_2$ are well known for their strong quenching of orthopositronium (Gree and Lee (1964)). These paramagnetic molecules have one and two unpaired electrons respectively and may exchange these electrons with the electron of the positronium atom. The physical nature of this electron exchange mechanism is discussed by Ferrell (1958). The cross section for such a mechanism should be of the order of atomic dimensions or comparable to the elastic scattering cross section. For NO this is indeed what is found (Heymann et al (1961)). For oxygen, on the other hand, Celitans et al (1964) obtained a value
about two orders of magnitude smaller - a result too small to be explained in terms of the exchange mechanism.

(iii) Induced Quenching

In the presence of a static magnetic field (Hughes et al (1955)), the $m = 0$ substate of orthopositronium contains an admixture of the $^1S$ (para) state, while the $m = \pm 1$ substates are unaffected. The extent of the admixture is governed by the strength of the magnetic field.

E. Direct Annihilation

1. Experimental Annihilation Rates

The work of Deutsch (1951b) was the first to clearly indicate that one component in the annihilation spectra of positrons in gases could be attributed to direct positron annihilation. Observing the decay rates of positrons in oxygen (a gas which strongly quenches the orthopositronium component), he found a mean positron lifetime inversely proportional to the pressure. On the basis of the discussion in Section C of this chapter, the annihilation rate for free positrons is expected to be proportional to the electron density and hence to the pressure of the gas.

No further experimental results were reported in the literature until Falk and Jones (1963) observed pressure-dependent annihilation rates in argon and krypton, and Paul and Saint-Pierre (1963), a similar dependence in hydrocarbon gases. The measured annihilation rates in argon and krypton were respectively a factor of 1.96 and 1.87 times greater than the Dirac rate computed from eq. (2), assuming that all the electrons of the atom participated in the annihilation process. For oxygen, the result of Deutsch yields an enhancement of about 1.7 above the Dirac rate.
Further results reported in this thesis for O₂, N₂ and CO₂ exhibit a very similar trend. On the other hand, the results for the hydrocarbons revealed annihilation rates roughly 100 times greater than that predicted by the Dirac cross section. This anomalously large value was interpreted in terms of the formation of positron-molecular complexes, a possibility which is considered further in Section F.

2. Theoretical Situation

Returning to the expression for the Dirac cross section, equation (1), we again note that it was based on the 'plane wave' approximation. The physical situation is clearly different in the following respects:

(i) The positron and electron experience the long range Coulomb interaction.

(ii) The electrons are bound to nuclei, and

(iii) The positively charged nucleus presents a strong repulsive potential to the incoming positron.

Deviations from the Dirac annihilation rate as represented by the quoted experimental results is not difficult to understand in the light of the above considerations. The effect of the Coulomb attraction between the positron and electron can be approximately accounted for by multiplying the Dirac cross section by a properly normalized Coulomb wave function (DeBenedetti and Corben (1954)). The present accuracy of the measured direct rates, however, requires a much more rigorous theoretical treatment of the problem before any basis for comparison is said to exist.

3. Velocity-Dependent Effects

In the foregoing discussion it was implicit that the direct annihilation component is represented by simple exponential decay, in
agreement with the experimental observations. Such a situation results when the annihilation rate, obtained by combining equations (1) and (2), is independent of the positron velocity. During the past year, three groups working independently, have reported results at variance with the above picture. On studying the annihilation rate of positrons in argon, Tao et al. (1964), Falk and Jones (1964), and Paul (1964), all observed the existence of a distinct 'shoulder' in the time spectrum of the annihilating positrons, as illustrated in Fig. 28. Failure of earlier investigations to reveal this behavior have been attributed both to poorer instrumental time resolution and the presence of polyatomic impurities in the argon.

A brief outline of the interpretation given by the above authors to the foregoing observation follows. In gases with no low-lying excitation levels, a situation characterizing the noble gases, the slowing-down time of a positron is very long compared to that in a diatomic molecule, say O₂. Tao et al. (1963) have shown that the slowing-down time for a positron in argon at 1 atm pressure from 9 eV to a final energy of 1 eV is of the order of 100 nsec. Since this time is of the same order as the mean life of a positron in the direct component, these observations have been interpreted in terms of a velocity-dependent direct annihilation rate in these gases. In the diatomic gases, however, the free positrons are assumed to thermalize so rapidly that the observed direct annihilation rate is that characterizing a velocity dependent rate averaged over the thermal equilibrium distribution of the positron population. A single exponential decay would thus be expected, as observed experimentally.

Another feature of the observed positron annihilation spectrum in argon is that the direct annihilation rate reverts to a constant value
after a time of about 340 nsec atm. This constant rate is manifested by
the single exponential decay, characteristic of this region of the spectrum.
A likely explanation of this observation is that the beginning of the
exponential decay corresponds to the time at which all the positrons are
thermalized.

The detailed shape of the annihilation spectrum depends both
on the velocity distribution of the positrons as a function of time and
the velocity dependent annihilation rate. In terms of more fundamental
quantities, the spectrum shape is determined by:

(i) The elastic positron-atomic scattering cross section (a
function of the positron velocity), $q(v)$.
(ii) The velocity dependent annihilation cross section, $\sigma(v)$.
(iii) The initial ($t = 0$) velocity distribution of free positrons.

Although the cross sections $q(v)$ and $\sigma(v)$ are of fundamental
interest, their measurement to date has defied experimental technique. The
work described in this thesis represents an experimental investigation of
some features of these velocity dependent cross sections as reflected in
the detailed shapes of the positron annihilation spectra reported in
this thesis.

The mathematical framework relating the fundamental quantities
mentioned above to the experimentally observable quantity, the annihilation
spectrum, is developed in Section H of this chapter and also in Appendix G.

F. Formation of Positron-Molecular Complex

In the foregoing section it was pointed out that the direct
annihilation rates observed in the hydrocarbon gases were about two orders
of magnitude greater than that predicted by the Dirac cross section. A
tentative explanation put forth by Paul and Saint-Pierre (1963) was that of the possible formation of a positron-molecular complex. Such a close association of a positron with a single atom would greatly enhance the annihilation probability because of the increased average electron density at the position of the positron. A similar hypothesis has been advanced by Green and Tao (1963) to explain the short lifetime of the direct component in freon-12. Here the annihilation enhancement factor compared to the Dirac rate is not as marked as for the hydrocarbons, being only one order of magnitude.

The tentative explanations of the large annihilation rates observed in some molecular gases has received some theoretical support. Khare et al (1964) have shown, by a variational calculation, that it is possible for a positron to form a bound state with a helium atom having a binding energy of about 0.55 eV. They further suggest that the presence of a bound state in helium, the substance in which it would seem least likely, makes it quite probable for bound states of the positron to exist with any atom or molecule.

G. Elementary Theory of the Slowing Down of Positrons in the Noble Gases

1. Introduction

In this section the slowing down behavior of positrons in the noble gases is considered in terms of simple elementary principles. Such considerations are greatly simplified by the fact that the ratio of the positron mass to the gas atom mass is very small. If positron kinetic energies in the range from zero to the positronium formation threshold (which is lower than the first excitation level in the noble gas atoms) only are considered, then except for the possible effect of virtual positronium
formation, only elastic collisions can occur. Neglecting all effects except those of elastic collisions, the theory governing neutron thermalization may be applied directly to this problem.

In the calculations which follow frequent use will be made of the quantities which are listed below. For convenience in making comparisons to the experimental results, all the numerical values shown have been calculated for a temperature of 25°C. This procedure has been followed consistently in all the work reported in this thesis.

(i) Ratio of positron mass to noble gas atom mass - \( m/M \).
\[
(m/M)_A = 1.376 \times 10^{-5}, \quad (m/M)_{\text{He}} = 1.371 \times 10^{-4}
\]

(ii) Density of atomic scatterers - \( N \).
\[
N = 2.464 \times 10^{19} \text{ P atoms/cc}
\]
where P is the gas pressure in atmospheres at 25°C.

(iii) Thermal velocity of a positron (at 25°C) - \( v_0 \).
\[
v_0 = \sqrt{\frac{3kT}{m}} = 1.164 \times 10^7 \text{ cm/sec}
\]

(iv) The positron velocity will frequently be written in terms of the dimensionless quantity, \( x = v/v_0 \). At a temperature of 25°C the thermal energy of a positron is 0.0386 eV and hence the energies 8.9, 11.6, and 15.68 eV (the positronium formation threshold, the first excitation level, and the ionization potential in argon respectively) correspond to values of \( x \) of 15.2, 17.3, and 20.2 respectively.

(v) Positron acceleration in an electric field - \( a \).
\[
a = 1.759 \times 10^{15} E \text{ cm/sec}^2
\]
where \( E \) is the electric field in volts/cm.

(vi) The elastic scattering cross section \( q(v) \) (in units of
cm\(^2\) is related to the cross section \(\sigma(v)\), expressed units of \(\pi a_0^2\) (\(a_0\) is the radius of the first Bohr orbit), by the expression
\[
\sigma(v) = 0.880 \times 10^{16} \, \text{cm}^2
\]

2. **Energy Loss of a Positron when Elastically Scattered from a Noble Gas Atom**

(a) **Colinear Elastic Collision**

Consider a one-dimensional elastic collision between a positron and an atom having velocities \(v\) and \(w\), and masses \(m\) and \(M\) respectively. The quantity \(m/M\) is very small and the atom is assumed to be in thermal equilibrium (at 25°C). For the case where the velocities of the two particles are colinear before and after the collision, the fractional change in energy of the positron can be easily shown to be
\[
\Delta E/E = -4 \left( \frac{m}{M} + \frac{w}{v} - \frac{w^2}{v^2} \right)
\]
correct to order \(w^2/v^2\). Here, \(w\) is understood to be positive for initial velocities parallel, and negative when antiparallel, with the result that the value of the second term in (5), averaged over many collisions, is zero. The average fractional energy loss in a single collision is thus very small indeed, of order \(m/M\), or approximately \(10^{-5}\) for the case of argon. The maximum fractional energy loss in a single collision is, however, much greater, being given by
\[
\Delta E_{\text{max}}/E = \left( \frac{4m}{M} \right) \left( \frac{v_0}{v} \right)
\]
where we used \(\frac{1}{2}mv_0^2 = \frac{1}{2}mv^2\).

(b) **Target Atom at Rest**

In the case where the target atom is at rest it can readily be shown that when scattered at an angle \(\theta\), the fractional energy loss of the
The positron is
\[ \Delta E/E = (2m/M)(1 - \cos \theta) \]  \hspace{1cm} (6)

(c) **Average Energy Loss Taking into Account the Thermal Motion of the Atoms**

In calculations of the slowing down time of a positron in a gas, the average energy loss of a positron in an encounter with a gas atom is required. Here the effect of the thermal motion of the atoms must be considered since the behavior of the positron at energies comparable to thermal energies is of particular interest. The solution to this problem has been given by Amaldi (1959) for the case of neutron thermalization and is applicable to the present discussion. He gives the result
\[ \Delta E/E = -(2m/M)[1 - (4/3)(E_t/E)] \]  \hspace{1cm} (7)

where \( \Delta E^- \) is the average energy loss in an elastic collision and \( E_t \) is the thermal energy of the atoms.

3. **Average Energy Gain of a Positron in an Electric Field**

If an electric field is applied to the positrons in a gas the positrons gain an average energy between collisions of (Marder et al. (1956))
\[ \Delta E^+ = \frac{m^2a^2}{4E(Nq)^2} \]  \hspace{1cm} (8)

where \( a \) is the acceleration of the positron produced by the electric field and \( N \) and \( q \) are the density of scatterers and the elastic scattering cross section respectively. This result is obtained by the following argument. Let the mean time interval between collisions be \( t \), and the mean free path, \( L \). Then
\[ t = L/v = 1/(Nqv) \]  \hspace{1cm} (9)

The energy gain in a mean free path length is
\[ \Delta E = \frac{1}{2}m(\bar{v} + \bar{a}t)^2 - \frac{1}{2}mv^2 = m\bar{v} \dot{a}t - \frac{1}{2}ma^2 t^2 \quad (10) \]

Substituting for \( t \) from equation (9) and noting that the average value of \( \bar{v} \bar{a} \) is zero, the expression (8) is obtained for the average energy gain in the presence of the electric field.

4. **Slowing Down in the Presence of an Electric Field**

(a) **General Case**

The general case in which an electric field is applied to the positrons in the gas is now considered, and the time calculated for a positron to slow down from an initial energy \( E_1 \) to some final energy \( E_2 \).

The rate at which collisions occur between the positron and the gas atoms is \( Nqv \) per sec. Using the expressions (7) and (8), the change in the positron energy in the time \( dt \) is given by

\[ dE = \left[ -\frac{2m}{M}(1 - \frac{4}{3} E)E + \frac{m^2 a^2}{4E(Nq)^2} \right] Nqvd \quad (11) \]

Rearranging, one obtains

\[ t = \int_{v_1}^{v_2} \frac{dv}{Nq \left[ -\frac{m}{M} v^2 - \frac{4}{3} \frac{m}{M} v_0^2 + \frac{a^2}{2(Nqv)^2} \right]} \quad (12) \]

for the time required to slow down from an initial velocity \( v_1 \) to a final velocity \( v_2 \). In this equation the thermal energy of the atoms, \( E_t \), has been replaced by the thermal energy of the positrons, \( \frac{1}{2}mv_o^2 \). From equation (11) it is noted that equilibrium is established when the expression in the brackets is zero. This equilibrium velocity is denoted by \( u \) and the corresponding energy by \( E_e \). Further, denoting the elastic scattering cross section at the thermal velocity \( v_0 \), by \( q_0 \), the following thermalization time constant is defined.
\[ T_c = \frac{1}{2Nq_0v_0m/M} = \frac{1}{(5.05 \times 10^{10}P_{q_0}m/M)} \]  

(b) **Special Cases of the Scattering Cross Section**

Consider the specific case in which the scattering cross section has the form

\[ q(v) = q_0 \frac{v}{v_0} \]  

(14)

For this case the equilibrium condition yields

\[ \frac{a^2}{2(Nq_0v_0)^2} = \frac{m}{M} \left( u^2 - \left(\frac{4}{3}v_0^2\right) \right) \]  

(15)

Introducing the quantities \( T_c, q(v), \) and \( u, \) equation (12) can be integrated to yield

\[ \left(\frac{v_2}{u}\right)^2 = 1 - \left[ 1 - \left(\frac{v_1}{u}\right)^2 \right] e^{-t/T_c} \]  

(16)

As a numerical example we consider the case of argon at a pressure of 10 atm and take for \( q_0 \) a value of 10 (in units of \( \pi a_o^2 \)). This value for \( q_0 \) is of the same order of magnitude as that calculated theoretically by Malik (1961). Further, taking the electric field to be zero so that \( u \approx v_0, \) the time required for a positron to slow down from an energy of 9 eV to 1 eV is found to be 32 nsec, and the thermalization time constant, \( T_c, \) is 14.4 nsec. Thus for the magnitude of the cross section assumed, a time, comparable to the direct annihilation mean lifetime, is required for a positron to slow down.

The case of a constant scattering cross section has been treated by Tao et al. (1963) and will not be repeated here. For the same example as above, but using the constant cross section of \( 1.5 \pi a_o^2 \) his calculations give a slowing down time to 1 eV of 23 nsec.

5. **Special Case of Slowing Down in the Absence of an Electric Field**

**the Neglecting the Thermal Motion of Scattering Atoms**

In the special case where no electric field is present and in the
energy region above about 1 eV where the thermal motion of the scattering atoms can be neglected (i.e. $v_2 \gg v_o$), the solution to equation (12) is particularly simple. The results for two different choices of the scattering cross section are

$$v_2 = v_1 e^{-t/2T_c}, \quad q(v) = q_0 v_0/v$$

and

$$v_2 = v_1/(1 + \frac{v_1 t}{2T_c}), \quad q(v) = q_0$$

where all the quantities have the same meaning as previously defined.

These forms for the slowing down expressions are particularly convenient in making estimates and comparisons among the various noble gases.

6. Collision Rate Between a Positron and the Scattering Atoms

The rate at which the positron suffers collisions with the scattering atoms is given by

$$r_c(v) = Nq(v)v/sec$$

At the thermal velocity, $v_o$, this reduces to

$$r_c(v_o) = 1/(2T_c m/M)/sec$$

For the earlier example where $T_c = 14.4$ nsec, the above thermal collision rate is $2.5 \times 10^{12}$/sec. This last result is of particular significance when we consider the effect of polyatomic impurities among the noble gas scattering atoms. Taking as a crude example a case where a low lying excitation level in the polyatomic impurity molecule has an inelastic scattering cross section of the same magnitude as the elastic scattering cross section of the noble gas atom, and assuming that the relative density of the impurity molecules is $10^{-4}$, it is found that the positron mean free time between collisions with the polyatomic molecules is about 4 nsec. Clearly, for the value of the excitation cross section assumed,
an impurity concentration of $10^{-4}$ would seriously alter the slowing down behavior of the positrons in the gas.

7. Importance of Diffusion Effects

In the foregoing slowing down calculations only the average energy losses and gains suffered by the positron have been considered. This approximation neglects diffusion effects which are required to take into account the random walk executed by the positron in velocity space. To appreciate the significance of such diffusion effects, the ratio of the maximum energy loss to the average energy loss in an elastic collision has been calculated and is denoted by $R_1$ in Table I. For a 9 eV positron in argon this ratio has a value of 35 and demonstrates the large "steps" that the positron can take in velocity space.

Similar quantities relating to the electric field are also shown in Table I for the general case, and also for the two specific cases considered for the elastic scattering cross section. The results shown in the table are valid for positron energies greater than about 1 eV since the thermal motion of the atoms has been neglected. For further simplification, the acceleration $a$ has been defined in terms of the equilibrium velocity $u$ (and the corresponding energy $E^*$) obtained from equation (11).

H. The Diffusion Equation and Calculation of the Positron Annihilation Rate

1. The Diffusion Equation

(a) Differential Equation

At the end of the foregoing section the importance of taking diffusion effects into account when determining the slowing down behavior
### TABLE I

Energy Loss and Gain of a Positron in a Uniform Electric Field in a Monatomic Gas. The Expressions Shown are Valid for $E \approx 1$ eV. The Equilibrium Energy and Velocity are $E^*$ and $u$ Respectively.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>General Expression</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Average energy loss in an elastic collision (from eq. (7)), $\Delta E^-$</td>
<td>$2(m/M)E$</td>
</tr>
<tr>
<td>2. Maximum energy loss in an elastic collision (from eq. (9a)), $\Delta E_m^-$</td>
<td>$4\sqrt{m/M} \left(\frac{v_0}{v}\right)E$</td>
</tr>
<tr>
<td>3. $R_1 = \frac{\Delta E_m^-}{\Delta E^+}$</td>
<td>$2\sqrt{M/m} \left(\frac{v_0}{v}\right)$</td>
</tr>
<tr>
<td>4. Average energy gain from an electric field in a mean free path length (from eq. (8)), $\Delta E^+$</td>
<td>$\frac{(ma)^2}{4E(Nq)^2}$</td>
</tr>
<tr>
<td>5. Maximum energy gain from an electric field in a mean free path length (from eq. (10)), $\Delta E_m^+$</td>
<td>$\frac{ma}{Nq}$</td>
</tr>
<tr>
<td>6. $R_2 = \frac{\Delta E_m^+}{\Delta E^+}$</td>
<td>$\frac{4NqE}{ma}$</td>
</tr>
<tr>
<td>7. $G_1 = \frac{\Delta E^+}{\Delta E^-}$</td>
<td>$\frac{E^*}{E}$</td>
</tr>
<tr>
<td>8. $G_2 = \frac{\Delta E_m^+}{\Delta E_m^-}$</td>
<td>$\frac{v}{u}$</td>
</tr>
</tbody>
</table>

Note: For $q(v) = \frac{q_0 v_0}{v}$ and $q(v) = q_0$.
of positrons in the noble gases, was noted. In Section E of the present chapter it was also pointed out that the detailed shape of the annihilation spectrum depended on the velocity distribution of the positrons as a function of time and the velocity dependent annihilation rate. The appropriate mathematical treatment of this problem is provided by the diffusion equation which is derived in Appendix G.

The positron velocity distribution function, $f(\vec{v},t)$, is defined in such a way that the number of positrons in the volume element $d\vec{v}$ of velocity space is given by $f(\vec{v},t)d\vec{v}$. For the special case of interest here in which the ratio of the mass of the positron to the mass of the scattering atom is much less than one, the differential equation for $f$ is shown in Appendix G, to be

$$\frac{\partial f}{\partial t} = \frac{1}{v^2} \frac{\partial}{\partial v} \left[ \left( \frac{a^2 v^2}{3} + \frac{m v^3 \mathcal{r}_c(v)}{M} \right) \frac{\partial f}{\partial v} + \frac{m v^3 \mathcal{r}_c(v)f}{M} \right] - \left[ r_a(v) - r_f(v) \right] f$$

where $r_c(v)$, $r_a(v)$, and $r_f(v)$ are the elastic scattering collision rate, the direct annihilation rate, and the positronium formation rate respectively. The function $f$ appearing in the above equation is isotropic in velocity space. Equation (21) can be solved in closed form only for a few special cases of the functions $r_c(v)$, $r_a(v)$, and $r_f(v)$. Thus for the general case, a numerical solution has to be obtained. This latter approach was the one followed here, and a brief description of the numerical method used is presented in Appendix G.

(b) Equilibrium Solution for Zero Annihilation Rate

The particular case where both the direct annihilation rate and the positronium formation rate are zero and the distribution is in
equilibrium, can be readily solved. In equation (21) the left hand side is thus zero and the term in the curly brackets equal to a constant. Since the term \( v^2 f \) and also its derivative approach zero rapidly for large \( v \), an examination of the resulting equation reveals that this constant must be zero. The solution is

\[
f = A \exp \left\{ -\int \frac{mvdv}{Ma^2/(3(nq)^2) + kT} \right\}
\]

(22)

where we have substituted \( Nq(v)v = r_c(v) \). To simplify the above expression further, the explicit velocity dependence of \( q(v) \) must be introduced. For the two specific cases of a constant cross section and a cross section inversely proportional to the velocity the distribution function has the form

\[
f = A \exp \left\{ -\frac{m}{kT} \left[ \frac{v^2}{2} - \frac{1}{2kT} \frac{Ma^2}{3(Nq_o)^2} \ln \left( \frac{Ma^2}{3(Nq_o)^2} + v^2kT \right) \right] \right\}
\]

(23)

and

\[
f = A \exp \left[ -\frac{mv^2/v}{Ma^2/(3(Nq_o)^2) + kT} \right]
\]

(24)

\[
q(v) = q_o \frac{v_o}{v}
\]

(c) Equilibrium Solutions for High Electric Fields

The solution of the above equations neglecting the finite temperature of the scatterers is now considered. The resulting expressions will be valid when the effect of the electric field represented by the first term in the denominator of (22), is much greater than \( kT \). For simplicity and convenience the electric field term has been expressed in terms of the equilibrium velocity \( u \), obtained from the elementary theory described in the previous section. Also, the expressions in terms of \( E \),
P, and Q on the right hand sides of (25) and (26) are for the specific case of argon. These relationships, obtained by putting $E_t = 0$ in equation (11), are

$$(u/v_o)^2 = \frac{M/(2m)}{NQ_o V_o^2} a = 1.142 \frac{E}{PQ_o} , \quad q(v) = q_o$$ (25)

and

$$(u/v_o) = \frac{M/(2m)}{NQ_o V_o^2} a = 1.142 \frac{E}{PQ_o} , \quad q(v) = q_o v_o/v$$ (26)

Here $E$ is the electric field in volts/cm, $P$ is the gas pressure in atmospheres, and $Q_o$ is the elastic scattering cross section at thermal velocities, expressed in units of $\pi a_o^2$. Introducing the above quantities into equation (22) and putting $T = 0$ one obtains for the normalized distribution functions

$$f = 0.816 (3/8)^{3/4} \frac{1}{\pi u^3} \exp \left[ -3/8 \left( v^4 / v_o^4 \right) \right] , \quad q(v) = q_o$$ (27)

and

$$f = \frac{3}{8\pi u^3} \sqrt{3/\pi} \exp \left[ -3/4 \left( v^2 / v_o^2 \right) \right] , \quad q(v) = q_o v_o/v$$ (28)

For comparison we calculate the distribution function for zero electric field, and noting that $kT = mv_o^2/3$, obtain the result

$$f = \left( \frac{3}{2\pi} \right)^{3/2} \frac{1}{v_o^3} \exp \left[ -3/2 \left( v^2 / v_o^2 \right) \right] , \quad \text{thermal dist.}$$ (29)

This latter distribution is immediately recognized as the Maxwell-Boltzmann distribution function. In the presence of an electric field the function $f$ is still Maxwellian for the case of equation (28), but with an 'effective temperature' determined by the electric field. The situation is wholly different for the case of a constant scattering cross section where the
exponential contains the term $v^4$ in contrast to the term $v^2$ in the Maxwellian distribution. This distribution, called the Druyvesteyn distribution, is characterized by a much narrower width of the velocity distribution than the Maxwellian distribution, as can be seen in a comparison of the two plots of $v^2f$ shown in Fig.3. The significance of this difference will become clearer in the discussion of the experimental results.

(d) Effect of Electric Field on Positronium Formation

As pointed out by Marder et al (1956), the effect of an electric field on positronium formation in gases can be understood in terms of the positron distribution function resulting under the action of an applied electric field. As the electric field increases the average velocity of the positrons increases, and hence the probability of a positron crossing the threshold for positronium formation increases correspondingly.

It is instructive to calculate the fraction of the positrons in the region of the distribution function above the positronium formation threshold (8.9 eV in argon), for the case when the annihilation and formation cross sections are assumed zero. Denoting this fraction by $F$, and taking for the magnitude of the electric field a value corresponding to $E/P = 122 \text{ V/(cm atm)}$, one obtains, from (27) and (28),

$$F = 0.056, \quad q(v) = q_0, \quad Q_0 = 1.5$$

and

$$F = 0.32, \quad q(v) = q_0 v_0/v, \quad Q_0 = 13$$

The values of the scattering cross section have been taken as $1.5 \pi a_o^2$ and $13 \pi a_o^2$ respectively for the two cases. The equilibrium energies which result for these values of $E/P$ and the cross sections are about 4.0 and
FIG. 3. Comparison of the Maxwellian and Druyvesteyn Velocity Distributions.
5.0 eV respectively. These results for $F$ demonstrate that even at these values of the equilibrium energy - about one half of the threshold value - the extension of the positron velocity distribution above the threshold is considerable. Furthermore, the much larger value for $F$ for the Maxwellian distribution than for the Druyvesteyn distribution is indicative of the different shapes of these distributions.

In the foregoing numerical calculation, tables of the Incomplete Gamma Function compiled by Pearson (1951) were employed.

2. Positron Annihilation Rate

(a) General Case

It is assumed for the present calculations that the time-dependent distribution function (equation (21)) is known so that the direct positron annihilation rate can be determined. The cross sections per atom for direct annihilation and positronium formation are denoted by $\sigma(v)$ and $s(v)$ respectively and are related to $r_a(v)$ and $r_f(v)$ by the expressions

$$r_a(v) = N\sigma(v)v, \quad r_f(v) = Ns(v)v$$

(30)

The quantities $r_a(v)$ and $r_f(v)$ represent respectively, the direct annihilation rate and the positronium formation rate of positrons with velocity $v$. The rates observed experimentally represent the velocity-averaged rates which we denote by $\lambda_a(t)$ and $\lambda_f(t)$ for direct annihilation and positronium formation respectively. Performing this averaging we obtain

$$\lambda_a(t) = \int_0^\infty 4\pi N\sigma(v)v^2 f^*(v,t) dv$$

(31)

and a similar expression for $\lambda_f(t)$, with $\sigma(v)$ replaced by $s(v)$ in the above expression. Here $f^*(v,t)$ denotes the normalized distribution at time $t$. 
(b) **Special Cases**

In general, $\lambda_a(t)$ is time dependent as indicated by equation (31). There are two notable exceptions however - one, when the annihilation cross section is inversely proportional to the velocity, and secondly, when the distribution function is stationary. Both cases result in a constant observed annihilation rate manifested by a single exponential decay.

The annihilation spectra recorded for argon reveal an exponential decay in the direct annihilation component in the region beyond the shoulder, both for the zero field case, and also for the non-zero field case. Furthermore, the observed variation of the direct annihilation mean lifetime with the electric field strength (Chapter V) indicates that the annihilation cross section is not inversely proportional to the velocity. Thus, one is led to the conclusion that the positron velocity distribution giving rise to the exponential decay, is in equilibrium.

Adopting this latter interpretation, the direct annihilation rates for the cases of the three stationary distribution functions, equations (27), (28), and (29) may be estimated. The assumption has to be made, however, that these distributions will not be seriously altered by the presence of the non-zero annihilation rate, although they were derived for the specific case of zero annihilation rate, only. For these calculations the direct annihilation cross section is assumed to have the functional form

$$\sigma(v) = \sigma_o(v_o/v)^n$$

and the positronium formation cross section is assumed to be zero.

The annihilation rate computed from equation (31) is the
reciprocal of the mean lifetime which we denote by $\tau_a(u)$ for equations (27) and (28), and by $\tau_a(v_0)$ for equation (29). Experimentally, the significant quantity is the ratio $\tau_a(u)/\tau_a(v_0)$ which, using equation (31), can be shown to be (for argon)

$$\frac{\tau_a(u)}{\tau_a(v_0)} = 0.346 \left[ 2.796 \frac{E}{P_0} \right]^{(n-1)/2} \left[ (4-n) \frac{\Gamma(2-n/2)}{\Gamma(2-n/4)} \right]$$  \hspace{1cm} (33)

and

$$\frac{\tau_a(u)}{\tau_a(v_0)} = \left[ 1.615 \frac{E}{P_0} \right]^{n-1} \hspace{1cm} (34)$$

where $\Gamma(x)$ denotes the Gamma function. The equilibrium velocity $u$ is related to $E, P$, and $Q_0$ by the equations (25) and (26). These results will be used in the analysis of the results presented in Chapter V.

The significance of undertaking a detailed program of investigation of the positron annihilation spectra in argon and the other noble gases when subjected to applied electric fields should be evident from the foregoing discussion. A series of experiments were planned in which the annihilation spectra could be recorded in detail for the case of zero electric field and also for uniform electric fields up to 1500 v/cm. For this purpose a large pressure chamber was built (Chapter III) that could withstand pressures of up to 70 atm and in which uniform electric fields could be produced.

I. The Elastic Scattering of Positrons from Atoms

1. Experimental Situation

Since the diffusion of positrons in the noble gases is governed
by the elastic scattering cross section, it is of interest to examine the present state of knowledge concerning the elastic scattering of positrons from atoms. Unfortunately, the experimental information is limited to the results of a single experiment - that of Marder et al (1956), while the theoretical situation is so inadequate that it can serve only as a general guide.

The experiment of Marder et al (1956) involved a measurement of the increase in positronium formation in the presence of an electric field. Detailed interpretation of their measurements by Teutsch and Hughes (1956) permitted a determination of the elastic scattering cross sections of positrons from several noble gases. Their results for He, Ne, and Ar were 0.23, 0.12, and 1.5 \( \pi a_0^2 \) respectively. These values can be considered averages of the cross sections in the neighborhood of the thresholds for positronium formation which occur at energies of 17.7, 14.7, and 8.9 eV respectively in these gases.

2. Theoretical Situation

(a) General

The theory of positron scattering differs from electron scattering in the following respects:

(i) The mean scattering potential seen by the positron is repulsive, whereas for electron scattering the mean scattering potential is attractive.

(ii) Positron scattering lacks the exchange effects which characterize electron scattering because, of course, the positron is distinguishable from the electron.

(iii) Positronium formation (real and virtual) can occur in
positron scattering.

A general feature of computed positron scattering cross sections is that they are normally at least an order of magnitude smaller than the corresponding electron cross sections. For helium, for example, La Bahn and Callaway (1964) calculate an electron cross section of about $20\pi a_o^2$ at an energy of 1 eV, compared to a calculated positron cross section of about $0.7\pi a_o^2$ (Malik (1961)).

(b) **Scattering by Hydrogen Atoms**

The majority of the theoretical work to date has concentrated on the problem of positron scattering from atomic hydrogen. The crucial problem in these calculations is that of adequately taking account of the polarization effects which are induced in the atomic electron cloud by the positively charged positron. According to Temkin (1962), the phenomenological aspects of this problem are clear: At large distances from the atom the positron experiences an attractive force due to the induced dipole moment; at close distances the positron feels the repulsion of the nuclear core. The many different methods of attacking the positron - hydrogen scattering problem have been discussed by Burke and Smith (1962) in a recent review article. We summarize here only some of the important conclusions of the various investigators.

An initial variational calculation by Massey and Moussa failed to reveal the existence of any significant polarization effects. Subsequently, further variational calculations by Moussa (1959) revealed that at energies below 5 eV the virtual formation of positronium was, however, of importance. In addition, Spruch and Rosenberg (1960) found that at low energies polarization effects were sufficiently large to produce a net
attraction of the positron to the atom, thus increasing the scattering cross section at these low energies. Using Kohn's variational method, Allison et al. (1961) found an increase in the cross section from 1.35 to 2.43 \( \pi a_o^2 \) (at zero energy) when allowance was made for polarization.

Results of other investigators in the field (Bransden (1962), Schwartz (1961), Rotenberg (1962), and Temkin (1962)) are in qualitative agreement with the conclusions of the other workers but large quantitative differences exist. Schwartz (1961) and Rotenberg (1962) give cross sections of about 9 and 7 \( \pi a_o^2 \) respectively at an energy of 1/4 eV which decrease to a value of \( \pi a_o^2 \) at about 1\(^\frac{1}{2}\) eV energy.

When the theoretical positron-hydrogen scattering cross section is compared to that of the electron-hydrogen case (Burke and Schey (1962)) one observes that the latter is again about an order of magnitude larger.

(c) Scattering from the Noble Gases

Turning to the scattering cross sections for the noble gases \(^{\text{A}}\) and \(\text{He} \), the available experimental and theoretical results are illustrated in Fig. 4. Variational calculations by Malik (1961) were performed using as the scattering potentials analytical approximations of the Hartree potentials. Similar calculations by Massey and Moussa (1958), which also neglected polarization effects, yielded results in qualitative agreement with those of Malik. These theoretical results are still much larger, however, than the experimental values (Teutsch and Hughes (1956)). A more detailed calculation by Allison et al. (1961) using the Kohn variational principle indicated that for helium the cross section (at zero energy) decreased from 0.70 to 0.10 \( \pi a_o^2 \) when allowance was made for polarization. This situation is in marked contrast with the case of atomic hydrogen,
discussed earlier, in which an increase in the cross section was obtained.

Summarizing these results, one concludes that

(i) Present theoretical methods are inadequate to predict accurately the elastic scattering cross sections.
(ii) There is a dire need for more experimental results.
(iii) It is possible that more rapid variations of the cross section with energy occur than predicted by either of the calculations of Massey and Moussa (1958) or Malik (1961).

Mctt and Massey (1952) indicate that for a repulsive potential the cross section cannot show as rapid a variation with energy as it can for an attractive potential, such as that characterizing electron scattering. The inclusion of polarization effects for the positron problem reveals, however, that the potential cannot be represented by a repulsive force alone, and that indeed, the potential may change from a repulsive one to an attractive one, as has recently been predicted for the positron-helium interaction by Khare et al (1964).
FIG. 4. Theoretical and Experimental Cross Sections for Elastic Scattering of Positrons from Argon and Helium.
CHAPTER III

ELECTRONIC AND MECHANICAL APPARATUS FOR
RECORDING OF POSITRON ANNIHILATION SPECTRA

A. Introduction

The electronic system used for obtaining the positron lifetime measurements in gases is shown schematically in Fig. 5. A chamber containing the gas under study and the positron source was mounted between the two gamma ray counters as indicated.

A discussion of the overall operation of the system when measuring the lifetime of a positron follows. A 1.28 MeV gamma ray emitted immediately following the beta-decay of a Na$^{22}$ nucleus activates the prompt limiter and produces two pulses as shown in Fig. 5. The positron associated with this beta-decay event loses energy rapidly in passage through the gas and in a time of the order of $10^{-9}$ sec (see Appendix E) has reached an energy of 10 - 20 eV. The eventual annihilation of the positron with an electron of a gas atom results in the emission of either two or three quanta, one of which activates the delayed limiter. Again two pulses are produced as was the case for the other limiter. The two signals appearing at the input of the time to amplitude converter (hereafter referred to as the time sorter, or just simply TS) from the 'prompt' and 'delayed' limiter define the 'birth' and 'death' of the positron respectively. A voltage pulse, proportional to the time separation of these two events and representing the positron lifetime is produced by the time sorter and fed into a 100-channel pulse height analyzer. A detailed description of the time sorter and its performance is given in Appendix B.
FIG. 5. Electronic System Used for Positron Lifetime Measurements in Gases.
In addition to a genuine event of the type described above there are several other possibilities in which the counters are activated and an output signal from the time sorter produced. These may consist of a legitimate signal in one counter and a photomultiplier noise pulse in the other, or a single gamma ray scattering from one counter to the other, activating both limiters. Since, however, a genuine time sorter output pulse can arise only if the initiating event in the prompt limiter is a 1.28 MeV gamma ray, nearly all the undesirable events may be eliminated by performing an energy selection on the gamma rays activating the limiters, using single channel pulse height analyzers. This is accomplished by using the standard "fast-slow" coincidence system (Bell et al (1952)). The outputs of the single channel analyzers are fed into a coincidence unit which produces a gating pulse for the 100 channel analyzer.

Another requirement which has to be satisfied by all genuine positron lifetime measurements is that the prompt limiter be activated before the delayed limiter. An inversion of this sequence resulting in a time sorter output corresponds to a random coincidence event. These random coincidences are termed "negative Time" events and constitute one half of all the random coincidences. By the use of an additional circuit called the "negative time eliminator", the kick sorter gating signal is inhibited whenever a negative time event has occurred and thus the corresponding time sorter output signal is not analyzed.

The fidelity of the time to amplitude conversion depends, among other things, on the complete restoration of the time sorter electronics to its quiescent state prior to the analysis of an event. If the pulse rates from the limiters are high, some of the events will
be processed by the time sorter before it has completely recovered from a previous event. In order to prevent registration of such events a pile-up rejector was incorporated in each of the counter channels. The function of this circuit is that of producing an output whenever an input pulse occurs within a predetermined time, \( T \), of a preceding input pulse. The output of the pile-up rejector is fed into an anticoincidence circuit together with the kick-sorter gating pulse thereby inhibiting the gate whenever a pulse pile-up has occurred.

In summary, we note that the kick-sorter is gated "on" and allowed to analyze a signal from the time sorter only when:

(i) The energy dissipated by the gamma ray in each counter falls within the energy interval defined by its respective single channel analyzer.
(ii) No pile-up of pulses has occurred in either counter.
(iii) The pulses from the limiters are in the proper time sequence.

A schematic diagram of the timing sequence of the pulses occurring in Fig. 5 is shown in Fig. 6 for a time separation of the input events of 400 nsec. In this latter diagram the number ascribed to each waveform refers to the corresponding points in Fig. 5 at which these waveforms would be observed.

A detailed description of the individual units of the system referred to above is given in the following section.

B. Description of Electronic Circuits

1. Photomultiplier and Limiter Circuit

A circuit diagram of the photomultiplier and limiter circuit is shown in Fig. 7.
FIG. 6. Timing Sequence of Pulses in System for Positron Lifetime Measurements. Time Separation of Input Events is 400 nsec.
FIG. 7. Photomultiplier and Limiter Circuit.
Preliminary positron lifetime measurements in gases were carried out using two RCA-6810 photomultipliers rather than the RCA-7264 photomultipliers shown. With these an overall time resolution of about 6 nsec was obtained. The desirability of better time resolution capabilities soon became apparent during these initial investigations and resulted in the replacement of these tubes by the type RCA-7264. This latter photomultiplier tube has a curved photocathode resulting in extremely small variations in electron transit time between the photocathode and the first dynode and is thus more suited for timing experiments. With this modification a time resolution of $3\frac{1}{2}$ nsec was achieved.

The resistor bleeder chain supplying the dynode potentials was of conventional design with capacitive decoupling between the last few dynode stages. The accelerating electrode and focus control were set to obtain the maximum amplitude dynode pulse and the best limiter pulse rise-time. With the value of the high voltage used for the photomultipliers, these conditions were obtained simultaneously for both tubes.

While the curved photocathode of the RCA-7264 possesses the important feature already noted, the coupling of the crystal to the photocathode presented some difficulty. Since commercially mounted NaI crystals were being used, an optical coupler was made from lucite in the form of a plano-concave lens designed to fit snugly between the photomultiplier and the face of the crystal. Dow Corning (QC-2-0057) grease was used between the various elements to increase the optical transmission from the crystal to the photomultiplier. The complete crystal and tube assembly was then encased in a light tight cylindrical magnetic shield.
NaI(Tl) crystals were used as the detectors because of their much higher efficiency compared to plastic or liquid scintillators, even though better time resolution capabilities are provided by these latter scintillators due to their short decay times. The dimensions of the NaI(Tl) crystals were 1\(\frac{1}{2}\) in. in diameter and 2 in. long. With these crystals a time resolution of 3.6 nsec was obtained (See Appendix B for details) using the 1.28 and 0.51 MeV gamma rays from a Na\(^{22}\) source deposited in aluminum.

Two pulses are derived from the photomultiplier as shown in Fig. 7. A total charge pulse obtained from one of the lower dynodes (dynode No. 9 in the case of the prompt limiter, and dynode No. 11 for the delayed limiter) is fed directly to the amplifiers preceding the pile-up rejector and the single channel analyzer. The charge arriving at the anode of the photomultiplier is collected on the stray capacity at the input of the limiter tube cutting it off quickly, thereby producing the limited pulse at the plate of the tube. Measurements with a sampling oscilloscope indicated limiter pulse risetimes of about 5 nsec for 500 KeV gamma rays and considerably longer risetimes for gamma rays of 100 KeV energy.

A detailed discussion of this type of limiter circuit is given by Jones (1960). The recovery time of the grid circuit of the limiter following detection of a 1.28 MeV gamma ray is about 50 nsec. Consequently the pile-up rejector circuit must make the system inoperative for at least this length of time. In order to maintain the grid circuit recovery time at a reasonable value, the grid of the limiter tube was connected to the positive supply via a 220 K resistor as shown. Under these conditions the tube normally drew about 350 \(\mu\)a of grid current.
leading to a plate current of about 50 ma. The appearance of the actual limiter pulses are shown in Figures 8(a)-(f) for excitation by Na$^{22}$ gamma rays as observed using a Tektronic Type 551 oscilloscope.

2. **Time Sorter**

A comprehensive discussion of the operation and performance of the time sorter is presented in Appendix B. In order to make the time sorter compatible with the ancillary equipment being used, two additional circuits were incorporated. One is the final output stage of the time sorter (the output inverter-driver) required to drive the CDC-100 channel analyzer, and the other the negative time eliminator coincidence circuit. Both circuits are shown in Fig. 9.

As indicated in Fig. 6, the time-to-amplitude converted pulse reaches its maximum before the kick sorter gating pulse occurs. It is thus necessary to delay the converter pulse in order to bring it into suitable time coincidence with the kick sorter gating pulse. The necessary delay is introduced at the input to the output driver stage using a 1.5 $\mu$sec length of HH 2000 delay line. Since the base of transistor $T_1$ is a virtual ground point in the circuit the proper termination of the delay line is provided by the 2.2K resistor. Additional gain within the feedback loop is achieved by boot-strapping the collector load of $T_1$ to the emitter of $T_2$ via the 0.1 $\mu$fd capacitor. Transistors $T_3$ and $T_4$ together form the white amplifier output stage which is used to drive the kick sorter. The overall gain of the output inverter-driver is -0.9.

The discussion in Appendix B indicates that the negative time eliminator produces an output whenever the time sorter input pulses are in the proper time sequence. This output is applied to the input of the
FIG. 8. Voltage Pulses at Various Points in the Circuits of the Limiter and the Pile-up Rejector (PUR) and its Associated Amplifier.
negative time eliminator coincidence circuit of Fig. 9 thus permitting
the entering gate pulse to pass on without interference to the kick
sorter. This is performed in the following manner. T₆ is in effect a
controlled series impedance between the emitter follower T₇ and the
output. In the absence of any signal from the NTE, T₆ is non-conducting
thus representing a very large impedance from collector to emitter. On
the other hand, when a NTE pulse is applied, T₆ is brought into saturation
thus providing a very low impedance path from the collector to the
emitter.

3. Pile-Up Rejector and Amplifier

The effect of pulse pile-up on the shape of the resolution
curve at high counting rates is shown by Schwarzchild (1963) to cause
a broadening of the tail of the resolution curve. A similar result is
indicated in Fig. 56 of Appendix B where the resolution curve is shown
with and without the pile-up rejector in operation.

The pulses used to drive the pile-up rejector (hereafter
referred to as the PUR) are obtained from a dynode of the photomultiplier
as shown in Fig. 5. These pulses have amplitudes of about 50 mV into
an impedance of 100 ohms for 0.51 MeV gamma rays and are insufficient
to operate the PUR directly. Furthermore, it is required that pulses much
smaller than this be able to operate the PUR since these are also able
to trigger the time sorter and contribute to the effective count rate.
The characteristics of the amplifier required for this purpose were a
current amplification of about 400 with a risetime of about 50 nsec.
A two stage feedback amplifier was constructed as shown in Fig. 10, with
the input to each stage forming a virtual earth point. The input
FIG. 10. Amplifier and Pulse Pile-up Rejector.
resistor to the first stage was later experimentally adjusted until an effective input impedance of 100 ohms was obtained. The RC network at the emitter of $T_9$ was empirically designed to yield the short, fast, current pulse required at the output of this amplifier stage.

A limiting action in the second amplifier stage is achieved by making the feedback network nonlinear with the use of a diode at the base of $T_{10}$. At saturation a $2\frac{1}{2}$ V pulse is produced at the emitter of $T_{11}$. Figures 8(g) and (h) show the voltage pulses at each of the amplifier stages. The variable emitter load resistor in $T_{11}$ is used to control the overall gain of the amplifier. A second diode was introduced between the emitter and base of $T_{11}$ to prevent the base voltage from rising above that of the emitter. This would otherwise occur whenever $T_{10}$ was cut off by the negative overshoot present in the input waveform.

The input of the PUR (Fig.10) employs a tunnel diode pulse shaper to produce a fast rising pulse (Fig.8(j)) to drive the pile-up rejection circuit. This latter circuit is similar in design to the one discussed by McGervey and Walters (1964) and produces an output pulse whenever an input pulse is preceded by another input pulse within some preset time interval, $T$. The pulse at the collector of $T_{13}$ (Fig.8(i)) is fed into two series tunnel diodes, both of which are normally in their low voltage state. The bias current of each diode is separately adjustable, the bias current of the first being set slightly higher than that of the second so that, upon receipt of an input pulse, tunnel diode $D_1$ is switched to its high voltage state. Immediately upon switching, the increased voltage at the input of $D_1$ prevents a further
inflow of current from the collector of T_{13}, this current being diverted to
ground via the 390 ohm resistor. If the bias current for D_2 is set at a
lower value than for D_1, the pulse current in D_2 does not reach the peak
current value before the input current is halted by the switching of D_1.
Thus a single isolated input event results in the switching of D_1 but not
of D_2.

The recovery of the tunnel diodes is governed by the time
constants L/R, which have values of about 100 $\mu$sec for both D_1 and D_2.
Recovery from the high voltage state occurs in two stages. Firstly, the
current through D_1 gradually drops to the valley point current from which
the transition to the low voltage state occurs. Immediately following
this transition, the tunnel diode current is still equal to the valley
point current, since the presence of the parallel inductance prevents a
sudden redistribution of the bias current shared by the two elements. Then,
secondly, the current in the tunnel diode builds up to its quiescent value
in a time again determined by the time constant L/R.

The action of the circuit in response to two closely spaced
pulses can now be understood from the foregoing discussion. If the
second input pulse occurs when D_1 is in the high voltage state and
sufficient time has elapsed for the 100 pf capacitor to become charged,
the input current from T_{13} will continue to flow until D_2 regenerates
producing an output pulse. On the other hand, if D_1 has returned to its
low voltage condition but has not yet fully recovered, the second input
pulse will cause a transition in D_2 if its bias current is greater than
the recovering current in D_1. In either case then, an output from D_2
occurs, thus indicating a pile-up event. The output from tunnel diode
D_2 is shown in Fig.8(k). By suitably adjusting the bias currents in the
two tunnel diodes, proper operation of the circuit can be achieved for a wide range of effective "dead times". Taking into consideration the recovery time of the limiters, the pile-up rejectors were set to register a pile-up event if the interval between pulses was less than 50 $\mu$sec. Using pulses from a scintillation counter, proper operation of the circuit was confirmed by measuring the input and output counting rates of the pile-up rejector. The circuit as shown in Fig.10 is incapable of registering pile-up events separated by less than 1 $\mu$sec because of the 1 $\mu$sec width of the pulse at the base of $T_{12}$. An attempt to decrease this pulse width produced difficulty with double triggering on a single input pulse from the scintillation counter.

4. Anticoincidence Circuit

The outputs of the two pile-up rejectors are fed into the anticoincidence circuit shown in Fig.11 where the occurrence of a pulse from either PUR blocks the incoming gate pulse. This action is achieved by allowing either PUR pulse to saturate its respective 2N706A transistor, thus producing a low impedance shunt between the base of $T_{15}$ and ground. The gate pulse is highly attenuated as a result, and no output is produced from the emitter follower $T_{15}$. In the absence of a pulse from either PUR, on the other hand, the gate pulse is faithfully reproduced at the emitter of $T_{15}$.

5. Slow Coincidence Circuit and Gate

The coincidence circuit which registers coincidence events from the energy selecting single channel analyzers is shown in Fig.11. A coincidence output triggers a monostable circuit to generate at 4.5 $\mu$sec, 18 V gating pulse for operating the multichannel analyzer.
FIG. 11. Anticoincidence Circuit and Slow Coincidence Circuit and Gate.
6. **Ancillary Equipment**

The following contains a brief discussion of some of the equipment not considered in detail in the foregoing sections. Firstly, it will be noted from Fig. 6 that the resolving time of the slow coincidence circuit must be at least as long as the time interval over which it is desired to measure the delayed annihilation events. This in turn, is governed by the width of the output pulses from the single channel analyzers. To insure proper operation, this resolution time was made about 1 μsec in the present experiment involving time measurements up to 600 nsec. Details of the single channel analyzers, except for revisions to the amplifier sections, have been given by Jones (1959).

The multichannel analyzer used in the experiments was a Computing Devices of Canada Ltd. 100-channel kicksorter (Type AEP2230). Experimental investigation of the gating requirements of this kicksorter indicated that a pulse of amplitude 8-18 V (positive) opened the gate fully and a negligible time delay (≤ 0.1 μsec) was required from the start of the input gate until the “fully open” state was achieved.

C. **Pressure Chambers**

In the various experiments reported in this thesis a total of three different pressure chambers were employed to obtain the positron annihilation spectra. The general arrangement used was to place the Na\(^{22}\) source in the geometrical centre of the chamber and position the two counters next to the chamber walls for maximum counting efficiency.

The general considerations that were taken into account in the design for a particular experiment were as follows:

(i) Firstly, of course, it was required that the chamber be
able to withstand the gas pressures encountered in the experiment.

(ii) On the other hand, it is desirable to have thin chamber walls because of the absorption of gamma rays. The resulting reduction of the coincidence counting rate is clearly undesirable. It is useful to define a "figure of merit", $\varepsilon$, for a particular metal as

$$\varepsilon = \mu \varphi / s$$

where $\mu$ is the gamma ray absorption coefficient (cm$^2$/gm), $\varphi$ is the density, and $s$ the maximum permissible working stress of the metal. For gamma ray energies between 200 - 500 KeV $\varepsilon$ has approximately the same value for stainless steel as for a "good" aluminum alloy. At energies lower than this the rapidly increasing photoelectric effect in iron results in much greater absorption than for aluminum. Aluminum is thus to be slightly preferred as a constructional material for these chambers.

(iii) For a high counting efficiency the placement of the counters relative to the source must be optimized and the physical shape of the chamber must allow for this. At the same time, counter to counter scattering must be kept to a minimum by adequate shielding of the counters. The deleterious effect of such scattering has been described in Appendix B.

(iv) In some of the experiments, measurements of the component intensities were made from the recorded spectra. To make such measurements possible the number of positrons annihilating in
materials other than the gas under study must be kept to an absolute minimum. In order to prevent annihilations with the walls of the chamber the size of the chamber must be sufficiently large so that for a particular gas and a given pressure all the positrons will reach the end of their range in the gas, rather than collide with the chamber walls. Calculations relating the range of positrons in various gases to their energy and the pressure of the gas are presented in Appendix E. A brief description of the three pressure chambers follows.

Preliminary experiments were all carried out in an aluminum cylindrical chamber of 2.8 litre volume. Fig. 12(a) shows a cross sectional view of this chamber indicating the essential dimensions. The size of this chamber restricted its use to cases where wall annihilation was not of primary consideration.

In argon the maximum range of positrons from the beta-decay of Na$^{22}$ is about 160 cm atm (Appendix E). For the experiments referred to earlier involving minimum wall annihilation, one thus requires a "free" spherical region of 6-10 in. radius surrounding the source, for experiments in the pressure range of 6-10 atm. The chamber constructed for this purpose was an aluminum sphere, made by welding together two 14 in. diameter aluminum hemispheres (spun by Spin Craft Mfg. Co., Van.). A diagram of this chamber is given in Fig. 12(b).

The desirability of carrying out measurements of the annihilation spectra of positrons in helium was pointed out in Chapter II. Experimentally, the requirements imposed by a gas with a density as low as that of helium are rather severe since the maximum range of Na$^{22}$ positrons in
FIG. 12. Cross Sectional View of Cylindrical and Spherical Pressure Chambers

(a) Cylindrical Pressure Chamber
- Scale: ½ Full Size
- Material: Dural

(b) Spherical Pressure Chamber
- Scale: ¼ Full Size
- Material: 2S, Half Hard Al.
this gas is about 1200 cm atm (Appendix E), or about 7 in. at a pressure of 1000 psi. A working compromise was a chamber designed to withstand pressures of up to 1000 psi and with an active volume, cylindrical in shape, of $9\frac{1}{2} \times 12$ in. The significant feature of this chamber was an elaborate grid structure with which to provide a uniform electric field over the large active volume up to a maximum of about 1500 V/cm. Thus in addition to the studies on helium, this chamber served for electric field experiments on other gases. A cross sectional view of this chamber is shown in Fig.13 and photographs of the actual chamber in various stages of assembly are shown in Fig.14. Details of the grid structure to provide the uniform electric field are clearly visible in this latter figure. The design calculations and further constructional details of the chamber are presented in Appendix A.

Table II summarizes the pertinent details of the three pressure chambers discussed in the foregoing paragraphs.
### TABLE II

Summary of the Characteristics of the Various Pressure Chambers

<table>
<thead>
<tr>
<th>Chamber</th>
<th>Construction</th>
<th>Hydrostatic Test Pressure psi (gauge)</th>
<th>Maximum Operating Pressure psi (gauge)</th>
<th>Chamber Volume in Litres</th>
<th>Calculated Wall Absorption of 500 kev Gamma Rays</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cylindrical Chamber</td>
<td>Aluminum (Dural)</td>
<td>-</td>
<td>200</td>
<td>2.8</td>
<td>13.5%</td>
</tr>
<tr>
<td>Spherical Chamber</td>
<td>Aluminum (2S half hard)</td>
<td>200</td>
<td>150</td>
<td>23.8</td>
<td>5.5%</td>
</tr>
<tr>
<td>High Pressure Chamber</td>
<td>Aluminum (See App. A)</td>
<td>1700</td>
<td>1000</td>
<td>23.6</td>
<td>16%</td>
</tr>
</tbody>
</table>
FIG. 13. Cross Sectional View of High Pressure Chamber.
FIG. 14. Photographs of High Pressure Chamber in Various Stages of Assembly.
CHAPTER IV

EXPERIMENTAL PROCEDURE AND PARTICULARS

A. Gases and Gas Purification

The annihilation of positrons was studied in a number of monatomic and polyatomic gases for the work described in this thesis. A table of these gases and their purity is given in Table III. The suppliers of these gases were The Matheson Co. of Canada and The Canadian Liquid Air Co.

A crucial factor in many of the experiments was that of gas purity as explained in Chapter II. This was particularly so in the case of the noble gases where an investigation of the velocity-dependent annihilation rate was being made. In general, the sensitivity of an experiment to a particular impurity in the gas depended on the nature of the experimental quantity being investigated. For example, the direct annihilation rates in oxygen and nitrogen are comparable, with the result that an impurity of $N_2$ in oxygen will not introduce any significant error in the measured direct rate in oxygen. An impurity of $O_2$ when added to nitrogen however, will, because of its strong orthopositronium quenching characteristics, greatly alter the ortho lifetime observed in the nitrogen. The results of Paul and Saint-Pierre (1963) on the direct annihilation rates in the hydrocarbons indicates the deleterious effect of these gas impurities in measurements of the direct rate in other gases. In view of this, several steps were carefully followed in assuring that effects of impurities in the gases under study were kept to a minimum.
(i) Firstly, efforts were made to procure the purest gases available whenever this was not prohibitively costly.

(ii) Great care was exercised in the cleansing and evacuation of the pressure chambers prior to gas filling.

(iii) For the detailed direct lifetime studies in argon and helium a gas purifier was used in conjunction with the pressure chamber to remove any residual impurities.

The purities of the gases, as quoted by their supplier are shown in Table III. For the diatomic gases no further effort was made to reduce the impurities and the gases were used as supplied. In the case of argon, mass spectrometer analysis were carried out on gas samples taken both from the gas cylinder and from the experimental chamber after purifying for several days. These results are also shown in the table and demonstrate the effectiveness of the gas purifier.

Prior to the gas filling of a pressure chamber, the chamber was cleaned with acetone and then baked at 250°F for about 24 hours. Neoprene O-rings were likewise cleaned and baked and on reassembly, vacuum grease was applied sparingly. Further baking of the assembled chamber was carried out with the chamber under vacuum until an equilibrium pressure was reached. Ultimate pressures of about $3 \times 10^{-6}$ mmHg were attained before the chamber was filled with the gas.

The purification of the gases argon and helium was carried out using the method described by Colli and Facchini (1952). A eutectic mixture consisting of 79% Ca and 21% Mg and contained in a stainless steel tube was fixed to the pressure chamber (see Appendix A) and was electrically heated to a temperature of 400°C. At this temperature
### TABLE III

**Gas Purities**

<table>
<thead>
<tr>
<th>Gas</th>
<th>Supplier</th>
<th>Quoted Purity(%)</th>
<th>Main Impurities</th>
<th>Mass Spectrometer Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>N₂</td>
<td>Matheson</td>
<td>99.996</td>
<td>O₂</td>
<td>N₂(%) 0.30  O₂(%) 0.00  CO₂(%) 0.03</td>
</tr>
<tr>
<td>O₂</td>
<td>Can. Liq. Air</td>
<td>99.5</td>
<td>N₂, A, H₂</td>
<td>Sample #1d 0.30 0.00 0.03  Sample #2e 0.00 0.00 0.00</td>
</tr>
<tr>
<td>CO₂</td>
<td>Can. Liq. Air</td>
<td>99.9</td>
<td>N₂, CO</td>
<td></td>
</tr>
<tr>
<td>He</td>
<td>Can. Liq. Air</td>
<td>99.997</td>
<td>Ne, N₂, O₂</td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>Can. Liq. Air</td>
<td>99.98</td>
<td>N₂, O₂, H₂</td>
<td></td>
</tr>
<tr>
<td>Kr</td>
<td>Matheson</td>
<td>99.99</td>
<td>Xe, N₂</td>
<td></td>
</tr>
</tbody>
</table>

a Prepurified Grade — typical oxygen content of 8 parts per million.
b Matheson of Canada, Ltd.
c Canadian Liquid Air Company, Ltd.
d Gas sample taken from gas cylinder.
e Gas sample taken from high pressure chamber after purifying for several days. The limit of sensitivity of the spectrometer for detecting the gases N₂, O₂, and CO₂ was about 0.02%. 
impurities of $N_2$, $CO_2$, $H_2$, and $O_2$ are quickly removed by the eutectic mixture. Prior to its use, outgassing and conditioning of the eutectic was carried out by heating it to 550°C under vacuum until all the entrapped gases had been driven off.

B. Positron Sources

A standard source for positron lifetime measurements is Na$^{22}$ which, with the useful half-life of 2.58 years, also satisfies the requirement of providing a positron "birth" signal in the form of a nuclear de-excitation gamma ray. Na$^{22}$ decays to the first excited state of Ne$^{22}$ - 10% via electron capture, and 90% via positron emission (Nuclear Data Tables, Part 4, U.S. Atomic Energy Commission, 1960). The transition from the excited state to the ground state of Ne$^{22}$ occurs in less than $10^{-11}$ sec (Alkhazov et al (1959)) via a 1.28 MeV gamma ray, essentially simultaneously with the positron, for our purposes.

The Na$^{22}$ activity used in the present experiments was obtained as a NaCl solution (from "The Radiochemical Centre", Amersham, England) with the high specific activity of 1.65 mc/mgm of Na. Sources were prepared by evaporating a small amount of the solution placed on thin aluminum sheets, $3 \times 10^{-5}$ in. in thickness. The thin aluminum sheets were supported by mounting them on a thin wire loop ($\frac{1}{2}$ in. diameter) or on an aluminum frame (.005 in. thickness) containing a $\frac{1}{2}$ in. diameter hole. From a consideration of the range of positrons in aluminum and the shape of the beta-spectrum from Na$^{22}$ it was estimated that about 2% of the positrons would annihilate in the source.

The factors determining the optimum source strength for use in a particular experiment depend on the experimental quantity being
investigated and on the overall counting efficiency of the system. Since the ratio of the random coincidences to the true coincidences is directly proportional to the source strength it is desirable to keep the source strength low. On the other hand, in order to obtain reasonable counting statistics in a practical length of time some compromise has to be made. Table IV shows the source strengths used for the three different chambers and also indicates the pertinent counting rate data under typical experimental situations.

C. Electronic Calibrations and Instrumental Arrangements

1. Counter Geometry

The effect of counter-counter scattering of gamma rays on the shape of the time resolution curve is pointed out in Appendix B. For this reason efforts were made to shield the counters from each other using lead shields while at the same time retaining good counter geometry. The geometrical arrangement of counters and chambers is shown in Fig. 15 for both the spherical chamber and the high pressure chamber. In the former case the shielding provided by the lead rings is only partial, but the 10 in. separation between the counters reduces to a large extent the effect of scattering. A similar geometry was employed for the cylindrical chamber as for the spherical chamber with the two lead shields replaced by a single lead block situated between the counters.

2. Calibration of Time Sorter Time Scale

A general discussion of the method used in calibrating the time scale of the time sorter is presented in Appendix B. Using the double pulse generator described in Appendix D an accurate integral calibration was obtained which yielded the overall length of the time scale. The
### TABLE IV

Source Strengths and Counting Rates Under Typical Experimental Conditions

<table>
<thead>
<tr>
<th>Source Strength (calculated from true-to-random coincidence rate)</th>
<th>Cylindrical Chamber (^a)</th>
<th>Spherical Chamber (^b)</th>
<th>High Pressure Chamber (^c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Positron Annihilations in Source</td>
<td>0.83 (\mu)C (Dec. 7/63)</td>
<td>4.6 (\mu)C (Aug. 24/64)</td>
<td>15.6 (\mu)C (Sept. 5/64)</td>
</tr>
<tr>
<td>Total Coincidence Counting Rate from Time Sorter (Counts/hr)</td>
<td>1230</td>
<td>1090</td>
<td>900</td>
</tr>
<tr>
<td>Random Coincidence Background Counting Rate (counts per hour per channel at 2.43 nsec/channel)</td>
<td>0.10</td>
<td>0.53</td>
<td>1.48</td>
</tr>
</tbody>
</table>

Counting rates in the Individual Counters:

(i) 1.28 MeV Single Channel Analyzer Output (counts per second)\(^d\)

(ii) 0.51 MeV Single Channel Analyzer Output (counts per second)\(^d\)

(iii) Triggering Rate of the Prompt Trigger (counts per second)

(iv) Triggering Rate of the Delayed Trigger (counts per second)

\(^a\) Counting rates quoted are for Argon at 8.3 atm (25°C)

\(^b\) Counting rates quoted are for oxygen at 10.6 atm (25°C)

\(^c\) Counting rates quoted are for Argon at 10.5 atm (25°C)

\(^d\) The energy windows of the single channel analyzers were set so that one analyzer spanned the 510 keV peak and the other the region between 0.6 to 1.4 MeV (see details in this chapter).
FIG. 15. Counter - Chamber Geometry for the Spherical Chamber and the High Pressure Chamber. Scale: 1/5 Full Size.
relative time widths of the individual channels were then measured using
the random time generator (Appendix C) in the arrangement shown in
Fig. 16(a). From the results obtained in these two independent cali-
brations an accurate absolute time width could be assigned to each
channel. This channel width data and the method of using this information
in the analysis of the results is presented in Appendix F.

3. Photomultiplier High Voltage and Trigger Circuit Sensitivity

With the high gain 14-stage photomultipliers employed it was
found that relatively low voltages had to be used if noise pulses were
not to be excessive in number and amplitude. The high voltages were
adjusted so that the largest noise pulse yielded pulses at the anode of
the limiter tube about 1/3 of the amplitude of the limited pulse. This
required values of -1500 and -1900V for the 0.51 MeV and the 1.28 MeV
gamma counter respectively. The sensitivity of the discriminators at
the input to the trigger circuits was then adjusted until only the larger
amplitude noise pulse were able to trigger the circuits. Triggering of
the circuits by some of the noise pulses was tolerated since it was
found that decreasing the sensitivity till all noise pulses were elimi-
nated resulted in a marked increase in the width of the resolution
curve.

4. Pile-Up Rej ector Sensitivity

In order to gain the maximum benefit from the PUR it must
respond to all pulses causing any appreciable dead time in the other
parts of the system. Very small noise pulses produced a negligible
dead time in the limiter circuit while the larger ones were able to
operate the trigger circuits. Thus the PUR sensitivity was matched to
that of the input threshold discriminators of the time sorter.

5. **Single Channel Analyzer Spectra**

With few exceptions, all the experiments described in this thesis were directed at an investigation of the direct annihilation rates of positrons. Since this component in the decay spectrum is predominantly characterized by 2-quantum annihilation, the single channel analyzer in the delayed channel was set on the 510 keV annihilation peak with an energy window of about 150 keV. In the prompt channel, only gamma rays originating from the 1.28 MeV transition were selected since only these correspond to the production of a positron. The portion of the energy spectrum selected by each of the single channel analyzers is shown in Fig. 17.

To facilitate the monitoring of the single channel analyzer settings a gating arrangement was devised to display on the kicksorter the portion of the spectrum selected by the analyzer. The arrangement is shown in Fig. 16(b). An inherent delay of about 0.7 μsec was present in the single channel analyzer output so that the signal to the kicksorter had to be delayed by about 2 μsec.
FIG. 16. (a) Experimental Arrangement to Check Differential Linearity of TS Using the Random Time Generator.

(b) Arrangement for Displaying the Gated Energy Spectrum.
FIG. 17. Na$^{22}$ Spectra Showing the Portion of the Spectrum Selected by Each of the Single Channel Analyzers.
CHAPTER V

PRESENTATION OF RESULTS

A. Selection of Gases for Study

As discussed in Chapter II, relatively little information is available on the direct annihilation rate of positrons in gases and only general qualitative features concerning the annihilation process have been established. It was further pointed out that recent experiments in argon have indicated an annihilation process in the noble gases more complex than expected. Thus the main objective of this work has involved a detailed study of the direct annihilation of positrons in argon and preliminary measurements have been obtained for the other noble gases, krypton and helium.

The lack of experimental data for the direct annihilation rates in polyatomic gases also motivated an investigation of several such gases. The choice of gases was largely determined by their availability in relatively pure form and at a reasonable cost, the gases nitrogen, oxygen, and carbon dioxide being selected.

B. Data Analysis

The experimental data obtained during an experiment with a particular gas consisted of a series of annihilation spectra recorded at different pressures and on different time scales. The time required to obtain a spectrum with adequate statistics was typically between 20 and 50 hours depending on the positron source strength and the time scale used. Since an investigation of the direct annihilation rate was the main object of these experiments, the orthopositronium component was
deliberately suppressed in the recording of the spectra as described in Section C of Chapter IV. It was estimated that the relative detection efficiency of a 3-quantum annihilation event to a 2-quantum event was typically about $1/2$.

A representative spectrum for carbon dioxide at a pressure of 5.29 atm is shown in Fig.18. The points shown indicate the raw data obtained from the kicksorter. The general features of this spectrum can be described as follows:

(i) The 'prompt peak' indicated on the figure represents events which occur simultaneously (within the instrumental resolution time) with the emission of the 1.28 MeV gamma ray and arise almost entirely from the annihilations of para-positronium atoms. Positron annihilations occurring within the source and in the chamber walls contribute the remainder of the counts in the prompt peak.

(ii) The region to the right of the prompt peak consists of two components, one representing direct annihilations and the other the annihilations from orthopositronium. In Fig.18 these two components are well separated and both are exponential in character.

(iii) Superimposed on the spectrum is a constant background consisting of random coincidence events.

The information to be derived from such curves consists of the time constants and intensities of the separate decay processes. To accomplish this the following procedure was adopted:

(i) Firstly, because of the nonlinearity of the time scale
(ie. unequal channel widths) the raw data was subjected to a linearization procedure as described in Appendix F.

(ii) From this linearized data the background was determined by averaging the counts to the left of the prompt peak position (See Appendix B).

(iii) A four parameter fit to the two exponential decay components was then performed on an IBM 7040 computer using the points to the right of the prompt peak position.

A description of the Maximum Likelihood program used for this four parameter fit is presented in Appendix F.

In the data presented in the following sections all the curves that are shown have had the time scales linearized and the random coincidence background subtracted.

C. Direct Annihilation Rates in Polyatomic Gases

1. Nitrogen

Typical annihilation spectra obtained in nitrogen at pressures of 11.2 and 5.08 atm respectively are shown in Figures 19 and 20. As indicated by the relative intensity of the prompt peak, relatively little positronium is formed in nitrogen. The increase in the intensity of the prompt peak in Fig.20 is due to the increased amount of wall annihilation at this lower pressure. The associated low intensity of the ortho-positronium component resulted in considerable difficulty in separation of the direct and ortho component. This difficulty could be largely overcome by using a three parameter fit to the data keeping the value of the orthopositronium lifetime fixed. The values for this lifetime were obtained from the results of Celitans and Green (1964) and Celitans
et al (1964) on the orthopositronium quenching rate in nitrogen. Their results can be represented equally well by either of the expressions

\[
\frac{\lambda(P)}{\lambda_o} = 0.894 + 0.029P \tag{1}
\]

or

\[
\frac{\lambda(P^2)}{\lambda_o} = 1.071 + 0.00102P^2 \tag{2}
\]

over the pressure range of interest, where \( \lambda \) is the observed orthopositronium annihilation rate, \( \lambda_o \) is the orthopositronium annihilation rate without quenching, and \( P \) is the gas pressure in atm (at 25°C). The value of \( \lambda_o \) used here is \( 7.21 \times 10^6 \) sec, obtained from the theoretical orthopositronium mean lifetime of 138.6 nsec given by Jauch and Rohrlick (1955). In the calculations, values for the ortho lifetime intermediate between the values given by the two expressions, were used. These values are indicated graphically in Fig. 21 together with the expressions (1) and (2). Fortunately, because of the small intensity of this component, the exact value selected for the lifetime had only a very small effect on the calculated mean life of the direct component. For example, for the spectrum of Fig. 19, a 35% change in the ortho lifetime resulted in only a 1.5% change in the calculated direct lifetime. Thus the spread of the points shown in Fig. 21 represents the range of values for the orthopositronium lifetime for which the calculated direct lifetime was approximately constant.

The calculation of the direct lifetimes for the spectra shown in Figures 19 and 20 used the experimental points in the region below channels 89 and 91 respectively as indicated by the vertical arrows in these figures.

The results for nitrogen are summarized in Table V. The
annihilation rate, which is equal to the reciprocal of the mean lifetime, is given in column 7 of this table and is plotted as a function of the pressure in Fig. 22. A least squares straight line fit to these points yields an annihilation rate, \((5.52 \pm 0.02) \times 10^6 \text{sec}^{-1} \text{atm}^{-1}\). In this latter calculation it has been assumed that the origin is a valid point on the line, since at zero gas pressure the direct annihilation rate should be zero. The excellence of the fit to these points indicates that in nitrogen the direct annihilation rate is directly proportional to the pressure within the accuracy of the experimental data.

For comparison, the annihilation rate given by the Dirac formula, equation (2) of Chapter II, has also been calculated and is shown by the dashed line in Fig. 22. Assuming that all the electrons of the atom participate in the annihilation process according to the Dirac cross section, the annihilation rate can be written as

\[
\Lambda_a = 1.844 \times 10^5 n_e P
\]

(3)

where \(n_e\) is the number of electrons per atom (or molecule) and \(P\) is the gas pressure in atm at 25°C.

2. Oxygen

Oxygen is known to quench orthopositronium strongly, as shown by Deutsch (1953), Heymann et al (1961), and Celitans et al (1964). At pressures of several atmospheres essentially all the orthopositronium events decay via two-quantum emission. The spectrum shown in Fig. 23 substantiates these findings, indicating the highly quenched orthopositronium as the shorter of the two exponential components. Although the lifetime of the quenched orthopositronium component could not be accurately determined from the recorded annihilation spectra, the
### TABLE V

Summary of N$_2$, O$_2$, and CO$_2$ Spectra and the Measured Direct Annihilation Mean Lifetimes, $\tau_a$. All these Spectra Were Recorded Using the Spherical Pressure Chamber Described in Chapter III. The Errors Indicated Represent the Statistical Errors Only.

<table>
<thead>
<tr>
<th>Gas Number</th>
<th>Spectrum Time per Channel (nsec)</th>
<th>Counting Time per Channel (hours)</th>
<th>Background Time per Channel (nsec)</th>
<th>$\lambda_a$ ($10^7$/sec)</th>
<th>Pressure in atm</th>
<th>Relative gas Density$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>N$_2$</td>
<td>2</td>
<td>2.43</td>
<td>4.00</td>
<td>25</td>
<td>16.2±0.13</td>
<td>6.18±0.051</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>4.68</td>
<td>25.42</td>
<td>24</td>
<td>16.3±0.15</td>
<td>6.14±0.058</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>2.43</td>
<td>46.17</td>
<td>31</td>
<td>19.6±0.10</td>
<td>5.09±0.026</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>4.68</td>
<td>24.08</td>
<td>28</td>
<td>20.5±0.23</td>
<td>4.88±0.056</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>2.43</td>
<td>43.08</td>
<td>29</td>
<td>25.3±0.13</td>
<td>3.95±0.021</td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>4.68</td>
<td>23.25</td>
<td>26</td>
<td>26.3±0.22</td>
<td>3.80±0.031</td>
</tr>
<tr>
<td></td>
<td>8</td>
<td>2.43</td>
<td>48.08</td>
<td>28</td>
<td>34.9±0.19</td>
<td>2.86±0.015</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td>4.68</td>
<td>23.83</td>
<td>26</td>
<td>35.5±0.29</td>
<td>2.82±0.023</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>2.43</td>
<td>47.75</td>
<td>25</td>
<td>60.7±2.4</td>
<td>1.65±0.066</td>
</tr>
<tr>
<td>O$_2$</td>
<td>24</td>
<td>2.43</td>
<td>43.83</td>
<td>23</td>
<td>19.9±0.16</td>
<td>5.03±0.041</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>2.43</td>
<td>64.50</td>
<td>34</td>
<td>25.9±0.41</td>
<td>3.87±0.062</td>
</tr>
<tr>
<td></td>
<td>26</td>
<td>2.43</td>
<td>36.67</td>
<td>23</td>
<td>31.6±0.32</td>
<td>3.16±0.032</td>
</tr>
<tr>
<td></td>
<td>27</td>
<td>2.43</td>
<td>53.92</td>
<td>33</td>
<td>50.1±0.73</td>
<td>1.99±0.029</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>13</td>
<td>2.43</td>
<td>48.00</td>
<td>29</td>
<td>6.42±0.17</td>
<td>15.6±0.41</td>
</tr>
<tr>
<td></td>
<td>14</td>
<td>2.43</td>
<td>49.33</td>
<td>31</td>
<td>8.36±0.32</td>
<td>12.0±0.14</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>2.43</td>
<td>46.50</td>
<td>25</td>
<td>11.5±0.15</td>
<td>8.72±0.11</td>
</tr>
<tr>
<td></td>
<td>16</td>
<td>4.68</td>
<td>22.42</td>
<td>25</td>
<td>11.7±0.33</td>
<td>8.58±0.25</td>
</tr>
<tr>
<td></td>
<td>17</td>
<td>2.43</td>
<td>48.67</td>
<td>30</td>
<td>17.2±0.17</td>
<td>5.83±0.058</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>4.68</td>
<td>27.83</td>
<td>33</td>
<td>16.9±0.29</td>
<td>5.93±0.10</td>
</tr>
<tr>
<td></td>
<td>19</td>
<td>2.43</td>
<td>44.00</td>
<td>24</td>
<td>29.9±0.24</td>
<td>3.34±0.027</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>4.68</td>
<td>27.00</td>
<td>24</td>
<td>30.0±0.40</td>
<td>3.33±0.044</td>
</tr>
</tbody>
</table>

$^a$The density of CO$_2$ at 25°C and a pressure of 1 atm was taken as unity. The density as a function of pressure was obtained from the 'Handbook of the American Inst. of Physics', second edition, McGraw-Hill, 1963.
estimated value was in agreement with the result of Celitans et al (1964), who found a quenching rate, \( \lambda_q = 3.1 \lambda_0 / \text{atm} \). For this result, \( \lambda_0 = 7.21 \times 10^6 / \text{sec} \), and the pressure is measured at 25°C.

With the ortho component well separated from the direct component, the lifetime of the latter could be readily determined using a two-parameter fit to the data. In order to check that essentially no orthopositronium component was contributing to the region of the spectrum used for calculating the direct lifetime, \( \tau_a \), several analyses of each spectrum were carried out with each analysis begun at a different channel.

A summary of the results for the direct component in oxygen is given in Table V. The annihilation rate as a function of the pressure is shown in Fig. 22, from which one obtains, by a least squares fit, the rate, \((4.70 \pm 0.06) \times 10^6 \text{ sec}^{-1} \text{ atm}^{-1}\).

3. Carbon Dioxide

Several annihilation spectra of positrons in CO₂ are shown in Figures 24 and 25. For carbon dioxide, no detailed investigations of the quenching rate of orthopositronium have been reported, and only an approximate value of \( \lambda_q = 0.08 \lambda_0 / \text{atm} \) has been given by Heymann et al (1961). In the analyses of the spectra for CO₂ it was thus necessary to determine both the direct and the ortho lifetime. The results for the orthopositronium rate are shown in Fig. 21. Although these results do not preclude the possibility of a nonlinear pressure dependence of this annihilation rate, an approximate fit to the points is obtained using the orthopositronium quenching rate, \( \lambda_q = 0.095 \lambda_0 / \text{atm} \). Using values for the orthopositronium lifetime calculated from this quenching rate,
the direct annihilation lifetimes were recalculated from the experimental data. In all cases but one, the lifetimes calculated in this way agreed with the initial values to within 1%. The only exception was the spectrum taken at a pressure of 3.04 atm where the difference was about 5%. The results of these two calculations were averaged and are presented in Fig. 26. Here, instead of plotting the direct annihilation rate against the pressure, the actual gas density has been used, since the behavior of CO₂ deviates considerably from that of an ideal gas even at pressures as low as 10 atm. The result is somewhat surprising since, even with this correction, a very marked deviation from a linear density dependence is still observed. For the two lowest pressures, a straight line through these points yields an annihilation rate, 1.07 x 10⁷ sec⁻¹ atm⁻¹. Most of the points within the pressure range examined can be fitted, however, by a linear plus a quadratic dependence,

\[ \lambda_a = (0.852 \rho + 0.040 \rho^2) \times 10^7 / \text{sec} \]

where \( \rho \) is the density of the gas, arbitrarily defined as unity at a pressure of 1 atm and 25°C.

D. Direct Annihilation Rate in Argon

1. Introduction

The positron annihilation spectra in argon were obtained using the high pressure chamber described in Chapter III. At a gas pressure of 10.5 atm (25°C) a detailed investigation of the dependence of the direct lifetime on the electric field was made over a range of electric field strengths from 0 to 1280 V/cm. A summary of the relevant details for these spectra is shown in Table VI. The data recorded for each of these spectra are tabulated in Appendix H.
### TABLE VI

Summary of Particulars on Argon and Helium Spectra. All Spectra were Recorded on the 2.43 nsec/ch Scale Except Where Noted Otherwise.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Spectrum</th>
<th>Pressure in atm(25°C)</th>
<th>Electric Field V/cm</th>
<th>Counting Time (Hours)</th>
<th>Background Counts per Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>29</td>
<td>10.5</td>
<td>0</td>
<td>52.10</td>
<td>77</td>
</tr>
<tr>
<td>A</td>
<td>30</td>
<td>10.5</td>
<td>682</td>
<td>44.50</td>
<td>62</td>
</tr>
<tr>
<td>A</td>
<td>31</td>
<td>10.5</td>
<td>329</td>
<td>47.75</td>
<td>74</td>
</tr>
<tr>
<td>A</td>
<td>32(^a)</td>
<td>10.5</td>
<td>0</td>
<td>24.83</td>
<td>73</td>
</tr>
<tr>
<td>A</td>
<td>33</td>
<td>10.5</td>
<td>0</td>
<td>44.33</td>
<td>67</td>
</tr>
<tr>
<td>A</td>
<td>35</td>
<td>10.5</td>
<td>158</td>
<td>65.17</td>
<td>97</td>
</tr>
<tr>
<td>A</td>
<td>37</td>
<td>10.5</td>
<td>493</td>
<td>48.17</td>
<td>79</td>
</tr>
<tr>
<td>A</td>
<td>38</td>
<td>10.5</td>
<td>0</td>
<td>40.17</td>
<td>59</td>
</tr>
<tr>
<td>A</td>
<td>39</td>
<td>10.5</td>
<td>979</td>
<td>45.58</td>
<td>57</td>
</tr>
<tr>
<td>A</td>
<td>40</td>
<td>10.5</td>
<td>1280</td>
<td>50.08</td>
<td>44</td>
</tr>
<tr>
<td>A</td>
<td>42</td>
<td>7.65</td>
<td>0</td>
<td>68.58</td>
<td>101</td>
</tr>
<tr>
<td>A</td>
<td>44</td>
<td>5.22</td>
<td>0</td>
<td>65.08</td>
<td>87</td>
</tr>
<tr>
<td>A + 0.1% N(_2)</td>
<td>45</td>
<td>5.22</td>
<td>0</td>
<td>71.70</td>
<td>80</td>
</tr>
<tr>
<td>A + 0.8% N(_2)</td>
<td>46</td>
<td>5.26</td>
<td>0</td>
<td>41.58</td>
<td>52</td>
</tr>
<tr>
<td>A + 2.7% N(_2)</td>
<td>48</td>
<td>5.36</td>
<td>0</td>
<td>58.00</td>
<td>69</td>
</tr>
<tr>
<td>He</td>
<td>52</td>
<td>49.6</td>
<td>1330</td>
<td>44.67</td>
<td>52</td>
</tr>
<tr>
<td>He</td>
<td>53</td>
<td>49.6</td>
<td>0</td>
<td>48.00</td>
<td>52</td>
</tr>
<tr>
<td>He</td>
<td>54</td>
<td>59.7</td>
<td>0</td>
<td>58.75</td>
<td>67</td>
</tr>
<tr>
<td>He</td>
<td>55</td>
<td>59.7</td>
<td>1360</td>
<td>58.25</td>
<td>60</td>
</tr>
<tr>
<td>He</td>
<td>56(^a),(^b)</td>
<td>59.7</td>
<td>0</td>
<td>41.83</td>
<td>348</td>
</tr>
<tr>
<td>He</td>
<td>57</td>
<td>59.7</td>
<td>296</td>
<td>40.12</td>
<td>48</td>
</tr>
<tr>
<td>He</td>
<td>59</td>
<td>20.6</td>
<td>283</td>
<td>78.00</td>
<td>115</td>
</tr>
<tr>
<td>He</td>
<td>60</td>
<td>20.6</td>
<td>0</td>
<td>68.62</td>
<td>93</td>
</tr>
<tr>
<td>He</td>
<td>61</td>
<td>20.6</td>
<td>1330</td>
<td>95.12</td>
<td>122</td>
</tr>
</tbody>
</table>

\(^a\)These spectra recorded on the 4.68 nsec/channel scale.

\(^b\)This spectrum was recorded with the single channel analyzer in the 500 keV channel set to accept gamma ray energies between 100-570 keV.
Figures 28-32 show representative annihilation spectra in argon at pressures of 5.22, 7.65, and 10.5 atm. Following the procedure outlined in Section B, these data have been linearized and the background subtracted before plotting. Many features of these spectra are similar to those obtained for the polyatomic gases N₂ and CO₂ but significant quantitative differences are evident. Firstly, the region to the right of the prompt peak exhibits a prominent 'shoulder' for the zero field case (Fig.28), preceding the more common exponential decay. The width of this 'shoulder' increases as the pressure is reduced as observed in Fig.29. Secondly, Figures 30-32 reveal a decrease in the prominence of this 'shoulder' at increasing electric fields. As in the case of the polyatomic gases, the presence of positronium formation is clearly indicated by the magnitude of the prompt peak and the long lifetime component evident in the 'tails' of the spectra.

An interpretation of these observations was given in Section E of Chapter II attributing the presence of the shoulder to a velocity dependence of the annihilation rate of free positrons in the gas. Support for this interpretation is provided by the features of the annihilation spectra noted above - the pressure dependent shoulder width, and the effect of an applied electric field. Further justification for this interpretation requires a more detailed quantitative analysis. For the present it will be assumed that the shoulder does arise from the annihilations of free positrons and an analysis made accordingly.

In considering the effect of an electric field, modifications to both the direct annihilation rate and the positronium annihilation rate need to be considered. The discussion which follows illustrates
these points in detail. Fig.27 illustrates the separate components assumed to comprise a typical annihilation spectrum in argon. The discussion in Section H of Chapter II revealed that an increase in positronium formation resulted from the application of an electric field. Even a preliminary analysis of the argon spectra requires a knowledge of the time distribution of the annihilation events arising from these 'electric field produced' positronium atoms. To facilitate the discussion, the positronium formed due to the action of the electric field will be referred to as 'delayed positronium'. Correspondingly, the positronium normally formed in the gas at zero electric field, will be called 'prompt positronium'. In the zero field case, the orthopositronium decay events appear as a single exponential on the spectrum as indicated by the 115 nsec lifetime component in Fig.27. Also, the parapositronium component, occurring within the prompt peak, verifies that positronium is formed in a time short compared to the instrumental time resolution. If an electric field is applied, the additional positronium so formed may be produced over a period of time much greater than this. While the detailed dependence of the delayed positronium formation rate as a function of time can, in principle, be obtained from a detailed numerical analysis of the diffusion equation (Section D.4 of this chapter), two limiting cases can be easily investigated. These are as follows:

(i) If the assumption is made that all the delayed positronium formation occurs at time, t = 0, then the resulting annihilation events will have the same time dependence as those resulting from the prompt positronium. For purposes of illustration, it has been assumed that a 25% increase in positronium resulted
from the electric field. The curves labelled $A_1$ and $B_1$ in Fig. 27 represent the delayed para and orthopositronium annihilation events respectively resulting from the increased positronium formation for this case.

(ii) If, on the other hand, the assumption is made that the relative probability of positronium formation to that of direct annihilation remains constant in time for a free positron, then the subsequent annihilation events for the para and orthopositronium components are represented by the curves $A_2$ and $B_2$ respectively.

For electric field strengths sufficiently low such that the mean energy of the positron velocity distribution as it tends toward equilibrium is below the positronium formation threshold, one would expect a physical situation intermediate between the limiting cases considered. The composite spectrum shown in Fig. 27 represents the sum of all the various components, assuming for the contributions from the delayed positronium, results intermediate between the limiting cases shown.

The effect of the instrumental time resolution on the shape of the annihilation spectrum is discussed in Appendix F. These investigations indicate that the distortion in the region of the shoulder is negligible, and hence that the observed spectra, for times greater than about 7 nsec beyond the prompt peak, can be considered as faithful reproductions of the true time distributions.

2. **Deduction of Direct Annihilation Mean Lifetimes**

To begin with, the detailed shape of the argon annihilation spectra in the region of the shoulder will be neglected, and only that
region beyond the termination of the shoulder considered. Assuming that in this region the direct annihilation component is exponential in character, the determination of the direct lifetime is obtained following a similar procedure to that previously outlined for the polyatomic gases. Now however, consideration must also be given to the orthopositronium events resulting from the applied electric field. Referring again to Fig. 27, it is observed that for times greater than 40 nsec, the contribution from the delayed orthopositronium annihilations, represented by either of the curves B₁ or B₂, is very similar. Thus in this region of the spectrum, the delayed orthopositronium component corresponds closely to an exponential decay having the same lifetime as the prompt orthopositronium component. The effect of the delayed orthopositronium events is thus simply to increase the intensity of the overall orthopositronium component for times greater than 40 nsec. For times shorter than this, the delayed orthopositronium events have an intensity (counts per channel) of only about 1% of the total spectrum. Hence very little error is incurred if the curve B₁ is assumed for all the delayed events. Consequently, in analyzing the spectra, a single exponential was assumed for all the orthopositronium events.

A unique determination of the orthopositronium lifetime proved to be very difficult for the spectra illustrated in Figures 28-32. For this reason, the orthopositronium mean lifetime was calculated from the quenching data measured by Celitans and Green (1964) and Celitans et al (1964). These authors report the following quenching rates for very pure argon (≤ 2 parts per 10^6 of impurities)
\[ \lambda_q = (2.1 \pm 0.3) \times 10^{-3} \lambda_0 / \text{atm}^2 \]

(from lifetime measurements)

\[ \lambda_q = (1.8 \pm 0.2) \times 10^{-3} \lambda_0 / \text{atm}^2 \]

(3 - \gamma coincidence measurements)

At greater impurity concentrations, Celitans et al. (1964) found a quenching rate proportional to the pressure:

\[ \lambda_q = 0.035 \lambda_0 / \text{atm} \]

This result is in good agreement with the value of 0.035 \lambda_0 / \text{atm} reported by Duff and Heymann (1962).

In the present experiments, it was considered that the argon was 'very pure' and thus a value for the quenching rate of

\[ \lambda_q = 1.9 \times 10^{-3} \lambda_0 / \text{atm}^2 \]

was adopted. In fact, numerous calculations for the spectra of Fig.28 using a range of values for the orthopositronium lifetime revealed that this latter quenching rate yielded the highest values for the Likelihood Function (Appendix F).

Initially, an analysis of the argon data was attempted by normalizing the intensity of the orthopositronium component for the annihilation spectra with electric field to those spectra shown in Fig.28 without electric field.

It was assumed that the effect of the increased positronium production could be taken into account using the experimental results of Marder et al. (1956), who measured the ratio of the fraction of positrons forming positronium in the presence of an electric field to the fraction forming positronium at zero field. This ratio, \( \bar{\varnothing} \), is shown in Fig.43 and in Table VIII for the electric field strengths used in the present
experiments. Unfortunately, however, this normalization procedure was found to be unworkable because of small instabilities in the single channel analyzer in the 500 keV channel. The relative detection efficiency of a 3-quantum to 2-quantum annihilation event is a very sensitive function of the energy 'window' defined by the single channel analyzer, and hence is difficult to maintain at a fixed value. Normally, this relative detection efficiency had a value of about 1/2. Consequently, any attempt at normalizing the intensity of the orthopositronium component between the various spectra was abandoned.

The following alternative procedure was thus adopted in the determination of the direct annihilation mean lifetimes.

(i) The data were linearized and the background subtracted, as previously described.

(ii) The orthopositronium lifetime was calculated using the quenching rate given by (4). The values of this lifetime, denoted by $\tau$, are shown in Table VII.

(iii) A three parameter fit (using the Maximum Likelihood program, Appendix F) was then made to the data to determine the direct annihilation mean lifetime and its corresponding intensity, and the intensity of the orthopositronium component. For this calculation the points beyond the termination of the shoulder region (denoted by the arrow in Figures 27-32) were used. Examples of these three parameter fits are shown in Figures 28-32. The line drawn through the points represents the sum of the two exponential components. Table VII summarizes the results for the direct annihilation mean lifetimes and also gives the intensity (counts per channel) of the
TABLE VII

Summary of the Direct Annihilation Mean Lifetime in Argon for Different Values of the Applied Electric Field. The Intensity of the Total Orthopositronium Component is Given in Column 5. The Errors indicated Represent the Statistical Errors Only.

<table>
<thead>
<tr>
<th>Spectrum Number</th>
<th>S/P (V cm⁻¹ atm⁻¹)</th>
<th>P (atm @ 25°C)</th>
<th>Orthopositronium Component Lifetime T(nsec)</th>
<th>Intensity I (10⁷/sec)</th>
<th>Direct Annihilation Component Tₐ(nsec)</th>
<th>λₐ(10⁷/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>29</td>
<td>0</td>
<td>10.5</td>
<td>115</td>
<td>110±5</td>
<td>17.7±0.27</td>
<td></td>
</tr>
<tr>
<td>32</td>
<td>0</td>
<td>10.5</td>
<td>115</td>
<td>108±5</td>
<td>18.1±0.41</td>
<td>5.57±0.062c</td>
</tr>
<tr>
<td>33</td>
<td>0</td>
<td>10.5</td>
<td>115</td>
<td>112±4</td>
<td>18.4±0.27</td>
<td></td>
</tr>
<tr>
<td>38</td>
<td>0</td>
<td>10.5</td>
<td>115</td>
<td>110±4</td>
<td>17.6±0.29</td>
<td></td>
</tr>
<tr>
<td>35</td>
<td>15.0</td>
<td>10.5</td>
<td>115</td>
<td>120±6</td>
<td>21.3±0.24</td>
<td></td>
</tr>
<tr>
<td>31</td>
<td>31.3</td>
<td>10.5</td>
<td>115</td>
<td>70±5</td>
<td>26.9±0.49</td>
<td></td>
</tr>
<tr>
<td>37</td>
<td>46.9</td>
<td>10.5</td>
<td>115</td>
<td>94±5</td>
<td>32.8±0.61</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>65.0</td>
<td>10.5</td>
<td>115</td>
<td>82±5</td>
<td>34.8±0.61</td>
<td></td>
</tr>
<tr>
<td>39</td>
<td>93.2</td>
<td>10.5</td>
<td>115</td>
<td>65±4</td>
<td>36.2±0.61</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>122.0</td>
<td>10.5</td>
<td>115</td>
<td>61±4</td>
<td>34.0±1.2</td>
<td></td>
</tr>
<tr>
<td>42</td>
<td>0</td>
<td>7.65</td>
<td>125</td>
<td>144±6</td>
<td>23.5±0.44</td>
<td>4.25±0.080</td>
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<tr>
<td>44</td>
<td>0</td>
<td>5.22</td>
<td>132</td>
<td>98±7</td>
<td>35.3±0.61</td>
<td>2.84±0.049</td>
</tr>
</tbody>
</table>

The orthopositronium lifetime was calculated using the quenching rate given by (4).

The intensity, I, is defined as 'counts per channel' at the position of the prompt peak (channel 88 in this case).

The mean value of Tₐ from spectra #29, #32, #33, and #38 is 17.9±0.20 nsec yielding the averaged annihilation rate of 5.57 x 10⁷/sec.
orthopositronium component extrapolated to the prompt peak position (channel 88).

The direct annihilation rates for the zero field spectra are plotted in Fig. 33 as a function of the pressure. The slope of the line obtained from these points yields the value

$$\lambda_a = (5.40\pm0.06) \times 10^6 \text{ sec}^{-1}\text{atm}^{-1}.$$  

3. Calculation of the Scattering and Direct Annihilation Cross Sections from Elementary Considerations

The observation of an exponential decay for the direct annihilation component (beyond the shoulder), even for the case of an applied electric field (Figures 30-32), suggests that the positron velocity distribution is in equilibrium (see Section H. 2 of Chapter II). In Chapter II, the direct annihilation mean lifetime resulting from such a stationary distribution was calculated for the Druyvesteyn and Maxwellian distributions assuming that these equilibrium distributions would not be seriously altered by the presence of a non-zero velocity-dependent annihilation cross section. With this proviso, and assuming that the direct annihilation cross section was of the form

$$\sigma(v) = \sigma_0(v_0/v)^n,$$

the equations (33) and (34) were obtained. Inspection of these equations reveals that a plot of $\ln (\tau_a(u)/\tau_a(v_0))$ vs $\ln (E/P)$ should yield a straight line. In the calculation of the ratio, $\tau_a(u)/\tau_a(v_0)$, two cases have to be distinguished.

(i) Case A - Delayed positronium formed at $t = 0$.

If all the delayed positronium is formed at time $t = 0$, then the lifetime of the observed direct annihilation component is
determined by the direct annihilation rate alone. Hence the appropriate ratio is that given in column 5 of Table VIII. These results, plotted in Fig. 34, clearly do not fall on a straight line, and hence, either the assumptions of the theory are poor, or, the assumption of the formation of the delayed positronium at \( t = 0 \) is wrong.

(ii) Case B - Delayed positronium formed over extended period of time. If, on the other hand, the relative probability of delayed positronium formation to that of direct annihilation remains constant in time for a free positron, then the observed mean lifetime for the direct component is given by

\[ \tau_{a} = \frac{1}{\lambda_{a}} = \frac{1}{(\lambda_{a}^{*} - \lambda_{f})} \]  

(5)

where \( \lambda_{a}^{*} \) and \( \lambda_{f} \) are the velocity-averaged direct annihilation rate and the positronium formation rate respectively. Also, denoting the fraction of positrons which form positronium in the presence of an electric field and in zero electric field by \( g^{*} \) and \( g \) respectively, it can readily be shown that

\[ g^{*} = g + (1-g) \frac{\lambda_{f}}{\lambda_{a}} \]  

(6)

Or, defining the ratio, \( \phi = g^{*} / g \), one obtains

\[ \frac{\lambda_{a}}{\lambda_{a}^{*}} = \frac{\tau^{*}_{a}}{\tau_{a}} = \frac{1}{(1-g) / (1-g\phi)} \]  

(7)

where \( \tau^{*}_{a} = 1/\lambda_{a}^{*} \). Equation (7) yields the direct annihilation mean lifetime, \( \tau_{a}^{*} \), that would be observed in the absence of the delayed positronium formation, in terms of the observed lifetime \( \tau_{a} \), the ratio \( \phi \), and the positronium formation fraction \( g \). The quantity \( \tau_{a}^{*} \) was calculated using values of \( \phi \) obtained from the work of Marder et al (1956), and the value \( g = 0.37 \),
<table>
<thead>
<tr>
<th>Spectrum Number</th>
<th>$\varepsilon/P$ (V cm$^{-1}$atm$^{-1}$)</th>
<th>$\phi^b$ (nsec)</th>
<th>$\tau_a^c$ (nsec)</th>
<th>$\tau_a (\varepsilon/P)$</th>
<th>$1-g^d$ (nsec)</th>
<th>$\tau_a^*(\varepsilon/P=0)$</th>
<th>$\tau_a^*(\varepsilon/P)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1.00</td>
<td>17.9</td>
<td>1.00</td>
<td>1.000</td>
<td>17.9</td>
<td>1.00</td>
<td></td>
</tr>
<tr>
<td>35</td>
<td>15.0</td>
<td>1.00</td>
<td>21.3</td>
<td>1.19</td>
<td>1.000</td>
<td>21.3</td>
<td>1.19</td>
</tr>
<tr>
<td>31</td>
<td>31.3</td>
<td>1.03</td>
<td>26.9</td>
<td>1.50</td>
<td>1.017</td>
<td>27.3</td>
<td>1.52</td>
</tr>
<tr>
<td>37</td>
<td>46.9</td>
<td>1.09</td>
<td>32.8</td>
<td>1.83</td>
<td>1.055</td>
<td>34.6</td>
<td>1.93</td>
</tr>
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<td>65.0</td>
<td>1.17</td>
<td>34.8</td>
<td>1.94</td>
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<td>38.6</td>
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</tr>
<tr>
<td>39</td>
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<td>1.36</td>
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<td>1.268</td>
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<td>40</td>
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<td>1.90</td>
<td>1.480</td>
<td>50.3</td>
<td>2.81</td>
</tr>
</tbody>
</table>

$^a$Average mean lifetime from spectra #29, #32, #33, and #38.

$^b$Results from Marder et al (1956).

$^c$Experimentally observed direct annihilation lifetime as given in Table VII.

$^d$Calculated from equation (7).

given by Falk and Jones (1964). The ratio $\tau_a^*/\tau_a (v_0)$ is given in the last column of Table VIII and plotted against $\varepsilon/P$ in Fig. 34. In this case the points lie very close to a straight line and thus permit a calculation of the quantity $n$ and the elastic scattering cross section, $Q_0$, by solving equations (33) and (34). In this way, one obtains

$$Q_0 = 16.9, \quad n = 1.83, \text{ for } q(v) = q_0 \quad (8)$$

and

$$Q_0 = 16.2, \quad n = 1.42, \text{ for } q(v) = q_0 \frac{v}{v} \quad (9)$$
These results lead to the conclusion that the assumption of a constant scattering cross section requires a direct annihilation cross section which varies more rapidly as a function of the positron velocity than does that of an inverse velocity-dependent scattering cross section. Also, the thermalization time constant calculated from equation (13) of Chapter II has values of 8.12 and 8.47 nsec respectively for a pressure of 10.5 atm. This yields slowing down times from an initial energy of 9 eV to a final energy of 0.1 eV (calculated from the analysis of Tao et al (1963) for a constant scattering cross section, and from equations (15) and (16) of Chapter II (with $a = 0$) for the inverse velocity dependent cross section) of 9.3 and 24.2 nsec respectively. Comparison of these slowing down times to the experimentally observed shoulder width of about 33 nsec (Fig.28) would suggest that the constant scattering cross section of 16.9 results in too rapid thermalization.

4. Numerical Solutions of the Diffusion Equation

(a) Choice of the Form of the Cross Sections and the Initial Distribution

The detailed shape of the annihilation spectra is determined by the velocity dependence of the following physical quantities: The cross section for elastic scattering, the cross section for direct annihilation, the cross section for positronium formation, and the initial velocity distribution of the positrons, $f(v,0)$. Unfortunately, it would appear that the set of experimental data presented in this thesis is not sufficient to determine all these functions uniquely. Furthermore, the nature of the numerical solution of the diffusion equation (Appendix G) employed specific forms for these functions and calculated the resulting
annihilation spectra, rather than performing the inverse operation of determining these functions from the observed annihilation spectra. For this reason, and because of the lack of any knowledge to the contrary, the velocity dependence of the cross sections was restricted to relatively simple forms and only elementary considerations given to the choice of \( f(v, o) \).

A description of the numerical solution of the diffusion equation is presented in Appendix G. In the following discussion, the positron velocity will always be specified in terms of the dimensionless quantity, \( x = v/v_o \), where \( v_o \) is the thermal velocity of the positron at 25°C, obtained from the relation, \( \frac{1}{2}mv_o^2 = (3/2) kT \). All the cross sections are given in units of \( \pi a_o^2 \) per atom.

(i) Initial Velocity Distribution, \( f(x, o) \)

The initial positron velocity distribution consists of those positrons which have been slowed down to an energy below the threshold for positronium formation, namely 8.9 eV in argon. The subsequent annihilation spectrum is determined by the time dependence of this initial distribution.

A calculation of this initial distribution requires a knowledge of all the ionization and excitation cross sections of the atom and also the positronium formation cross section. Since no such information whatever is presently available, this calculation cannot be performed. Consequently, for most of the computer solutions of the diffusion equation the simplest assumption was adopted, namely that the initial distribution had a constant density in phase space. Hence, \( f(x, o) = 1 \), and the number of positrons in the velocity interval \( dx \), about \( x \), is proportional
A modified form of this initial distribution function was also investigated. In this case it was assumed that the positrons initially in the energy interval between 11.6 and 15.7 eV (the excitation and ionization energy respectively, Fig. 2) have a high probability of being scattered inelastically into the energy interval from 0 to 4.1 eV rather than forming positronium. If the assumption is made that **all** the positrons in the energy interval between 11.6 and 15.7 eV are so scattered it can be shown that the modified distribution, $f^*$, has the form

$$f^*(x, o) = 1 + \frac{1}{(x_2/x)^2 + 1}, \quad 0 \leq x \leq 10.3$$  

$$f^*(x, o) = 1, \quad 10.3 < x \leq 15.2$$  

where $x_2 (= 17.36)$ is the positron velocity corresponding to the energy of the excitation level, namely 11.6 eV. Here it has been assumed that prior to this last inelastic scattering (from the region between 11.6 and 15.7 eV), the interval between 0 and 15.7 eV was uniformly populated in phase space.

(ii) **Elastic Scattering Cross Section**

Only two functional forms for the elastic scattering cross section were assumed in the various calculations. These were

$$Q(x) = a/x$$  

and

$$Q(x) = b$$

involving one adjustable parameter for each case. For the latter case of a constant scattering cross section, furthermore, it was found that only cases of zero electric field could be treated with the program available
because of divergence of the numerical solution of the diffusion equation for those cases where an electric field was applied.

The theoretically predicted shapes of the scattering cross sections obtained by Massay and Moussa (1958) and Malik (1961) are shown in Fig.4. These cross sections decrease approximately linearly with the positron velocity. However, since these calculations ignored polarization effects, their validity is questionable. Hence these results were not considered further.

(iii) **Direct Annihilation Cross Section**

The form for this function used in all the calculation was

\[ Z(x) = \alpha/x^n \]  

(13)

where \( \alpha \) and \( n \) were adjustable parameters.

(iv) **Positronium Formation Cross Section**

Obviously, this cross section is zero below the threshold energy for positronium formation. Above the threshold, for simplicity, a linearly rising ramp was assumed, terminating in a constant amplitude at an energy of a few electron volts above the threshold. Thus

\[ S(x) = \begin{cases} 
0 & , \quad x < 15.2 \\
= k(x-15.2), & 15.2 \leq x \leq x^* \\
= k(x^*-15.2), & x > x^* 
\end{cases} \]  

(14)

Again, two parameters, \( k \) and \( x^* \), were adjustable.

(b) **Characteristic Behavior of the Diffusion Equation Solutions**

It was noted in (a) that the four functions \( Q(x) \), \( Z(x) \), \( S(x) \), and \( f(x,\gamma) \) determine the shape of the annihilation spectrum. If the approximate representations of these functions discussed in the preceding
sections bear any resemblance to reality, the problem is now to determine if a particular set of the 5 parameters will yield a good fit to all the experimental data available. Fortunately, some aspects of the experimental curves (such as the slope of the exponential portion) are much more sensitive to the detailed velocity dependence of certain of these functions than to the others. Thus hopefully, some of these cross sections can be determined in an approximately unique way.

A series of computer calculations were performed using as a guide the results of equations (8) and (9) of Section D.3 for the scattering and annihilation cross sections. Also, trials were made using different initial distribution functions and different cross sections for positronium formation. From these calculations the following qualitative characteristics and features of the annihilation spectra were established.

(1) Shape and Width of the Shoulder

Considering for the present only those situations for which the electric field is zero, it is observed that the width of the shoulder is determined primarily by the 'average' magnitude of the cross section, \( Q(x) \). Thus the constant in either of the equations (11) or (12) is determined.

The shape of the shoulder, on the other hand depends not only on the scattering cross section but also on the functions \( f(x, \sigma) \) and \( Z(x) \). The dependence of the shoulder shape on \( f(x, \sigma) \) is not as marked as one might first expect. The difference between the spectra obtained using the initial distribution of equation (10) and the distribution \( f(x, \sigma) = 1 \) is, for example, quite small and is shown in Fig.35 by the curves labelled 'a' and 'b'. The values of the other functions used in calculating these curves are shown in Table IX.
The dependence of the shape of the shoulder on the annihilation cross section is observed to be much stronger, however. Curves 'c', 'd', and 'e' demonstrate the effect of changing the exponent, n, in the cross section (13).

Similarly, the effect of changing the form of the elastic scattering cross section is indicated by curves 'f' and 'g' of Fig.36. Another pair of curves in which both the scattering cross section and the annihilation cross section are different, are those designated 'b' and 'h' of this figure.

A qualitative understanding of the effect of the scattering cross section is obtained from an examination of Fig.37(b). Here the distributions, \( x^2 f \), are shown at different stages of thermalization for a constant scattering cross section, and a cross section proportional to \( 1/x \). At times of 5 and 20 nsec respectively, these distributions both have a maximum at \( x \approx 4 \), but the former distribution has a much narrower width than the latter. The velocity-averaged annihilation rate thus represents an effective average over a smaller interval of \( x \) for the constant scattering cross section than for the inverse velocity-dependent cross section.

(ii) Mean Lifetime of the Exponential Portion of the Curves

Again considering the zero field case, the value of the direct annihilation mean lifetime in the exponential portion of the spectrum is determined primarily by the annihilation cross section and to a lesser degree by the scattering cross section (cases f and g of Fig.36.). Further, once the exponent 'n' in equation (13) is selected, the constant \( \alpha \) is determined by the observed mean lifetime.
Values of the Cross Sections $Q(x)$, $Z(x)$, and $S(x)$, and the Initial Distribution, $f(x,0)$ for Numerical Solutions of the Diffusion Equation. These Cross Sections Represent the Elastic Scattering, the Direct Annihilation, and the Positronium Formation Cross Sections Respectively.

<table>
<thead>
<tr>
<th>Case</th>
<th>$P$ (atm)</th>
<th>$E/P$ (V cm$^{-1}$atm$^{-1}$)</th>
<th>$Q(x)$</th>
<th>$Z(x)$</th>
<th>$S(x)^a$</th>
<th>$S_m$</th>
<th>$x^*$</th>
<th>$f(x,0)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>10.5</td>
<td>0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.000050</td>
<td>15.25</td>
<td></td>
<td></td>
</tr>
<tr>
<td>b</td>
<td>10.5</td>
<td>0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.010</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>c</td>
<td>10.5</td>
<td>0</td>
<td>15/x</td>
<td>0.000209/x</td>
<td>0</td>
<td></td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>d</td>
<td>10.5</td>
<td>0</td>
<td>15/x</td>
<td>0.000209/x$^{1.4}$</td>
<td>0</td>
<td></td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>e</td>
<td>10.5</td>
<td>0</td>
<td>15/x</td>
<td>0.000209/x$^{2}$</td>
<td>0</td>
<td></td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>f</td>
<td>10.5</td>
<td>0</td>
<td>10</td>
<td>0.000225/x$^{1.83}$</td>
<td>0</td>
<td></td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>g</td>
<td>10.5</td>
<td>0</td>
<td>17/x</td>
<td>0.000225/x$^{1.83}$</td>
<td>0</td>
<td></td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>h</td>
<td>10.5</td>
<td>0</td>
<td>8.0</td>
<td>0.000174/x$^{1.83}$</td>
<td>0</td>
<td></td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>i</td>
<td>5.22</td>
<td>0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.0025</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>j</td>
<td>7.65</td>
<td>0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.0025</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>k</td>
<td>10.5</td>
<td>15.0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.0025</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>l</td>
<td>10.5</td>
<td>31.3</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.0025</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>m</td>
<td>10.5</td>
<td>46.9</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.0025</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>n</td>
<td>10.5</td>
<td>65.0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.0025</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>p</td>
<td>10.5</td>
<td>93.2</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.0025</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>q</td>
<td>10.5</td>
<td>122.0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.0025</td>
<td>20.0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>r</td>
<td>10.5</td>
<td>65.0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.000050</td>
<td>15.25</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>s</td>
<td>10.5</td>
<td>93.2</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.000050</td>
<td>15.25</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>t</td>
<td>10.5</td>
<td>122.0</td>
<td>20/x</td>
<td>0.000225/x$^{1.5}$</td>
<td>0.000050</td>
<td>15.25</td>
<td>1</td>
<td></td>
</tr>
</tbody>
</table>

$^a$The form of the cross section, $S(x)$ was taken as a ramp function, as given by equation (14). $S_m$ denotes the maximum value of the cross section, equal to $k(x^* - 15.2)$

$^b$This initial distribution function given by equation (10).
At low fields, say for $\varepsilon/P \leq 40 \text{ V cm}^{-1}\text{atm}^{-1}$, delayed positronium formation is negligible and can be ignored. Also, since the exponential portion of the curve corresponds to an equilibrium situation, the shape of the initial distribution is irrelevant. The scattering cross section, however, determines the equilibrium velocity of the distribution and thus, together with the annihilation cross section, determines the mean lifetime, $\tau_a$. For a given scattering cross section, the parameters $\alpha$ and $n$ can now be adjusted to yield the best fit to the zero field and low field results. As an example of this kind of adjustment and fitting, consider the case where $Q(x) = 20/x$, which requires a value for $\alpha$ in equation (13) of $2.25 \times 10^{-4}$ in order to fit the zero field results. Then for a value of $\varepsilon/P = 31.3 \text{ V cm}^{-1}\text{atm}^{-1}$, and for values of $n$ of 1.42 and 1.50, $\tau_a$ has values of 25.0 and 26.9 nsec respectively. The latter value is in agreement with the experimentally observed value. Thus for a given form of the scattering cross section $n$ can be determined quite accurately.

At high electric fields, the lifetime $\tau_a$ depends on both the direct annihilation rate and the positronium formation rate as given by (5). Again, the scattering cross section is important here, since it determines the equilibrium velocity of the distribution.

(iii) **Delayed Positronium Formation**

At low electric fields the increase in positronium formation is strongly dependent on the shape of the initial velocity distribution. This can be understood from the fact that at these fields nearly all the delayed positronium is formed in the first few nanoseconds. For example, for case m corresponding to a value of $\varepsilon/P$ of $46.9 \text{ V cm}^{-1}\text{atm}^{-1}$ an increase in positronium formation of 4.3% results, with about 95% of this increase
occurring in the first 10 nsec. Fig. 37(a) illustrates why this is so. For times greater than about 10 nsec, the extension of the positron velocity distribution above the positronium formation threshold is very small indeed, and hence little positronium formation results. For smaller times (as indicated by the curve labelled '5 nsec'), however, the density of positrons in the region of the threshold is much larger, and hence the probability of a positron crossing the threshold and forming positronium is much greater.

At higher electric fields the initial velocity distribution is of less importance since delayed positronium is formed from the equilibrium distribution as well as from the initial non-equilibrium distribution.

In addition to the initial distribution, each of the cross sections Q(x), Z(x), and S(x) is important in determining the amount of delayed positronium formation. Here again, the scattering cross section determines the shape and equilibrium velocity of the distribution, and the relative magnitude of the direct annihilation cross section and the positronium formation cross section determines the fractional increase in positronium formation.

Furthermore, the shape of the positronium formation cross section affects the low field and high field cases somewhat differently. At low fields, the magnitude of S(x) immediately above the threshold only, is important, since few positrons attain higher velocities. On the other hand, at high fields the velocity distribution extends significantly further above the threshold and thus the value of S(x) at high velocities is also important.

(c) Calculated Fits to the Experimental Annihilation Spectra
An attempt was made to fit all the argon annihilation spectra consistently, using one set of parameters. This was done for a scattering cross section of the form $Q(x) = a/x$, only, since the case of a constant cross section with an applied electric field produced the divergence difficulties previously noted. The form assumed for the initial velocity distribution was $f(x, o) = 1$, since little theoretical justification existed for selecting anything more complex.

From the zero field and low field fits to the data the following parameters were obtained

$$Q(x) = \frac{20}{x} \quad (15)$$
$$Z(x) = 2.25 \times 10^{-4}/x^{1.5} \quad (16)$$

The calculated curves for several of the spectra are shown in Figures 38, 39, and 40. The orthopositronium component has been subtracted from each of the experimental spectra using the orthopositronium lifetime and intensity given in Table VII. From these calculated fits it is immediately apparent that the main region of disagreement with the experimental results is in the initial region of the shoulder following the prompt peak. A discussion of this is given in the following chapter. The other features of the curves show good agreement with the data, although it appears that a slightly larger value of the scattering cross section would have improved the fit somewhat.

The relevant parameters that have to be fitted for the electric field cases are the direct annihilation lifetime, $\tau_a$, and the fractional Increase in positronium formation, $\phi$. Calculations for two widely differing values of the positronium formation cross section were performed.
Form A: \[ S(x) = 0 \quad , \quad x < 15.2 \]
\[ = 5.21 \times 10^{-4}(x-15.2) \quad , \quad 15.2 \leq x \leq 20 \]  \( (17) \)
\[ = 2.5 \times 10^{-3} \quad , \quad x > 20 \]

Form B: \[ S(x) = 0 \quad , \quad x < 15.25 \]
\[ = 5 \times 10^{-5} \quad , \quad x \geq 15.25 \]  \( (18) \)

The results are shown in Fig. 41 for spectra #30 and #40 whose values of \( \varepsilon/P \) are 65.0 and 122 V cm\(^{-1}\)atm\(^{-1}\) respectively. At these fields, the increase in positronium formation is sufficiently large that the contribution from the parapositronium annihilations must be considered. Consequently, for comparison to the experimental data, the parapositronium component was added to the direct component before plotting the curves shown in this figure. For spectrum #30, both cases of the positronium formation cross section yield results in good agreement with the observed spectrum. For spectrum #40, however, form A results in a value for \( \mathcal{T}_a \), much shorter than that observed.

A summary of the calculated values of \( \mathcal{T}_a \) and \( \phi \) using form A and form B for the positronium formation cross section is given in Table X. The experimental values of \( \mathcal{T}_a \) and \( \phi \) (from Marder et al. (1956)) are also shown for comparison. A graphical presentation of these results is shown in Figures 42 and 43. In Fig. 42 is revealed the excellent agreement between the experimental and calculated values of \( \mathcal{T}_a \) at low fields for form A of the positronium formation cross section. The calculated value of the lifetime at the highest field is much smaller however than the experimental value.

The calculated values of the lifetime, \( \mathcal{T}_a \), can be increased significantly at the higher fields by decreasing the positronium formation cross section, \( S(x) \), to that given by form B. For
TABLE X

Summary of the Experimental and Calculated Values of $\tau_a$ and $\phi$ for Argon. The Calculated Values are Shown for the Two Forms of the Positronium Formation Cross Section Given in Equation (17) and (18).

<table>
<thead>
<tr>
<th>Spectrum Number</th>
<th>$\varepsilon/P$ ($V \text{ cm}^{-1}\text{ atm}^{-1}$)</th>
<th>$\tau_a$ (nsec)</th>
<th>$\phi$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Experimental</td>
<td>Form A</td>
<td>Form B</td>
</tr>
<tr>
<td>0</td>
<td>17.9</td>
<td>18.0</td>
<td></td>
</tr>
<tr>
<td>35</td>
<td>21.3</td>
<td>21.5</td>
<td></td>
</tr>
<tr>
<td>31</td>
<td>26.9</td>
<td>26.9</td>
<td></td>
</tr>
<tr>
<td>37</td>
<td>32.8</td>
<td>31.4</td>
<td></td>
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<tr>
<td>30</td>
<td>34.8</td>
<td>36.0</td>
<td>36.1</td>
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<tr>
<td>39</td>
<td>36.2</td>
<td>35.9</td>
<td>39.0</td>
</tr>
<tr>
<td>40</td>
<td>34.0</td>
<td>22.9</td>
<td>30.7</td>
</tr>
</tbody>
</table>

$^a$Results from Marder et al (1956)

$\varepsilon/P = 65 \text{ V cm}^{-1}\text{ atm}^{-1}$,
the difference in $\tau_a$ for the two cases is very small. It is gratifying that the calculated value of $\tau_a$ for form B of the formation cross section which shows the large deviation from the experimental value, also corresponds to a calculated value of $\phi$ (Fig. 43) which deviates markedly from the experimental results.

A further examination of these results is presented in the discussion of the following chapter.

E. Direct Annihilation Rates in Helium and Krypton

1. Helium

Preliminary investigations of the direct annihilation rate of
positrons in helium were also undertaken. A summary of the experimental particulars for these spectra is given in Table VI.

The results observed were rather surprising, in the light of the argon results, since very little evidence of a shoulder was present. Furthermore, the effect of an electric field on the direct annihilation lifetime, as indicated by spectra #55 and #57 of Fig.44 and #59 and #61 of Fig.46, appears slight indeed. The small variation of lifetime which is observed can probably be explained in terms of delayed positronium formation since values of $E/P$ of about 20 V cm$^{-1}$ atm$^{-1}$ (Marder et al (1956)) are sufficient to produce saturation in the formation of delayed positronium. To investigate this situation further would require obtaining a series of annihilation spectra having much better statistics than the present ones. In view of these limitations, no diffusion equation calculations were attempted for helium.

The lifetime of the direct annihilation component was calculated, however, for the zero field spectra. In order to obtain a value for the orthopositronium lifetime, one spectrum was recorded on the 4.68 nsec/channel scale, with the single channel analyzer (in the 500 keV channel) window set to accept gamma ray energies between 100 to 570 keV. The result is shown in Fig.45. If a linear pressure-dependent quenching rate is assumed, the observed orthopositronium lifetime of 95.5 nsec yields a quenching rate

$$\lambda_q = (7.5 \pm 0.5) \times 10^{-3} \lambda_\text{atm} \quad (19)$$

This value is considerably smaller than the values of $(1.4 \pm 0.7) \times 10^{-2}$ $\lambda_\text{atm}$ and $(1.23 \pm 0.11) \times 10^{-2} \lambda_\text{atm}$ given by Heymann et al (1961) and Duff and Heymann (1962) respectively, and is probably attributable to the
use of a purer gas in the present experiments. Hence, the value given by (19) was adopted for the present calculations.

For spectrum #54 shown in Fig.44 and for spectrum #53, a three parameter fit to the data was made using the fixed value of the orthopositronium lifetime given in Table XI. For spectrum #60 shown in Fig.46 only a two parameter fit could be made since the two lifetimes were comparable in this case. The intensity of the orthopositronium component was estimated in this case after normalizing the intensities of the various spectra. Fortunately, since the two lifetimes differed by less than a factor of two, the calculated value for $\tau_a$ was quite insensitive to changes in the orthopositronium intensity, a 100% change in this latter quantity affecting $\tau_a$ by less than 2%.

The results for $\tau_a$ from these calculations are summarized in Table XI. Also shown in this table is the value of $\tau_a$ for spectrum #55, for which $E/P$ has the value 22.8 V cm$^{-1}$atm$^{-1}$. For this spectrum, $\tau_a$ has a value of 24.9 nsec compared to the mean value of 23.8 nsec for the zero field spectra, #54 and #56. The annihilation rates calculated from the values of $\tau_a$ are plotted as a function of the pressure in Fig.47. The slope of the straight line fitted to these points has the value $(7.00\pm0.08) \times 10^5$ sec$^{-1}$atm$^{-1}$.

2. Krypton

Preliminary investigations indicated the existence of a shoulder in the annihilation spectrum for krypton as reported by Falk and Jones (1964) (Appendix I). No further measurements on krypton have since been made. From the results given in Appendix I the shoulder width can be estimated to have a value of about 170 nsec atm, or about one half the
TABLE XI

Summary of the Direct Annihilation Mean Lifetime in Helium. The Lifetime and Intensity of the Orthopositronium Component are Shown in Columns 4 and 5 Respectively.

<table>
<thead>
<tr>
<th>Spectrum Number</th>
<th>$E/P$ (V cm$^{-1}$atm$^{-1}$)</th>
<th>$P$ (atm@25°C)</th>
<th>Orthopositronium Component</th>
<th>Direct Annihilation Component</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Lifetime</td>
<td>Intensity</td>
</tr>
<tr>
<td>52</td>
<td>26.8</td>
<td>49.6</td>
<td>101</td>
<td>66±5</td>
</tr>
<tr>
<td>53</td>
<td>0</td>
<td>49.6</td>
<td>95.5±6.6</td>
<td>91±5</td>
</tr>
<tr>
<td>54</td>
<td>0</td>
<td>59.7</td>
<td>95.5±3.9</td>
<td>14±5</td>
</tr>
<tr>
<td>55</td>
<td>22.8</td>
<td>59.7</td>
<td>95.5±1.8</td>
<td>740±14</td>
</tr>
<tr>
<td>56</td>
<td>0</td>
<td>59.7</td>
<td>95.5</td>
<td></td>
</tr>
<tr>
<td>57</td>
<td>4.96</td>
<td>59.7</td>
<td>95.5</td>
<td></td>
</tr>
<tr>
<td>58</td>
<td>13.7</td>
<td>20.6</td>
<td>120±18</td>
<td>77±6</td>
</tr>
<tr>
<td>60</td>
<td>0</td>
<td>20.6</td>
<td>120</td>
<td></td>
</tr>
<tr>
<td>61</td>
<td>64.5</td>
<td>20.6</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$The intensity, I, is defined as 'counts per channel' at the position of the prompt peak which was in channel 92$\frac{1}{2}$ for spectrum #56 and channel 89$\frac{1}{2}$ for the other spectra.

The value observed in argon. The value of the direct annihilation rate, also calculated from these results, is

$$\lambda_a = (1.11±0.10) \times 10^7 \text{ sec}^{-1}\text{atm}^{-1}$$
FIG. 18. Annihilation Spectrum of Positrons in CO₂ at 5.29 atm (25°C). The Points Shown Represent the Raw Data Obtained From the Kicksorter.
FIG. 19. Positron Annihilation Spectrum in Nitrogen at 11.2 atm (25°C). The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
FIG. 20. Positron Annihilation Spectrum in Nitrogen at 5.08 atm (25°C). The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
FIG. 21. Orthopositronium Annihilation Rates in Nitrogen and Carbon Dioxide as a Function of Pressure.
FIG. 22. Direct Annihilation Rates in Nitrogen and Oxygen as a Function of
the Pressure. The Dashed Lines Indicate the Annihilation Rates Computed From
the Dirac Cross Section Assuming all the Electrons of the Atom Participate.
FIG. 24. Positron Annihilation Spectrum in CO$_2$ at 9.20 atm (25°C). The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
FIG. 25. Positron Annihilation Spectrum in CO$_2$ at 3.04 atm (25°C). The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
FIG. 26. Direct Annihilation Rate in CO₂ as a Function of the Gas Density. The Dashed Line Indicates the Annihilation Rate Computed From the Dirac Cross Section Assuming all the Electrons of the Atom Participate.
FIG. 27. Hypothetical Annihilation Spectrum in Argon with an Applied Electric Field. Annihilation Events from Delayed Para and Orthopositronium are Indicated by Curves $A_1$ and $B_1$ Respectively for One Limiting Case of Delayed Positronium Formation and by Curves $A_2$ and $B_2$ for Another Limiting Case.
FIG. 28. Positron Annihilation Spectrum in Argon at 10.5 atm (25°C) and \(\varepsilon/P = 0\). The Three Different Spectra were Obtained Under Identical Experimental Conditions, But at Different Times. The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
ARGON, $\epsilon/P = 0$

- Spectrum #44, $P = 5.22\, \text{atm}$
- Spectrum #42, $P = 7.65\, \text{atm}$

$2.43\, \text{nsec/ch}$

FIG. 29. Positron Annihilation Spectrum in Argon at 5.22 and 7.65 atm ($25^\circ\text{C}$) and $\epsilon/P = 0$. The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
FIG. 30. Positron Annihilation Spectrum in Argon at 10.5 atm (25°C) and $\varepsilon/P = 31.3$. The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
FIG. 31. Positron Annihilation Spectrum in Argon at 10.5 atm (25°C) and ε/P = 65.0. The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
FIG. 32. Positron Annihilation Spectrum in Argon at 10.5 atm (25°C) and $\varepsilon/P = 122$. The Sum of the Two Exponential Components is Indicated by the Solid Line Drawn Through the Points.
FIG. 33. Direct Annihilation Rate in Argon at Zero Field as a Function of the Pressure. The Rates are Calculated from the Exponential Portion of the Direct Component in the Annihilation Spectra. The Dashed Line Indicates the Annihilation Rate Computed from the Dirac Cross Section Assuming all the Electrons of the Atom Participate.
FIG. 34. Relative Direct Annihilation Mean Lifetime in Argon as a Function of $\varepsilon/P$. 

- Case A: Delayed Positronium Formed at $t=0$
- Case B: Delayed Positronium Formed Over Extended Period of Time
FIG. 35. Calculated Direct Annihilation Spectra in Argon from Computer Solutions of the Diffusion Equation. The Effect of Changing the Initial Velocity Distribution is Demonstrated by Cases a and b. Cases c, d, and e Indicate the Effect of Changing the Exponent, n, in the Annihilation Cross Section. The Values of the Other Functions and Parameters Used in Calculating these Curves are Shown in Table IX.
FIG. 36. Calculated Direct Annihilation Spectra in Argon Showing the Effect of Changing the Scattering Cross Section (Cases f and g), and a Combination of the Scattering Cross Section and the Annihilation Cross Section (Cases b and h). The Values of the Other Functions and Parameters Used in Calculating these Curves are Shown in Table IX.
FIG. 37. Calculated Positron Velocity Distributions in Argon as a Function of Time. The Line Marked $\bar{v}$ Denotes the Most Probable Velocity of the Equilibrium Distribution. The Values of the Other Functions and Parameters Used in Calculating these Curves are Shown in Table IX.
FIG. 38. Diffusion Equation Fit to Direct Annihilation Component in Argon at 10.5 atm and ε/P = 0. The Orthopositronium Events Have Been Subtracted from the Experimental Data.
FIG. 39. Diffusion Equation Fits to Direct Annihilation Components in Argon at 5.22 and 7.65 atm (25°C) for $\varepsilon/P = 0$. The Orthopositronium Events Have Been Subtracted From the Experimental Data.
FIG. 40. Diffusion Equation Fit to Direct Annihilation Component in Argon at 10.5 atm (25°C) and \( \varepsilon/P = 31.3 \text{ Vcm}^{-1}\text{atm}^{-1} \). The Orthopositronium Events Have Been Subtracted From the Experimental Data.
FIG. 41. Diffusion Equation Fits to Direct Annihilation Components in Argon at 10.5 atm (25°C) for \( \epsilon/P = 65.0 \) and 122. Cases \( n \) and \( g \) were Calculated Using the Values for the Positronium Formation Cross Section Given by (17), While Cases \( r \) and \( t \) Used the Values Given by (18). The Orthopositronium Events Have Been Subtracted From the Experimental Data.
FIG. 42. Comparison of Experimental and Calculated Values of $\tau_a$ as a Function of $\epsilon/P$. The Calculated Values of $\tau_a$ are Shown for the Two Forms of the Positronium Formation Cross Section Given by Equations (17) and (18).
FIG. 43. Comparison of Experimental and Calculated Values of $\phi$ as a Function of $\epsilon/P$. The Calculated Values of $\phi$ are Shown for the Two Forms of the Positronium Formation Cross Section Given by Equations (17) and (18).
FIG. 44. Positron Annihilation Spectrum in Helium at 59.7 atm (25°C) for ε/P = 0, 4.96, and 22.8.
FIG. 45. Positron Annihilation Spectrum in Helium at 59.7 atm (25°C) and ε/P = 0. For this Spectrum the Single Channel Analyzer (in the 500 keV Channel) was Set to Accept Gamma Ray Energies Between 100 to 570 keV. The Solid Line Drawn Through the Points Represents the Sum of the Two Exponential Components.
FIG. 46. Positron Annihilation Spectrum in Helium at 20.6 atm (25°C) for $\varepsilon/P = 0$, 13.7, and 64.5.
FIG. 47. Direct Annihilation Rate in Helium as a Function of the Pressure. The Dashed Line Indicates the Annihilation Rate Computed From the Dirac Cross Section Assuming all the Electrons of the Atom Participate.
CHAPTER VI

DISCUSSION OF RESULTS AND CONCLUSIONS

A. Discussion of Errors

1. Effect of Gas Impurities

(a) Direct Lifetime

A crucial factor in many of the experiments described was that of gas purity. As noted in Chapters II and IV, the sensitivity of an experiment to a particular gas impurity depended on the experimental quantity being investigated. The experimental observation, of comparable direct annihilation rates in $N_2$, $O_2$, and $CO_2$ leads to the conclusion that an impurity of one of these gases in any of the other gases of this group would affect the measured annihilation rate to a negligible degree. The precautions described in Chapter IV which were followed while filling of the pressure chambers should have reduced further contamination of the gas to a minimum. Thus it is felt that these results should be free from any gross errors attributable to gas impurities. In the case of argon, it was observed that the lifetime of the exponential portion of the direct component was quite insensitive to polyatomic impurities such as nitrogen, as illustrated in Fig. 48.

(b) Orthopositronium Lifetime

The effects of impurities, however, may have resulted in relatively large errors in the determination of the orthopositronium lifetime. This lifetime is strongly affected by such quenching gases as $O_2$ and $NO$, the former of which is a common impurity in most gases (see Table III). Where reliable comparisons of the orthopositronium lifetime could be made with
FIG. 48. Effect of Nitrogen Impurities on the Annihilation Spectrum in Argon at 5.22 atm (25°C).
those of other workers, the agreement between the results indicated that these errors were not large (see Chapter V). Since an accurate determination of the orthopositronium lifetime itself was not the object of the present experiments, no special efforts beyond those described in Chapter IV were made in purifying the polyatomic gases $N_2$, $O_2$, and $CO_2$.

(c) Shoulder Width in the Noble Gases

Of much greater significance from the point of view of the present experiments were the effects of impurities on the shape of the shoulder in the annihilation spectra of $He$, $A$, and $Kr$. While a mass spectrometer analysis of a sample of argon (taken from the pressure chamber) revealed no detectable impurities (Chapter IV), it was felt that a further investigation should be made. For this purpose, nitrogen was added to the argon in amounts of 0.1%, 0.8%, and 2.7% (by volume), and annihilation spectra recorded at each of these nitrogen concentrations. The results are shown in Fig. 48. For a nitrogen concentration of 0.1% the spectrum is identical to spectrum #44 (Fig. 29) recorded at zero nitrogen concentration. For a concentration of 0.8% nitrogen, the width of the shoulder has decreased noticeably, and at 2.7%, both the width and the shape of the shoulder have been altered drastically. From these investigations it appears that nitrogen impurities at concentrations of about 0.1% or less produce a negligible effect on the observed annihilation spectrum. The observation of a wider 'shoulder' width (see Section C1(c) of this chapter) in these experiments than that found by Paul (1954) and Tao et al. (1964) would suggest that some polyatomic impurities were present in their gases.

Although similar investigations were not made in helium, it is felt that these results should also be free from any gross errors, since
the gas purifier was also operated while these spectra were recorded.

2. Statistical Errors and Errors Due to Uncertainty in Orthopositronium Lifetime

The errors quoted in Chapter V for the direct annihilation rates represented the statistical counting errors only. In addition to these errors, the uncertainty in the orthopositronium lifetime contributes a further error to the calculated direct lifetime. This error was estimated by examining the results of the Maximum Likelihood calculations (Appendix F), and is given in Table XII.

3. Uncertainty of Time Calibrations

Over the period of time during which all the annihilation spectra reported in this thesis were recorded, the total shift in the position of the prompt peak was less than 5 nsec, or approximately 0.7% of the total length of the time scale - 700 nsec. In order to include the uncertainty in the knowledge of the relative channel widths (see Appendix F), an overall probable error of ±1% was allowed for the time sorter calibration.

4. Errors in Measuring $E$ and $P$

The error in determining the electric field strength was about ±4%, consisting entirely of the error in measuring the high voltage applied to the chamber.

The probable error in determining the gas pressure in the various experiments is shown in Table XII.

5. Error in the Calculated Values of $\phi$

Calculation of the fractional increase in positronium formation, $\phi$, presented in Table X, requires a knowledge of the fraction of positrons forming positronium at zero field. Uncertainty in this fraction $g$ results
in an error in the calculated value of \( \phi \) which, using equation (31) of Appendix G, can be shown to be

\[
\frac{d\phi}{\phi} = \frac{(1 - \phi)}{(1 - g)\phi} \frac{dg}{g}
\]

Thus, for example, for \( g = 0.37 \pm 0.03 \) (Falk and Jones (1964)) and \( \phi = 1.4 \), a probable error of 3.7% occurs in \( \phi \).

6. Total Error Ascribed to Direct Annihilation Rates

In the final column of Table XII the total probable errors in the calculated values of the direct annihilation rates are shown.

B. Direct Annihilation Rates in \( N_2, O_2, CO_2, He, A, Kr \)

1. General Discussion

For all the gases investigated, the direct annihilation component, excluding the initial portion of the spectrum ('the shoulder') in the noble gases, was characterized by an exponential decay. This observation leads to the conclusion that either

(i) The positrons are in thermal equilibrium, that is, the positron velocity distribution is stationary, or

(ii) The annihilation cross section is inversely proportional to the positron velocity over the velocity range characterizing this portion of the slowing-down process.

This latter conclusion is unsatisfactory in the light of the evidence provided by the argon results which clearly point to a velocity dependence of the annihilation cross section other than that of an inverse dependence. Furthermore, in the polyatomic gases, thermalization of the positrons is expected to occur in times comparable to the width of the prompt peak in the annihilation spectra. Thus, one is led to the conclusion that the
TABLE XII

Errors in the Determination of the Direct Annihilation Rates per Atmosphere for the Gases $N_2$, $O_2$, $CO_2$, $A$, $He$, and $Kr$.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Statistical Counting Error (%)</th>
<th>Error Due to Uncertainty in Ortho Lifetime (%)</th>
<th>Uncertainty in Time Sorter Calibration (%)</th>
<th>Error in the Pressure P (%)</th>
<th>Total Error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_2$</td>
<td>0.36</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1.8</td>
</tr>
<tr>
<td>$O_2$</td>
<td>1.3</td>
<td>-</td>
<td>1</td>
<td>1</td>
<td>1.9</td>
</tr>
<tr>
<td>$CO_2$</td>
<td>2</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>3.1</td>
</tr>
<tr>
<td>$A$</td>
<td>1.1</td>
<td>3</td>
<td>1</td>
<td>2</td>
<td>3.9</td>
</tr>
<tr>
<td>$He$</td>
<td>1.1</td>
<td>3</td>
<td>1</td>
<td>3</td>
<td>4.5</td>
</tr>
<tr>
<td>$Kr$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>10</td>
</tr>
</tbody>
</table>

It is assumed, here, that these errors are all uncorrelated and can be combined by taking the square root of the sum of the squares.
exponential decay is attributable to annihilation from a thermalized positron distribution. A further discussion of this matter for the case of argon is presented in Section C.

Secondly, with the exception of CO₂, the gases investigated revealed an annihilation rate directly proportional to the atomic (or molecular) density of the gas (Figures 22, 26, 33, and 47). Such a dependence of the annihilation rate on the gas density is expected if a positron interacts with an individual atom (or molecule), rather than with several atoms (or molecules) at the same time. At pressures much higher than those used in the present experiments it is not unlikely that a departure from this linear density dependence would be observed.

2. Dependence of the Direct Annihilation Rate on the Number of Electrons per Molecule

A summary of the direct annihilation rates in the monatomic and polyatomic gases is presented in Table XIII. Where available, the results of other workers have also been included for comparison. In Fig. 49 the direct annihilation rate per atmosphere has been plotted as a function of the number of electrons per molecule, Z. Somewhat surprisingly, it is seen that if the point for CO₂ is neglected, the remaining points (from
the present work) can be represented approximately by a straight line with a slope, $0.33 \times 10^6 \text{sec}^{-1}\text{atm}^{-1}$ per electron, which is about 1.8 times the Dirac annihilation rate. This line, however, is clearly outside the statistics on the points for $N_2$, $O_2$, and $A$. Since one would expect the annihilation rate characteristic of a particular gas to depend on the detailed electronic structure of the atom (or molecule) deviations from this line for individual cases is not unexpected.

Whereas six gases were investigated, comparisons with the results of other workers could only be made for three of these gases. The result for helium, obtained by Duff and Heymann (1962), is about 14% lower than the annihilation rate obtained in the present experiments, although this comparison is complicated by the uncertainty in the temperature appropriate to their result. For argon, values of the annihilation rate have been given by Tao et al (1964), Paul (1964), and Falk and Jones (unpublished). These values are $4.55$, $4.75$, and $5.1 \times 10^6 \text{sec}^{-1}\text{atm}^{-1}$ respectively compared to the value presented here of $(5.4 \pm 0.21) \times 10^6 \text{sec}^{-1}\text{atm}^{-1}$. It is believed that the earlier value of Falk and Jones (unpublished) is somewhat low because part of the shoulder was probably included with the exponential component when analyzing the data. Whether this explains the lower values obtained by the two other workers is not known.

3. **Comparison With Direct Annihilation Rates in Liquids and Solids**

Liu and Roberts (1963) have performed measurements of the direct annihilation rates of positrons in a number of condensed gases. These results are summarized in Table XIV. For comparison with our results, their values of the lifetime have been normalized to correspond to the density of the gas at 25°C and atmospheric pressure, assuming that the
TABLE XIII

Summary of Direct Annihilation Rates in the Monatomic and Polyatomic Gases. For the Monatomic Gases, the Results Refer to the Exponential Portion of the Direct Component, Only.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Molecular Z</th>
<th>Direct Annihilation Rate ((10^6 \text{ sec}^{-1}\text{atm}^{-1}), 25^\circ\text{C})</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>2</td>
<td>(0.70 \pm 0.032) (0.60 \pm 0.04)</td>
</tr>
<tr>
<td>(N_2)</td>
<td>14</td>
<td>(5.52 \pm 0.10) (4.8)</td>
</tr>
<tr>
<td>(O_2)</td>
<td>16</td>
<td>(4.70 \pm 0.09) (5.4 \pm 0.21)</td>
</tr>
<tr>
<td>(A)</td>
<td>18</td>
<td>(5.4 \pm 0.21) (5.1) (4.55) (4.75)</td>
</tr>
<tr>
<td>(\text{CO}_2)</td>
<td>22</td>
<td>(10.7 \pm 0.33) (11.1 \pm 1.1)</td>
</tr>
<tr>
<td>(\text{Kr})</td>
<td>36</td>
<td></td>
</tr>
</tbody>
</table>

\(^{a}\) The errors indicated are the total errors given in the last column of Table XII.

\(^{b}\) Duff and Heymann (1962). The temperature was not indicated for this result.

\(^{c}\) Calculated from results given by Deutsch (1953).

\(^{d}\) Falk and Jones, unpublished.

\(^{e}\) Tao et al (1964)

\(^{f}\) Paul (1964)

\(^{g}\) The result for \(\text{CO}_2\) was obtained from the low pressure measurements as shown in Fig. 26.
TABLE XIV

Direct Annihilation Lifetimes in Condensed Gases from the Work of Liu and Roberts (1963). The Values for the Gaseous State are those Reported in this Thesis.

<table>
<thead>
<tr>
<th>Substance</th>
<th>State</th>
<th>Temp (°K)</th>
<th>$\tau_a$ (nsec)</th>
<th>Density (g/cm$^3$)</th>
<th>$\tau_a(\rho_{gas})^c$</th>
<th>$\tau_a = 1/\tau_a(\rho_{gas})$ (10$^{-6}$ sec$^{-1}$ atm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H$_2$</td>
<td>Gas</td>
<td>298</td>
<td>8.25x10^{-5}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Liquid</td>
<td>20.4</td>
<td>0.92±0.04</td>
<td>0.07</td>
<td>780</td>
<td>1.28</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>13</td>
<td>0.80±0.03</td>
<td>0.0763</td>
<td>740</td>
<td>1.35</td>
</tr>
<tr>
<td>He</td>
<td>Gas</td>
<td>298</td>
<td>1.63x10^{-4}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Liquid$^a$</td>
<td>4.2</td>
<td>1.83±0.15</td>
<td>0.125</td>
<td>1430</td>
<td>0.70</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4.2</td>
<td>1.90±0.06</td>
<td>0.125</td>
<td>1450</td>
<td>0.69</td>
</tr>
<tr>
<td>Ne</td>
<td>Gas</td>
<td>20.4</td>
<td>8.25x10^{-4}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>20.4</td>
<td>0.43±0.03</td>
<td>1.20</td>
<td>626</td>
<td>0.60</td>
</tr>
<tr>
<td>N$_2$</td>
<td>Gas</td>
<td>298</td>
<td>1.14x10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Liquid</td>
<td>77.3</td>
<td>0.56±0.02</td>
<td>0.804</td>
<td>393</td>
<td>2.54</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>62</td>
<td>0.48±0.02</td>
<td>0.87</td>
<td>365</td>
<td>2.74</td>
</tr>
<tr>
<td>O$_2$</td>
<td>Gas</td>
<td>298</td>
<td>1.30x10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Liquid</td>
<td>90.1</td>
<td>0.45±0.02</td>
<td>1.14</td>
<td>392</td>
<td>2.55</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>4.2</td>
<td>0.38±0.02</td>
<td>1.52</td>
<td>441</td>
<td>2.27</td>
</tr>
<tr>
<td>A</td>
<td>Gas</td>
<td>298</td>
<td>1.63x10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Liquid</td>
<td>86</td>
<td>0.50±0.02</td>
<td>1.41</td>
<td>431</td>
<td>2.32</td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>77.3</td>
<td>0.43±0.03</td>
<td>1.45</td>
<td>381</td>
<td>2.62</td>
</tr>
<tr>
<td>Kr</td>
<td>Gas</td>
<td>298</td>
<td>3.43x10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Xe</td>
<td>Gas</td>
<td>298</td>
<td>5.37x10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Solid</td>
<td>77.3</td>
<td>0.40±0.03</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$Paul and Graham, Phys. Rev. 106, 16 (1957)

$^b$The densities were obtained from the "Smithsonian Physical Tables", Vol. 120, p291, and the "Handbook of Chemistry and Physics", 42 Edition, The Chemical Rubber Publishing Co.

$^c$ $\tau_a(\rho_{gas})$ is the calculated lifetime after normalizing to the corresponding gas density at 25°C and 1 atm.
lifetime is inversely proportional to the density. These results, 
illustrated in Fig. 49, reveal that in all cases where comparisons can be 
made, the annihilation rate in the liquid or solid is less than or equal 
to the annihilation rate in the gas. Furthermore, for nitrogen, oxygen, 
and argon these rates differ by a factor of approximately two. Intuitively, 
one would expect a larger annihilation rate in the denser media since the 
positron interaction with the atomic electrons should extend to several 
molecules simultaneously. The question arises whether the polarization 
effect produced by the positron on the atomic electron cloud is diminished 
when the intermolecular distances are reduced to those found in the liquid 
and solid state. An interesting case is presented by helium for which 
the annihilation rate per atmosphere is the same in the liquid and the gas. 
This suggests that the interaction of the positron with the helium atom 
remains substantially the same over a range of densities differing by a 
factor of $10^3$.

4. Nonlinear Density Dependence of the Annihilation Rate in Carbon 
Dioxide

It was noted earlier that the direct annihilation rate of positrons in carbon dioxide could be represented by the equation

$$\lambda_a = (0.852 \rho - 0.040 \rho^2) \times 10^7/sec$$

where the density, $\rho$, is expressed in terms of the density at 25°C and 
atmospheric pressure. It is tempting to interpret such a dependence of 
the annihilation rate on the gas density in terms of a positron-molecular 
interaction which extends to an increasing number of molecules as the gas 
density is increased. In the light of the evidence provided by the low 
anihilation rates in the condensed gases, however, a more detailed in-
FIG. 49. Direct Annihilation Rate Per Atmosphere for Substances in the Gaseous, Liquid, and Solid State. The Annihilation Rates in the Liquids and Solids have been Normalized to Correspond to the Density of the Gas at 25°C and 1 atm. The Results for Helium by Duff and Heymann (1962) and Paul and Graham (1957) Fall Within the Circle Shown.
interpretation must await further experimental investigation.

5. **Conclusions**

Summarizing the results on the direct annihilation rates in gases, the following conclusions can be made:

(i) The exponential portion of the direct annihilation component arises from the annihilation of free positrons in thermal equilibrium with the molecules of the gas.

(ii) At present, it is impossible to predict quantitatively the direct annihilation cross section for positrons in an arbitrary gas. The information provided by the results for carbon dioxide and the condensed gases indicates that complex mechanisms are involved.

(iii) The group of gases consisting of He, N$_2$, O$_2$, A, and Kr are characterized by an annihilation rate approximately proportional to the number of electrons per molecule (Fig. 49). It would be instructive to investigate other gases such as hydrogen and neon to determine whether they belong to this 'group'. In addition it would be valuable to have data for other gases such as CO, NO, N$_2$O, and SO$_2$.

(iv) An investigation of the annihilation rate both as a function of the temperature and over a greater range of pressures, especially in a gas such as CO$_2$, should yield significant information regarding the annihilation process.

C. **Direct Annihilation of Positrons in Argon**

1. **Qualitative Discussion**

   (a) Identification of the Shoulder as Part of the Direct Component
While the tacit assumption was made in the analysis of the data that the shoulder was attributable wholly to the direct annihilation of free positrons, the results of such an analysis itself provides strong support for the validity of this interpretation. The evidence supporting this interpretation is as follows:

(i) Firstly, as noted earlier, the width of the shoulder varies inversely as the pressure, as one would expect for free positrons.

(ii) The effect of an electric field both on the shape of the shoulder, as well as on the lifetime of the exponential component, suggests a strong correlation between these two components of the annihilation spectrum.

(iii) The effect of the addition of 2% CO₂ to the argon has also been shown (Falk and Jones (1964)) (Appendix I), to support this interpretation.

(iv) The experimental evidence (Chapter V) suggests that at zero fields, all the positronium is formed in a time of the order of several nanoseconds. Calculations from the diffusion equation confirm this observation. Thus a delay in the formation of positronium (at zero fields) cannot be postulated to explain the shoulder. Furthermore, as Paul (1964) points out, the intensity of the orthopositronium component is insufficient to produce an effect of the magnitude observed.

(b) The Exponential Decay in the Direct Component

In the early reports on the observation of a shoulder in the
direct annihilation of positrons in argon by Tao et al (1964), Falk and Jones (1964) (Appendix I), and Paul (1964), no definite conclusions were made regarding the nature of the transition from the shoulder to the exponential decay. This can be attributed to the lack of knowledge regarding the velocity dependence of both the elastic scattering cross section and the annihilation cross section, knowledge of which is required to determine the width of the shoulder.

Calculations of the positron slowing-down time based on a constant scattering cross section of $1.5\pi a_0^2$ (Marder et al (1956)) requires that the shoulder width corresponds to an energy decrease of the positron from 8.9 eV to approximately 1 eV. If the start of the exponential decay corresponds to a positron energy of 1 eV, one is forced to postulate either that the annihilation cross section is inversely proportional to the positron velocity for energies less than or equal to 1 eV, or that the positron does not slow down below the energy of 1 eV. Both of these conclusions are unsatisfactory. The evidence presented in this thesis suggest that the exponential decay corresponds to the direct annihilation of positrons which are in thermal equilibrium with the atoms of the gas. This is most readily observed by examining the effect of weak electric fields on the direct annihilation spectrum. From the diffusion equation (equation (21) of Chapter II) it can be shown that the relative magnitude of the term involving the temperature of the scatterers to that involving the electric field term, is given by

$$R(T/E) = 2.794 \times 10^4 \left( \frac{m}{M} \right) \left( \frac{P_0}{E} \right)^2$$

for $q(v) = q_0 v_0 / v$.
The lowest field strength at which measurements were made in argon corresponded to $\mathcal{E}/P = 15.0 \text{ V cm}^{-1}\text{atm}^{-1}$. Using $Q_0 = 20$ (the value used in the calculations in Chapter V for the inverse velocity dependent cross section), the above equation yields $R(T/E) = 0.68$. Thus, for this value of the field strength, the shift of the equilibrium energy of the positrons due to the electric field is only of the order of $kT$. However, the lifetime of the direct component has increased by 19% over that at zero field (Table VII). Clearly, such a large effect would not be produced if

(i) the positrons all had energies of the order of 1 eV,

or if

(ii) the positron annihilation rate was constant for positron energies below 1 eV.

(c) **Comparison With Results of Other Workers**

As noted earlier, two other groups, Tao et al (1964) and Paul (1964), have reported observations in argon at zero electric field similar to those presented here. The overall interpretation given by these authors parallels that given by Falk and Jones (1964). A useful parameter for comparing the various results is the width of the shoulder. The shoulder width is here defined as the time interval between the midpoint of the prompt peak and the 'end' of the shoulder, the 'end' being defined by the intercept of the straight line drawn tangent to the central portion of the shoulder and the straight line extension of the exponential component, as shown in Fig. 39. Measurements from the results presented by Paul (1964) yield a value of about 300 nsec atm, for this width, in approximate agreement with the mean value $340 \pm 10$ nsec atm, obtained from Figures 38 and 39. The results of Tao et al (1964), however yield a width
of only about 100 nsec atm. Furthermore, their annihilation spectra reveal a distinct minimum immediately following the prompt peak. These observations suggest that some polyatomic impurity was present in their argon gas, since similar results were apparent in the investigations reported here when a nitrogen impurity was added to the argon (Fig. 48).

2. **Diffusion Equation Fits to the Annihilation Spectra**

(a) **General Features**

Although only a few simple forms for the various cross sections were investigated in the solutions of the diffusion equation, it is possible to arrive at several general conclusions. In addition, it is possible to obtain a set of functions, namely the cross sections for elastic scattering, direct annihilation, and positronium formation which yield results consistent with all the experimental data available at the present time. It is of great interest, first, to determine the consistency of these predictions with other types of experimental results, such as the temperature dependence of the shape of the direct annihilation component and secondly, to determine the extent by which these functions may be varied and still yield results consistent with experiment.

The best fit to the experimental data was obtained using the following values for the cross sections

\[ Q(x) = \frac{20}{x} \text{ (Scattering cross section)} \]  \hspace{1cm} (1)

\[ Z(x) = 2.25 \times 10^{-4}/x^{1.5} \text{ (Direct annihilation Cross Section)} \]  \hspace{1cm} (2)

\[ S(x) = 0 \quad \text{, } x \leq 15.2 \]
\[ = 10^{-3}(x - 15.2) \quad , \quad 15.2 < x \leq 15.25 \]  \hspace{1cm} (3)
\[ = 5 \times 10^{-5} \quad \text{, } x > 15.25 \]

(Positronium Formation Cross Section)
Also, the initial distribution, \( f(x,0) \), was taken as 1 in the energy interval 0 - 8.9 eV. Although calculations were not performed with this value of \( S(x) \) for the three 'low field' cases (\( \mathcal{E}/P = 15.0, 31.3, \) and 46.9 V cm\(^{-1}\)atm\(^{-1}\)), it can be inferred from the calculation for \( \mathcal{E}/P = 65.0 \) V cm\(^{-1}\)atm\(^{-1}\) that the quantities \( \phi \) and \( \tau_a \) would not be altered significantly from the values obtained when using for \( S(x) \), the value given in equation (17) of Chapter V. Using this set of functions the following features regarding the quality of the fit to the experimental data are noted.

(i) In the initial portion of the shoulder following the prompt peak, the calculated curves exhibit the greatest deviation from the experimental data (Figures 38 - 40).

(ii) The experimental annihilation spectra (Figures 38 and 39) reveal a more sudden transition from the shoulder to the exponential component than do the calculated curves.

(iii) The direct annihilation mean lifetime, \( \tau_a \), calculated from the diffusion equation agrees with the experimental values to \( \pm 4\% \) for values of \( \mathcal{E}/P \leq 70 \) V cm\(^{-1}\)atm\(^{-1}\), and to within \( \pm 10\% \) at the higher fields.

(iv) The fractional increase in positronium formation, \( \phi \), calculated from the diffusion equation is in agreement with the experimental results of Marder et al (1956) within the experimental errors.

The following discussion examines these features in more detail.

(b) Shoulder Shape

The curves presented in Fig.35 revealed that only a small effect
on the shape of the shoulder occurred if different initial distributions, 
\( f(x, o) \), were assumed. However, these curves do illustrate that the use of the modified distribution, 
\( f(x, o) = f^* \), in which the density of positrons at small values of velocity is greater than that given by 
\( f(x, o) = 1 \), yields a slightly improved fit in the initial portion of the shoulder. Furthermore, the shoulder width is slightly reduced, improving the agreement in Figures 38 and 39. A better quantitative estimate of the initial distribution function would therefore be highly desirable.

A more marked effect on the shoulder shape is produced by changing the direct annihilation cross section as shown in Fig. 35. The initial portion of the shoulder is brought into better agreement with experiment if the exponent \( 'n' \) (in equation (13) of Chapter V) is reduced from the value given above of 1.50. Such a change, however, results in a serious disagreement of the calculated direct annihilation lifetime, \( \tau_a \), with the experimental values at the different field strengths.

The calculations performed by Paul (1964) to fit the observed annihilation spectra employed both a constant scattering cross section and a scattering cross section decreasing linearly with the positron velocity. For either case, he was able to show that the resulting annihilation cross section was approximately inversely proportional to the positron velocity in the energy range between 1 and 9 eV. At some lower energy (whose value depended on the magnitude of the scattering cross section) the annihilation cross section increased rapidly. Further calculations with annihilation cross sections having such a velocity dependence should be investigated, but it is questionable whether the experimental annihilation
spectra at intermediate field strengths could be fitted assuming such a velocity dependence.

(c) Effect of the Choice of the Scattering Cross Section

Teutsch and Hughes (1956) performed a detailed theoretical investigation of the experimental results of Marder et al. (1956) on the effect of an electric field on positronium formation. They were able to obtain a good fit to the experimental data using a constant value for the scattering cross section and assuming that the positronium formation cross section was of the same order of magnitude as the elastic scattering cross section (i.e., $\approx \pi a_o^2$). This value is greater by a factor of about $10^5$ than the value of $5 \times 10^{-5} \pi a_o^2$ obtained from the calculations in this thesis. Such widely differing values for the positronium formation cross section are explicable in terms of the drastically different shapes of the positron velocity distributions that are obtained using a constant scattering cross section and a cross section inversely proportional to the positron velocity (Fig. 37(b)). In addition, since the only data they had available was the fractional increase in positronium formation, $\phi$, the magnitude of the constant scattering cross section was adjusted from this information alone.

Also, their analysis rejects the possibility of a 'small' positronium formation cross section after assuming that the formation of the delayed positronium occurs only from an equilibrium positron distribution. It was noted in Chapter V that at low fields essentially all the delayed positronium is formed in times of the order of 5 nsec - much shorter than the times required to reach equilibrium. Thus their rejection of a small positronium formation cross section is not justified.
The lack of knowledge concerning the positronium formation cross section limits the conclusions that can be made regarding the 'high field' results. Indeed, if the positronium formation cross section is as small as that found from these calculations, that is, of the same order of magnitude as the direct annihilation cross section, then the assumption that the initial distribution, \( f(x,0) \), is zero above the positronium formation threshold, needs to be reexamined.

Theoretical calculations on positronium formation have been carried out only for helium (Massey and Moussa (1961)) and atomic hydrogen (Cheshire (1964)). These theoretical cross sections are characterized by a relatively steep front edge reaching a maximum value at about 7 eV above the threshold. The maximum values in helium and atomic hydrogen were about \( 0.4\pi a_o^2 \) and \( 4\pi a_o^2 \) respectively.

3. **Conclusions**

The observed annihilation spectrum of 'free' positrons in argon can be described in terms of the time-dependent positron velocity distribution and a velocity dependent annihilation rate. It is evident that more solutions of the diffusion equation will have to be examined in order to obtain a more comprehensive picture of the dependence of the annihilation spectrum on the scattering cross section, the direct annihilation cross section and the positronium formation cross section. The results of the calculations described in this thesis show that a consistent description of the experimental results is obtained using the values for the cross sections given by equations (1) - (3), but it cannot be concluded that this set of functions is unique.

In order to make further progress it would be of great benefit if
the dependence of $\phi$ on $E/P$ were known to a higher degree of accuracy. Also a more detailed knowledge of the shape of the annihilation spectra, using higher counting rates, and lower random coincidence backgrounds should be obtained.

Another line of investigation which would shed light on the mechanisms involved would be an examination of the effect of changing the temperature of the gas. This should produce an effect similar to that produced by an electric field, and would serve to check the general interpretation of the results. Further investigations using applied electric fields in gases such as nitrogen, hydrogen, oxygen, and neon would be profitable. Although the polyatomic gases investigated revealed no evidence of a 'shoulder', sufficiently strong electric fields should alter the equilibrium positron distribution sufficiently to produce observable effects if the annihilation rates in these gases are also velocity dependent.

D. Direct Annihilation of Positrons in Krypton and Helium

1. Krypton

The information obtained for krypton, although limited, suggests a mechanism very similar to that in argon. The shoulder is characterized by a width of about 170 nsec atm, or about one half the value in argon. Noting that the positron-atom mass ratio, $m/M$, for krypton is $\frac{1}{2}$ the value for argon, and also taking into account the lower positronium formation threshold (7.1 eV in Kr), the observed shoulder width can be interpreted in terms of a scattering cross section somewhat less than 4 times the magnitude of the scattering cross section in argon.
2. Helium

On the basis of the experimental data presented in this thesis the evidence for a shoulder in helium is inconclusive. It would be highly desirable to obtain more detailed measurements in helium since the problem of the positron-helium interaction is likely to be more amenable to theoretical treatment than for any other gas for which experimental data can be obtained.

The electric field experiments in helium exhibited only a small dependence of the direct annihilation mean lifetime on the strength of the applied field. This suggests that in helium the direct annihilation cross section is closely approximated by an inverse velocity dependence. Such a velocity dependence is consistent with the observation that over the temperature range from 4.2°K to 298°K (Table XIV) the annihilation rate in helium is linearly dependent on the density.
APPENDIX A

HIGH PRESSURE CHAMBER DESIGN

A. Introduction

Detailed investigations into the design of a high pressure chamber were soon restricted to a vessel having spherical walls, since any other type of construction considered was either uneconomical, or inconvenient because of the excessive weight of metal required. Three primary specifications were set down.

(i) The chamber must withstand a working pressure of 1000 psi.
(ii) Adequate provision for a convection circulating gas purifier was to be incorporated.
(iii) Provision for producing a uniform electric field over a large active volume of the chamber was required.

Taking into account the general chamber requirements discussed in Chapter III, the overall design illustrated in Fig. 13 was decided upon.

Basically, the chamber consisted of two heavy aluminum flanges, to each of which was welded an aluminum hemisphere. A third similar flange contained the outlet pumping connection, the high voltage lead-through, and the purifier. Purification of the gas was carried out by passing the gas over a hot calcium-magnesium eutectic mixture contained in a corrosion resistant stainless steel tube. A water jacket at each end of the tube was provided to remove the heat conducted to the purifier ends. The design calculations are given briefly in the next section.

B. Design Calculations

In the calculations which follow frequent reference was made to
the detailed analysis presented by S. Timoshenko in "Strength of Materials - Part II". A gas pressure of 1000 psi is used in all the numerical calculations to obtain the working stresses at the various points in the chamber. The numerical values of the chamber dimensions labelled in Fig. 50, which depicts the region at the joint of the shell and the flange, are as follows: c = 5.5 in., a = 6.25 in. (= (d-c)/2), d = 7.0 in., x = 2.562 in., h = 2.25 in., h₁ = 0.5 in. The value given for h₁ is its mean value over the region of the weld area, the actual thickness varying from about 0.30 in. just above the weld area to 0.75 in. at the base of the weld. Construction of the metal hemispheres, which is discussed later, proved to be difficult, resulting in some nonuniformity in thickness of the finished shells. A conservative estimate of 0.30 in. is used for the mean shell thickness in the calculations. The sealing of the two outer flanges to the central flange is accomplished by an O-ring gasket seal whose mean diameter is 11.75 in.

A straightforward calculation yields the following membrane stress, $S_m$, in the hemispherical shell

$$S_m = \frac{pd}{2t} = 9,170 \text{ psi}$$

Here we have put $t = 0.30 \text{ in.}$ (the shell thickness), and $p = 1000 \text{ psi}$. The membrane stress is the pure tensile stress resulting from the internal gas pressure, in the region of the shell where edge effects are of no importance.

In addition to the membrane stress, other large stresses are produced at the joint of the hemispherical shell and the flange. The situation is depicted in Fig. 50. Due to the force $R$, the force per unit
FIG. 50. Schematic Diagram of the Joint Area Between Shell and Flange in the High Pressure Chamber in the Normal and Stressed Conditions.
length of the circumference produced by the internal gas pressure, and the opposing moment $M$ and other internal stresses in the flange, an equilibrium condition is achieved with the flange rotated through an angle $\theta$. The internal flange stresses are in the circumferential direction arising from the lengthening of the fibers on the inside edge of the flange and the compression of the fibers at the outer edge of the flange, as the flange rotates. An estimate of the moment $M$ is difficult to make since the degree of effectiveness of the bolting of the flanges in preventing rotation of the flange region outside of the bolt circle is not known. An approximate estimate for this moment is obtained by assuming that the fibers at the top and bottom edge of the flange are compressed and stretched respectively by an amount $(h/2)\theta$, yielding the result

$$M = \frac{1}{12} \frac{E h^3}{x}, \quad (1)$$

where $E$ is the modulus of elasticity.

The presence of the internal gas pressure would result in a radial expansion of the shell by an amount $y_1$, if indeed the shell was free to expand. The presence of the flange prevents such expansion at the edge of the shell. A small displacement of the shell, $y_2$, occurs at the base due to the rotation of the flange leaving a net "stress producing displacement" of $y = y_1 - y_2$. The resulting stresses are $P_o$ and $M_o$, the shearing force and moment per unit length of the circumference respectively. For the analysis which follows we assume that the spherical shell may be replaced by a cylindrical shell of the same thickness and radius. A longitudinal strip of this cylinder supported along its entire length by the cylinder walls on either side, results in a situation analogous to that
of a beam on an elastic foundation. This latter problem is well known and yields the results (equations (11) and (12) of Timoshenko)

\[ y = \frac{1}{2B^2D} (P_0 - BM_0) \]  

(2)

\[ \theta = \frac{1}{2B^2D} (P_0 - 2BM_0) \]  

(3)

for the deflection and slope of the end of the longitudinal strip. The quantities \( D \) and \( B \) are defined by

\[ D = \frac{Eh_1^3}{12(1-u^2)} \]

and

\[ B = \left[ \frac{2(1-u^2)}{c^2h_1^2} \right]^\frac{1}{2} \]

where \( u \) is Poisson's ratio, assumed to have a value of 0.316. The result for the deflection of the strip is readily found to be

\[ y = y_1 - y_2 = \frac{pc^2(1-u)}{2h_1E} - \frac{h \theta}{2} \]  

(4)

Returning to Fig. 50 and considering the equilibrium condition of the flange, we find that the net moment per unit length of the center line of the flange, \( M_c \), responsible for the rotation through the angle \( \theta \), is given by

\[ M_c = \frac{c}{a} \left[ R(d-c) + M_0 + P_0 h/2 - M \right] \]  

(5)

From equation (128) of Timoshenko, \( M_c \) is related to \( \theta \) by the expression

\[ \theta = \frac{12M_c a}{Eh_1^3 \ln(d/c)} \]  

(6)

Using the calculated quantities \( R = 3,140, D = .01157E, \) and \( B = 0.774, \) the equations (1) to (6) are readily solved yielding \( M_0 = 316 \text{ lb-} \)}
in., \( P_0 = 309 \) lb, \( M = 4,840 \) lb-in., and \( \theta = 13,080/E \). The resulting stresses obtained from these results are summarised in Table XV, together with the formulae used to obtain them.

The results for the purifier pipe and purifier connection given at the end of the foregoing table require some further comment. In both cases large stresses arise at the ends of the pipes where they are kept rigid and so prevented from free expansion under pressure. The bending stresses which occur at the end of a pipe whose end is fixed, when added to the other stresses occurring in the pipe, yield a total maximum stress given by

\[
s_{\text{max}} = 2.27 \frac{P}{r/e}
\]

where \( r \) is the inside radius of the pipe and \( e \) the wall thickness. For the aluminum pipe joining the purifier to the chamber and the stainless steel purifier tube, these quantities had values 0.412 in. and 0.113 in., and 0.525 in. and 0.133 in. respectively.

C. Chamber Materials and Construction

Construction of the chamber, except for the purifier tube, was entirely of aluminum alloy. The desirability of using aluminum was pointed out in Chapter III. In the construction of the three main flanges, wrought plate aluminum, Alcan 65ST6, was used because of its high strength and its availability in thicknesses up to 3 in. The aluminum hemispheres were "spun" by Art Metal Spinning Co., Vancouver. The spinning process requires the original plate material to be in the "O" temper so that cracking will not occur during the forming process. Considerable work hardening of the material occurs during spinning resulting in a much harder and stronger metal in the final hemispheres. Unfortunately, during the
TABLE XV

Summary of Stresses Occurring at Various Points in the High Pressure Chamber at a Gas Pressure of 1000 psi.

<table>
<thead>
<tr>
<th>Stress Location</th>
<th>Formula</th>
<th>Stress (psi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(i) Membrane stress is hemispherical shell.</td>
<td>$s = pc/2t$</td>
<td>9,170</td>
</tr>
<tr>
<td>(ii) Shear force at base of shell.</td>
<td>$s = P_o/h_1$</td>
<td>618</td>
</tr>
<tr>
<td>(iii) Stress due to bending moment $M_o$, at base of shell.</td>
<td>$s = 6M_o/h_1^2$</td>
<td>7,580</td>
</tr>
<tr>
<td>(iv) Stress due to bending moment $M$, at bolt circle of flange</td>
<td>$s = 6M/h^2$</td>
<td>5,740</td>
</tr>
<tr>
<td>(v) Stress at inner edge of flange due to rotation through angle $\theta$.</td>
<td>$s = E\theta h/2c$</td>
<td>2,680</td>
</tr>
<tr>
<td>(vi) Maximum stress in shell approximately equal to sum of (i) and (iii)</td>
<td></td>
<td>16,000</td>
</tr>
<tr>
<td>(vii) Maximum stress in aluminum pipe joining purifier to chamber.</td>
<td>eq. (7)</td>
<td>8,270</td>
</tr>
<tr>
<td>(viii) Maximum stress in stainless steel purifier pipe (neglecting thermal stresses).</td>
<td>eq. (7)</td>
<td>8,950</td>
</tr>
</tbody>
</table>
welding process some annealing of the metals in the weld zone occurs, again reducing their strength.

Upon the recommendation of Mr. Malloy (1964) the welding of the hemispherical shells to the flanges was carried out using a 56s filler wire. A full 100% penetration weld was achieved.

A summary of the relevant elastic properties of the materials used in the chamber construction appears in Table XVI. Comparison of this table with the results given in Table XV quickly reveals that the crucial stress area is at the base of the shell. In fact, at the pressure of 1000 psi, it is possible that the base of the shell is stressed to its yield point.

D. Chamber Fittings and Details

1. High Voltage Lead-Through

The electric fields anticipated in the course of the experiments required a high voltage source of about 25 kV. A satisfactory high voltage lead-through was obtained using an ordinary ignition engine spark plug which, with the addition of an epoxy filler surrounding the center electrode, also served adequately as a high vacuum seal.

2. Main Outlet Valve

In order to be able to evacuate the chamber efficiently and thoroughly a valve with a large orifice and good high vacuum properties was required. The additional requirement of high pressure operation necessitated some compromise. A hydromatics Flo-Ball valve (series 715) with a 3/8 in. orifice and equipped with teflon seats and O-ring seals was obtained for this purpose.

3. Flange Bolts
### TABLE XVI

Summary of the Relevant Elastic Properties of the Materials used in the Construction of the High Pressure Chamber

<table>
<thead>
<tr>
<th>Part</th>
<th>Original Material</th>
<th>Yield Strength (psi)</th>
<th>Ultimate Strength (psi)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hemisphere #1</td>
<td>B53S - 0</td>
<td>17,000</td>
<td>36,000</td>
<td>B53S-H32, work hardened condition of hemisphere</td>
</tr>
<tr>
<td>3/8 in. plate Al.</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hemisphere #2</td>
<td>D54 - 0</td>
<td>18,000</td>
<td>40,000</td>
<td></td>
</tr>
<tr>
<td>½ in. plate Al.</td>
<td></td>
<td></td>
<td></td>
<td>D54-H, ½ hard, work hardened condition of hemisphere</td>
</tr>
<tr>
<td>Flanges - Al</td>
<td>65ST6</td>
<td>21,000</td>
<td>45,000</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>16,000</td>
<td>30,000</td>
<td>65ST4, approximate weld zone condition</td>
</tr>
<tr>
<td>Aluminum pipe connecting</td>
<td>65S</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>purifier to chamber</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stainless steel purifier</td>
<td>304</td>
<td>87,000</td>
<td></td>
<td></td>
</tr>
<tr>
<td>pipe</td>
<td></td>
<td>44,500</td>
<td></td>
<td>Strength at 1200°F</td>
</tr>
</tbody>
</table>
The pressure load at 1000 psi carried by each of the flange bolts is 9,050 lb. Additional loading occurs during tightening of the nuts so that an adequate margin of safety must be provided. To achieve this, steel bolts ("Grade 5") with a minimum breaking load of 70,000 lb. were obtained.

E. Pressure Testing of Chamber

A hydrostatic pressure test of the chamber was carried out at the Civil Engineering Dept., U. B. C. The pressure was cycled five times up to a maximum of 1700 psi with no apparent difficulties developing. Further testing was carried out with a gas filling of nitrogen at a pressure of 1075 psi in order to test the purifier at elevated temperatures. Again no difficulties were apparent with the purifier at a temperature of 550°C.

F. Design of Electric Field Grid Structure

The discussion in Chapter II pointed out the need for making measurements of positron annihilation spectra at electric field strengths of at least 1000 V/cm (in argon at 10 atm). The grid structure designed to supply a uniform field of this magnitude over a large active volume of the chamber is illustrated in the photographs of Fig.14. The equatorial plane of the chamber was made an equipotential by the use of a fine wire mesh wound on a brass ring, to which a high positive voltage could be applied. On either side of this center plane, copper rings situated at regular intervals of ½ in. were connected consecutively by high voltage resistors. The ends of the cylindrical volume formed by these rings were closed by pieces of sheet aluminum electrically grounded to the chamber. A uniform axial electric field was thus established which was directed away from the center on either side of the central equatorial plane. The complete grid
assembly was mounted on strips of teflon which served both to position the rings and provide electrical insulation.

A technical difficulty encountered as a result of the high electric fields existing between the central rings and the chamber walls was that of electrical breakdown of the gas. The literature dealing with the electrical breakdown characteristics of gases provided limited information, and the results which were found were used only as a guide. A summary of the results of the dielectric strengths of gases obtained from Druyvesteyn and Penning (1940), and Von Hippel (1954) is shown in Table XVII. The large spread in the reported values for air likely result from the range of experimental conditions encountered. With a spacing of 0.55 in. between the grid rings and the chamber walls, an electric field of 45 kV/cm is developed when 25 kV is applied to the central ring. For the breakdown voltages quoted above for argon and helium, working pressures of 10 atm and 70 atm in these gases respectively should yield no difficulties.

Construction of the grid rings was performed by winding 0.204 in. diameter copper wire on a form (9 1/2 in. in diameter), cutting the spiral so formed and then soldering the ends of the individual wire loops. In order to reduce corona discharge all the rings were chromium plated, and the chamber itself was highly polished and all sharp edges rounded.

The high voltage resistors were Type : BAEW (1/2 watt, 2500V), 20 megohm, precision (3%) resistors, obtained from "Resistor Products Company". In all, twenty four resistors were used in the grid giving a net input resistance to the grid of 120 megohms.

The requirements placed on the fine wire mesh in the center plane
TABLE XVII

Dielectric Strengths of Gases in a Uniform Electric Field

<table>
<thead>
<tr>
<th>Gas</th>
<th>Pressure (atm)</th>
<th>Breakdown Voltage V/(cm atm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>1</td>
<td>28</td>
</tr>
<tr>
<td>Air</td>
<td>1</td>
<td>40</td>
</tr>
<tr>
<td>Air</td>
<td>27</td>
<td>18.5</td>
</tr>
<tr>
<td>N₂</td>
<td>40</td>
<td>8.7</td>
</tr>
<tr>
<td>A</td>
<td>1</td>
<td>8</td>
</tr>
<tr>
<td>He</td>
<td>68</td>
<td>2.3</td>
</tr>
</tbody>
</table>

of the chamber were twofold. Firstly, an equipotential had to be established as uniformly as possible in this plane, and secondly, the mesh was to have a high transmission for positrons crossing this boundary. To achieve the desired result, thirteen holes were evenly spaced along the inner circumference of the center brass ring, and, using number 30 nichrome wire, the mesh was formed by connecting each of the thirteen points to every other point. A check of the uniformity of the field produced by the grid structure was obtained by using a two-dimensional analogue constructed on conducting paper. The potential plot obtained for this model revealed that irregularities in the field along planes parallel to the central mesh were restricted to within 0.1 in. on either side of this equatorial plane. Irregularities produced between the grid rings and the chamber walls became negligible at a distance of about 0.2 in. from the inside edge of the rings. The overall effective volume thus formed inside
the chamber had a diameter of 9.5 in. and a length of 12.0 in.

Initial attempts to operate the grid at potentials of 15 kV and above in argon at 10 atm failed because of discharges occurring inside the chamber. Since the high voltage supply was protected by an overload relay which opened at a current of about 1.5 ma, small momentary discharges were sufficient to interrupt the circuit. A 5 megohm series resistor placed in the external circuit virtually eliminated this phenomena and satisfactory operation was achieved up to 21 kV in argon at 10 atm pressure. Similarly, no difficulty was encountered using helium at 50 atm pressure with 22 kV on the grid.
A. Introduction

The time sorter described in this paper has been used over the past two years in a program involving the measurement of the lifetime of positrons in gases. Although the time sorter was designed primarily for such measurements in the range of 1 - 600 nsec, the time scale of the instrument can be readily extended to cover a range of many microseconds. The principle of pulse overlap is utilized in the time to amplitude conversion to generate an output pulse whose amplitude is proportional to the time separation of the input pulses. Flexibility in the choice of the desired time scale is achieved by replacing the normal delay line shaped square pulses by fast trigger circuits which generate fast rising square pulses of adjustable length.

The advantages and disadvantages of the various time conversion methods used in time sorters have been discussed extensively by Bonitz (1963). A frequent problem encountered in experiments using time sorter experiments is that of count rate effects since the 'singles' rate in the individual counters is generally high. Use of the overlap principle minimizes this difficulty since it automatically requires a coincidence between the input pulses before the converter is actuated. Thus, high counting rates are restricted to the nonlinear pulse circuits which precede the actual converter.
An electronic resolution (full width at half amplitude) of less than $2 \times 10^{-10}$ sec is obtained with this circuit, and the overall time resolution of the system using NaI crystals (2 in. long, $1\frac{2}{3}$ in. diameter) and RCA 7264 photomultipliers is about $3\frac{1}{2}$ nsec for the $1.28 - 0.51$ MeV gamma rays from $^{22}$Na (in aluminum).

A schematic diagram of the time sorter indicating the basic units of the system is shown in Fig. 51. The pulses appearing at the lettered points in this diagram are displayed in the correspondingly lettered parts of Fig. 52. This latter figure ((a)-(g)) portrays the timing sequence of the pulses appearing in the circuit for a time separation of the input events of 400 nsec.

The two trigger circuits, driven by the threshold discriminators, generate the fast rising square pulses which activate the time to amplitude conversion circuit. A recovery circuit restores the converter to its quiescent state about 6 μsec after the occurrence of a coincidence event. In order to prevent analysis of input events occurring in the wrong timing sequence, a negative time eliminator is incorporated into the system. This circuit produces a coincidence signal whenever the order of the input events is correct.

B. Time to Amplitude Conversion Circuit

The time to amplitude conversion circuit, similar in principle to the one described by Simms (1961), is shown in simplified form in Fig. 53 and in detail in Fig. 55. In the quiescent state the two switching transistors, $T_1$ and $T_2$, share the constant current $I$ from the 3.9K resistor, whereas the transistor $T_3$ is cut off. A single input pulse, sufficient to cut off the corresponding switching transistor, only switches the current.
to the other member of the $T_1$ - $T_2$ pair so that no output pulse results. If $T_1$ and $T_2$ are simultaneously cut off, however, the current $I$ is switched via $T_3$ to the integrating capacitor $C$. Thus if $I$ is constant the amplitude of the voltage pulse appearing on $C$ is directly proportional to the time during which both $T_1$ and $T_2$ are off. Transistor $T_3$ (a silicon 2N2411) was normally biased so that the emitter was $\approx 1/10$ V above the base. The foregoing arrangement proved to yield sufficient tolerance that such precautions as matching the two switching transistors $T_1$ and $T_2$ was found unnecessary.

The necessity of utilizing devices with very small leakage currents in the circuitry associated with the integrator governed the choice of the 2N2411 transistor ($T_3$), the 1N3604 silicon junction charging diode, and also the 2N2219 silicon junction transistor serving as the first stage of the output amplifier. This output stage consisted of a stabilized feedback amplifier with a gain of about 5.

C. Threshold Discriminators and Trigger Circuits

As evident in Figs. 52(a) - (c), the region of overlap of the two input pulses is defined by the leading edge of the delayed input and the trailing edge of the prompt input. In order to achieve the desired timing stability the width of the prompt input pulse must therefore be accurately defined. To first approximation, operation of the circuit is insensitive to the amplitude of the two inputs, but because of the finite risetime of the pulses, maximum stability results only if the amplitudes of the inputs are maintained constant.

The input pulses to the current switch are produced by the fast trigger circuits illustrated in Fig. 54. Incorporated in the input of each
trigger circuit is a tunnel diode discriminator which provides an accurate discrimination threshold and also produces a fast pulse to operate the trigger circuit. The threshold level, which is readily adjustable by the bias resistor is set to eliminate the photomultiplier noise pulses. The input pulses to these discriminators are produced by conventional limiter circuits (Jones (1960)) delivering 25 mA of current.

A detailed description of the circuit used for the delayed trigger has been given by Jones (1963). This circuit is capable of producing pulses whose rise and fall times are less than 10 nsec, and whose width can be varied over a wide range by changing the value of the single timing capacitor, $C_1$. The triggering sensitivity of the circuit is normally between 0.15 and 0.25 V and is thus readily triggered by the tunnel diode discriminator.

The desired time stability in the width of the prompt trigger output was obtained by relying on the propagation time of a delay line in a feedback circuit. The circuit used is similar to the one described by Sugarman and Higinbotham (1962). Thus, although the frequency response of the delay time itself may be relatively poor, its use in a circuit of this type enables the production of an output pulse with an accurately determined pulse width, yet maintaining very short rise and fall times.

The rise and fall times of the trigger pulses (when connected to the remaining circuitry in the normal operating manner) were measured with a Tektronix Type 661 sampling oscilloscope and found to be 10 and 15 nsec respectively for each of the pulses.

The large voltage pulses appearing at the collector of $T_4$ (see Fig. 52(k)) require the use of a transistor with a large collector-base
voltage rating. A 2N2904 transistor, which also has a 5V emitter-base rating, was selected for this purpose.

In each of the trigger circuits a series combination of an inductance and a resistor is placed across the tunnel diode to insure that the tunnel diode is in its low voltage state in the DC condition.

D. Negative Time Eliminator (NTE)

The random coincidence background counts which are always present in coincidence experiments consist equally of events in which the prompt trigger is activated before the delayed trigger and events in which the delayed trigger precedes the prompt trigger. This latter group of random coincidences which are termed 'negative time' events are always distinguishable from the true coincidences because of the different time sequence of the input pulses. The circuit denoted as the Negative Time Eliminator (or NTE) in Fig. 55 serves to eliminate this group of coincidences and consequently reduces the random coincidence background by a factor of two. Operation of this circuit is as follows. The prompt trigger pulse is sharply differentiated at the base of $T_9$ which is normally conducting. Thus the leading edge has no effect whereas the back edge turns $T_9$ on and causes current to flow through tunnel diode $D_2$. $D_2$ is biased so that a coincidence is required between this pulse and the delayed trigger pulse before regeneration occurs. When fired by $D_2$, the trigger circuit consisting of $T_{10}$ and $T_{11}$ produces a $3 \mu \text{sec}$ output pulse at the collector of $T_{11}$ which can then be used in the gating circuitry for the kicksorter.

From the foregoing it is noted that if the back edge of the delayed trigger pulse precedes the back edge of the prompt trigger in time, no output from the NTE is produced. In addition, for the proper operation
of the pulse overlap system employed, it is essential that the width of
the delayed trigger pulse be greater than that of the prompt trigger.
The pulse widths used for the two triggers were 0.8 and 0.7 μsec respectively
as shown in Fig. 52(h) and (i). A consequence of this choice of widths does
mean, however, that those negative time coincidences characterized by a
time separation of 0 - 100 nsec do lead to a NTE output and are therefore
interpreted as positive time events. This is not a serious limitation
however, since all such events produce a constant amplitude output (the
maximum) from the time sorter corresponding to complete overlap of the
two trigger pulses. Nevertheless, it is undesirable to have all these
pulses occurring at the point in the spectrum corresponding to the real
zero of time (the prompt peak). For this reason a delay of about 35 nsec
was inserted between the delayed trigger and its limiter so that the
position of the prompt peak and the position corresponding to maximum
pulse overlap were well separated on the spectrum.

E. Recovery Circuit

The integrating time constant at the collector of $T_3$ is
sufficiently long that negligible leakage occurs during the entire inte-
gration period. It is thus necessary to discharge the integrating
capacitor after each coincidence event. The recovery circuit is fired
by the negative pulse appearing at the common collectors of $T_1$ and $T_2$ when-
ever the current switch is operated. After amplification by $T_{14}$, this
pulse is applied to the trigger circuit consisting of $T_{15}$ and $T_{16}$ which
produces a 6 μsec pulse. $T_{18}$ is turned on at the back edge of this pulse,
thus discharging the integrating capacitor until it is clamped to the 5V
line by the 1N3604 diode. The discharge pulse appearing at the collector
of $T_{18}$ is shown in Fig. 52(g). In a time of about 8 µsec after application of the input pulses the time sorter circuit is completely restored.

**F. Performance Characteristics**

The performance characteristics of the time sorter were measured with the normal fast-slow coincidence system. In order to compensate for the intrinsic time delay in the slow channels the time sorter output pulse was delayed prior to entering the kicksorter. This delayed time sorter pulse and the kicksorter gating pulse are shown in Fig. 52(d) and (f) respectively. A triple coincidence between both slow channels and the NTE was required to produce the gating pulse.

The time resolution of the time sorter (obtained for Na$_{22}$ in aluminum) is shown in Fig. 56 together with resolution curves obtained under several different experimental conditions. NaI(Tl) crystals, $\frac{1}{2}$ in. in diameter and 2 in. long, mounted on RCA 7264 photomultipliers were used as the detectors. In normal operation the single channel energy selectors were set in such a manner than one analyzer spanned the 510 keV full-energy peak (with a window of about 150 keV) and the other the energy region from 0.65 to 1.4 MeV to accept the 1.28 MeV gamma ray. A pile-up rejector (McGervey and Walters (1964)) was also incorporated in the system to eliminate effects due to high count rates. Each of the curves shown in Fig. 56 was obtained for pulse rates of 2500/sec and 2000/sec in the delayed and prompt trigger circuits respectively. Under these conditions a time resolution of 3.6 nsec (full width at half amplitude) was obtained.

Under certain experimental conditions somewhat harmful effects on the resolution curve were observed. A widening of the resolution curve
in the region of channel 86 occurred if the lead shield between the counters was removed thereby permitting scattering from one counter to the other. Thus the geometrical positioning of the detectors is important if good resolution is desired. Removal of the pile-up rejector from the system resulted in effects very similar to that described above. It was further found, that an increase in the trigger circuit count rates to about 4000/sec again resulted in a widening of the resolution curve in the same region, even with the pile-up rejector in operation. This latter effect has not been fully investigated.

G. Time Calibration

The time calibration of the time sorter, shown in Fig. 57, was carried out using a double pulse generator (Appendix D) whose pulse separation was accurately determined by a passive variable delay line. From such integral calibrations however, it is difficult to obtain accurate values for the relative channel widths, which are required for the accurate determination of lifetimes and intensities from complex spectra. What is required is a measure of the differential linearity of the time scale. A standard technique for measuring the differential linearity is to use independent gamma ray sources for each of the counters and record the random coincidence spectrum. Unfortunately, this method is characterized by very slow coincidence counting rates and necessitates very high single channel counting rates if adequate statistics are to be obtained in a reasonable length of time. To overcome this difficulty a circuit (which we refer to as a random time generator) was built (Appendix C) which, when triggered by a pulse from one of the counters, subsequently produced an output which was random in time with respect to the triggering pulse.
Results using this circuit are shown in Fig.58 together with the random coincidence spectra obtained using independent gamma ray sources. The latter are included for comparison only. The deviation from constancy of the channel widths is a maximum of ±10% over the entire time range and considerably less over most of the range. Nonlinearities in the kicksorter itself, amounting to ±4%, also contribute to the observed overall nonlinearities. The rapid rise in the curve of Fig.58(a) at the upper end of the time scale in the region of channel 100 is an effect due to the random time generator.

The purpose of the delay inserted between the delayed trigger and its limiter, it was noted earlier, was to separate, on the time spectrum, the position of the prompt peak and the position corresponding to maximum pulse overlap. An additional advantage is gained from this arrangement since the coincidence counts recorded between these two positions represent the random coincidence background counts added to the spectrum.

H. Stability

The overall stability of the instrument was found to be very good under ordinary laboratory conditions. Over a period of many weeks in actual operation the shift in the position of the prompt peak was less than 3 nsec on a time scale 700 nsec in length, and the time calibration could be reproduced to within ±1%. Variations in the position of the peak were found to be directly proportional to the variation of the current in the current switch. For this reason a well stabilized power supply (Technipower, Model M-31.5 - 0.200 A) with 0.05% regulation was used for the time sorter.
FIG. 51. Schematic Time Sorter Diagram Indicating the Basic Units of the System. The Pulses Appearing at the Lettered Points are Shown in Fig. 52.
FIG. 52. Voltage Pulses at Various Points in the Circuit of the Time Sorter. Figures (a) - (g) Show the Timing Sequence of Pulses for Input Events Separated by 400 nsec.
FIG. 53. Simplified Circuit Diagram of Time-to-Amplitude Converter.
FIG. 54. Trigger Circuits.
FIG. 55. Detailed Circuit Diagram of Time - to - Amplitude Converter.
FIG. 56. Time Sorter Resolution Curve (for Na\textsuperscript{22} in Al) Under Different Experimental Conditions.
FIG. 57. Time Sorter Calibration Curve. Measurements on Two Different Time Scales are Indicated.
FIG. 58. Time Sorter Differential Linearity Curves Obtained by the Two Methods Discussed in the Text.
APPENDIX C

A RANDOM TIME GENERATOR FOR TIME SORTER LINEARITY MEASUREMENTS

W. Falk, G. Jones, R. Orth

Submitted to "Nuclear Instruments and Methods" for consideration as a letter to the editor.

In order to determine the performance of a time sorter it is necessary to know the linearity of the time scale. The integral linearity measured by making a normal time calibration curve is easily obtained, but detailed analyses of time distributions frequently require knowledge of the differential linearity. Previously published methods for establishing the differential linearity rely either on high precision integral linearity measurements (Roddick and Lynch (1964)) or on coincidences between uncorrelated pulses (Weisberg and Berko (1964)). The former type of measurement is extremely difficult to perform, while the latter yields statistically significant results only over long counting periods.

We report here the construction of a pulse generator capable of producing output pulses random in time compared with the start of one of the input pulses, at a rate comparable with that of this same input pulse.

The system operates in the following fashion. Square pulses of width equal to the desired time range, $T$, are produced at a random rate. In our case, shaped pulses from a scintillation detector were employed. These pulses are distributed both to one channel (start) of the time sorter and to the random time generator, henceforth called RTG. A second input of the RTG is connected to a pulse generator producing pulses of very short duration, at a maximum frequency, $I/T$. Output pulses from the RTG then
occur for each coincidence between these two input pulses. These coincidence pulses are then shaped and distributed to the other channel (stop) of the time sorter.

The circuit diagram of the RTG is illustrated in Fig. 59. The sensitivity of the tunnel diode \( D_1 \) is so adjusted that it triggers only when the two input pulses are coincident, and not at all for single pulses at either input. Since the two pulses are uncorrelated, the duration of the time interval between arrival of the first pulse and production of the coincidence pulse is random over the period 0 to \( T \) secs. In order to achieve this in practice, it was necessary to isolate the DC voltage supplies to the two input transistors using low pass filters.

The system described here, however, suffers from one minor difficulty. Whereas the random coincidence pulses used in the calibration of the time sorter are produced when the pulse from the pulse generator (pulse B), follows that from the scintillation counter (pulse A), coincidence pulses are also produced whenever a pulse B precedes a pulse A by a time interval less than the width of pulse B. The output pulses, in this situation, are simultaneous with the start of pulse A, and result in excessive counts near the prompt channels of the time sorter. In order to reduce the number of prompt counts to a value comparable with the number over the rest of the time scale, it is necessary to reduce the width of pulse B to as small a value as possible. By using the shaping circuit involving the tunnel diode \( D_2 \), a width of \( \leq 20 \) nsec was achieved.

Calibration of a time sorter was obtained using the circuit described in this paper in conjunction with the ancillary pulse generators already referred to. The block diagram of the system is illustrated in
Fig. 16(a). The differential linearity of the time sorter measured in this way is shown in Fig. 58. The rapid rise in the curve of Fig. 58(a) in the region of channel 100 is evidence of the finite width of pulse B. Fig. 58 also illustrates the differential linearity obtained using two uncorrelated gamma ray sources.
FIG. 59. Circuit Diagram of Random Time Generator. Pulse A is Produced by a Photomultiplier Viewing a Na$^{22}$ Source. Pulse B is a 100 KC Pulse approximately 50 nsec Wide. All Capacitors are in Microfarads.
APPENDIX D

DOUBLE PULSE GENERATOR WITH PRECISION PULSE SEPARATION

The circuit shown in Fig. 60 is a double pulse generator for producing fast pulses whose time separation can be controlled with great precision. The separation of the two pulses is determined by a passive delay line (General Radio Type 314 - S86, 200 ohms impedance) of 1/2 \(\mu\)sec total delay. A square pulse (10\(\mu\)sec in length) generated by the monostable circuit consisting of \(T_2\) and \(T_3\) produces a prompt trigger and a delayed trigger at the points in the circuit labelled "A" and "B" respectively. Both of these trigger pulses feed identical stages containing tunnel diode inputs, which, upon regeneration, produce outputs with very fast rise-times from emitter followers \(T_4\) and \(T_5\). The accurate triggering threshold provided by the tunnel diodes results in negligible timing jitter of the output even though the input pulse has a relatively slow rise time. Measurements with a sampling oscilloscope indicated output pulse rise-times of about 7 nsec.

A detailed calibration of the delay line was carried out over the entire 500 nsec delay using a Tektronix Type 661 sampling oscilloscope on the 5 nsec/cm scale. This procedure, in conjunction with a calibration of the oscilloscope time scale (using a Hewlett-Packard Model 5243L Electronic Counter and a Model 5251A Frequency Converter to Calibrate the 100 Mc signal of the sampling oscilloscope) permitted the time separation between the prompt and delayed outputs to be determined to an absolute accuracy of \(\pm 0.3\%\) (of the full delay).
FIG. 60. Double Pulse Generator With Precision Pulse Separation.
APPENDIX E

RANGE AND STOPPING TIME DISTRIBUTIONS OF Na\(^{22}\) POSITRONS IN GASES

In this appendix, the range and stopping time distributions of positrons (emitted by a Na\(^{22}\) source) which have slowed down in a gas are calculated for a number of different gases. The data for the energy loss and range of positrons was obtained from NBS Circular 577 (National Bureau of Standards, 1956). Since the data tabulated in this circular are only shown down to energies as low as 10 keV, the calculated range (and similarly the stopping time) represents the range of a positron having an initial energy \(E\) and a final energy of about 10 keV. However, in the energy region below 10 keV the rate of energy loss by the positron is so large that the additional contribution to the range and stopping time in slowing down to an energy of about 50 eV is assumed to be negligible. It should be clarified that the "range" calculated here actually corresponds to the total path length of the positron, whereas the actual range of the positron would in all cases be less than that indicated.

Fig. 61 shows the dependence of the positron range and stopping time in argon (at 25°C) as a function of the initial positron energy. The range and stopping time are expressed in terms of the pressure independent units, 'cm atm' and 'nsec atm' respectively. Since the positrons emitted from Na\(^{22}\) are distributed over a range of energies up to a maximum of 542 keV (Macklin et al. (1950)) it is desirable to know what fraction of the positrons has a range within a certain range interval \(dR\), and similarly, what fraction has a stopping time within the interval \(dT\). If we let \(N(E)\) represent the energy spectrum (the beta-spectrum of Na\(^{22}\)), \(N(x)\) the positron range distribution, that is, the number of positrons stopping
per unit range interval, and \( N(t) \) the stopping time distribution, then the following relationships exist between the distributions.

\[
N(x) = N(E) \frac{dE}{dx}
\]

where \( x \) is the range at the initial energy \( E \) and \( \frac{dE}{dx} \) is the differential energy loss. Also

\[
N(t) = N(E) v \frac{dE}{dx}
\]

where \( t \) is the stopping time for a position of initial energy \( E \) and \( v \) is its velocity at this energy. The stopping time is obtained by numerical integration of the expression

\[
t(E) = \int_E^0 (v \frac{dE}{dx})^{-1} \, dE
\]

The above distributions are not normalized, and their expression in terms of the pressure independent units referred to earlier remains to be carried out. Introducing the appropriate factors for argon, the two distributions shown in Fig. 62 are obtained. It is noted that the stopping time distribution has a maximum at 1.5 nsec atm and extends to a maximum of 7.5 nsec atm. Thus in a typical experimental situation with a pressure of about 10 atm, the maximum stopping time of the positrons is 0.75 nsec and the bulk of the positrons stop in a time of approximately half this value. Thus, in most experimental situations the slowing down time is so short as to be within the time resolution of the instrumentation used in these experiments.

In the case of the range distribution, a comparison can be made to the measured values of Celitans and Green (1963) who find that the distribution ends at about 130 cm atm compared to the calculated maximum path length of 156 cm atm (at 20°C). It should be remarked however that use of these measured values in the experiments of Tao et al. (1964) lead to
conclusions which are inconsistent with those of Falk and Jones (1964).

The design of the pressure chambers for minimum positron annihilation in the chamber walls employed these results for determining the optimum physical size.

Of further usefulness is a knowledge of the fraction of positrons with a range greater than some value R and the fraction with a stopping time greater than T. This is readily obtained by integrating the curves of Fig. 62, the results being displayed in Fig. 63.

While the results plotted in Figures 61-63 were determined specifically for argon, they may easily be generalized to apply to other gases. An examination of the differential energy loss, dE/dx of the different gases, reveals that in the energy range between 50 keV and 550 keV the dE/dx curves, as a function of energy, are to a good approximation the same shape for different gases, the various curves differing only by a constant (within ±3%) scale factor. At lower energies somewhat greater variations occur but this is of small consequences. Table XVIII lists the appropriate scaling factors for various gases relative to argon. Use of the graphs for an arbitrary gas requires that the range and the stopping time be multiplied by the scaling factor appropriate to the gas in question.

**TABLE XVIII**

Range and Stopping Time Scaling Factors for Various Gases Relative to Argon

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<td>CO₂</td>
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FIG. 61. Positron Range and Stopping Time in Argon at 25°C.
FIG. 62. Range and Stopping Time Distributions of Na$^{22}$ Positrons in Argon (at 25°C) During Initial Slowing Down Period. The Distributions are Not Normalized.
FIG. 63. Fraction of Na\textsuperscript{22} Positrons, F(R), with Range Greater than R and Fraction, F(T), with Stopping Time Greater than T.
APPENDIX F

DATA ANALYSIS

A. Linearization of Time Scale

Time distributions recorded with a time sorter suffer from distortions due to nonlinearities in the electronic analyzing and recording equipment. A schematic kicksorter histogram representing an annihilation spectrum is shown in Fig. 64 where the nonlinearities are indicated by the unequal widths of the channels. As a result of these unequal channel widths a twofold distortion occurs. Firstly, the number of counts registered in a channel is proportional to the width of that channel rather than proportional to the amplitude of the function alone, and secondly, the time scale itself is distorted.

For purposes of making compensating corrections to the data it is assumed that the number of counts registered in a channel represents the intensity (or amplitude) of the spectrum at the midpoint of that channel. In addition, each channel is assigned a relative width, $r_i$, obtained by the method described in Appendix C (see also Appendix B and Chapter IV.) The relative width, $r_i$, is defined as $w_i/\bar{w}$, where $\bar{w}$ is the average width of all the channels. Then, denoting the number of counts in channel 'i' by $N_i'$, a first correction to the data is obtained by replacing $N_i'$ by $N_i'/r_i = N_i$. This gives the corrected intensity (counts per unit channel) at the midpoint of each channel. However, since the midpoints of these channels are not equally spaced, a further correction is required. This correction is made as follows. The dashed lines labelled a, b, c,... etc. in Fig. 64 represent the midpoints of a new set of regularly spaced
FIG. 64. Schematic Annihilation Spectrum Depicting the Nonlinearities Produced by the Unequal Time Widths of the Channels.
channels whose width is $\bar{w}$. The intensity at each of these points, a, b, c, .... etc. is then obtained by making a linear interpolation between the corrected counts per channel, $N_i$. If the spectrum changes rapidly over the interval of one channel such a linear interpolation introduces a significant distortion. For this reason the interpolation procedure was begun at the peak of the distribution by making the midpoints of the 'old' and 'new' set of channels coincide at the peak of the distribution as shown in Fig.64. However, since the detailed shape of the prompt peak was of no particular interest in the present experiments, slight distortions introduced into this part of the spectrum were of little concern.

The data on the relative channel widths is shown in Table XIX.

B. Calculation of Mean Lifetimes and Intensities from Annihilation Spectra

Considering times greater than those characteristic of the shoulder and/or the prompt peak region of the spectra, the annihilation spectra are characterized by two exponential components - the direct annihilation component, and the orthopositronium component. A determination of the direct annihilation mean lifetime also requires the simultaneous determination of the orthopositronium lifetime and the intensities corresponding to these two components. The following method used in determining these four quantities, involves maximizing the likelihood function as discussed by Orear (1958).

Let the observed number of counts in channel $k$ be $N_k$. Then, from the Poisson Distribution, the probability, $p$, that this number $N_k$ is observed is

$$p = (\gamma_k)^{N_k} e^{-\gamma_k/N_k}$$  \hspace{1cm} (1)

where $\gamma_k$ is the average number of counts that would be observed in channel
TABLE XIX

Relative Channel Width Data for the Time Sorter - Kicksorter System. The Data are Entered Beginning with Channel 100 and Read from Left to Right Across the Page

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<sup>1</sup>The sum of the relative channel widths for channels 11-100 is 88.98. From the calibration curve of Fig.57 the time interval between these channels is 216.3 nsec. Thus the time width of a channel whose relative width is 1, is 2.43 nsec.

<sup>2</sup>By the same method as above, the time width of a channel whose relative width is unity is 4.68 nsec. The channel width data presented for this time scale does not correspond identically to the curve shown in Fig.58 which was obtained at a later date. The analysis of the data was performed with the tabulated values shown above, however.
k in many measurements of the quantity \( N_k \). Thus, the overall probability of obtaining a particular distribution of counts, \( N_1, N_2 \ldots N_k \), is given by

\[
L = \prod_k (y_k)^{N_k} e^{-y_k/N_k}
\]

(2)

where \( \prod_k \) denotes the product of the terms over the range of values of \( k \).

The quantity \( L \) is called the likelihood function for the distribution (Orear (1958)). Also,

\[
W = \ln L = -\sum_k y_k - \ln(N_k!) \tag{3}
\]

Now assuming that the counts in the distribution consist of counts from the two exponential components and a known constant background \( B \), one may write

\[
y_k = I_1 e^{-(k-1)/\tau_1} + I_2 e^{-(k-1)/\tau_2} - B \tag{4}
\]

where \( \tau_1 \) and \( \tau_2 \) are the lifetimes of the two exponential components and \( I_1 \) and \( I_2 \) the counts per channel in channel 1 for each of these components respectively. This latter equation remains valid even when the mean lifetimes are comparable to the time-widths of the channels. For the particular observed distribution, \( N_k \), the problem is now to find those values of \( \tau_1, \tau_2, I_1, \) and \( I_2 \) which will yield the maximum value of the function \( L \). Clearly, a maximum value for \( L \) also corresponds to a maximum value for \( W = \ln L \). Thus, introducing the expression for \( y_k \) from (4) into equation (3) and differentiating with respect to these four unknown parameters to yield the extremum conditions, four equations are obtained.

Unfortunately, these equations are not linear in the unknowns, and hence cannot be solved directly to yield the values of \( \tau_1, \tau_2, I_1, \) and \( I_2 \).
However, if approximate initial estimates for these quantities are known, an iterative procedure can be used which yields first order corrections to these quantities from the four extremum-condition equations. This iterative procedure using these corrected estimates then yields the best values for the lifetimes and intensities. The computational time on the IBM 7040 was about 20 sec for a calculation involving six iterations.

Initial estimates for $\tau_1$, $\tau_2$, $I_1$, and $I_2$ were obtained graphically and were typically within $\pm 10\%$ of the computer calculated values. Under some conditions, initial estimates which deviated by much more than $\pm 10\%$ from the best values resulted in a divergence of the iterative procedure. Also, if the lifetimes of the two exponentials were comparable, as for example in Fig. 29, poor convergence resulted. In such cases it was usually necessary to keep one of the lifetimes, usually the ortho-positronium lifetime, fixed and then determine the three remaining parameters using the Maximum Likelihood procedure.

A useful feature of the Maximum Likelihood technique is that the actual probability for obtaining the particular observed distribution can be obtained (i.e., the Likelihood function, $L$). This latter information proved very valuable in selecting the best values for the parameters when the program convergence was poor.

It can be shown (Orear (1958)) that the standard deviation of a parameter $x$, is given by

$$\Delta x = \left(-\frac{\partial^2 L}{\partial x^2}\right)^{-\frac{1}{2}}$$

This error was also calculated for each of the four parameters $\tau_1$, $\tau_2$, $I_1$, and $I_2$. 
C. Effect of Instrumental Time Resolution on the Shape of the Annihilation Spectra

The effect of the instrumental time resolution on the shape of the annihilation spectra has been investigated by folding the experimental instrumental time resolution into a number of different input spectra. A convolution integral was performed using the resolution curve, labelled 'A' in Fig.56 and several exponential functions, and also with a function (curve 'a' of Fig.65) simulating the 'shoulder' in the argon spectra. Fig.65 shows the calculated output spectra. For the two exponential functions, curves 'c' and 'd', a significant deviation from a pure exponential occurs only in the region within about 5 nsec of the 'zero of time', as indicated by the arrow in the diagram. Also, for the input function 'a', the output curve 'b' is remarkably faithful reproduction of the input except for the initial portion of the curve. In this region, however, this discrepancy between the two curves is of minor importance since the presence of the prompt peak in the normal spectrum obscures this region in any case. For clarity, the origin of the time axis has been displaced for curves 'a' and 'b'.

The data for the resolution curves shown in Fig.56 are given in Table XX.
<table>
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<tr>
<th>Channel Number</th>
<th>Curve A</th>
<th>Curve B</th>
<th>Curve C</th>
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FIG. 65. Effect of Instrumental Time Resolution on Hypothetical Annihilation Input Spectra. Curve "b" Represents the Output Spectrum for the Input 'a'. Curves 'c' and 'd' are the Outputs for Single Exponential Inputs.
A. Derivation of the Diffusion Equation Neglecting the Thermal Motion Of the Scattering Atoms

1. Introduction

The diffusion equation governing the velocity distribution of positrons in a monatomic gas is derived for the case when the thermal motions of the scattering atoms can be neglected. Provision is made for including the effects of a uniform DC electric field applied to the gas. While the general theory of such diffusion processes has been discussed by Chapman and Cowling (1952) and other writers, in this appendix is presented a summary of the relevant parts of the general theory required to develop a diffusion equation appropriate to the case of positrons in a monatomic gas when subjected to a uniform DC electric field.

Let \( f(\vec{v}, t) \) be the average number of positrons at time \( t \) in the volume element \( d\vec{v} \) around \( \vec{v} \). The number of positrons in this volume element will change with time due to the following mechanisms:

(i) Elastic collisions of the positrons with the atoms of the gas. This will result in positrons being scattered both into and also out of the volume element.

(ii) Annihilation of free positrons (direct annihilation).

(iii) Positronium Formation.

(iv) Drift in velocity space due to the presence of an electric field.

The effects of excitation and ionization levels in the scattering atoms will
not be considered since the energy region of interest here ranges from zero to just above the positronium formation threshold which is lower in energy than any of the excitation levels in the noble gases.

2. Elastic Collisions

Let the density of atomic scatterers be $N$ per unit volume. Then the time rate of change of the number of positrons in the volume element $d\mathbf{v}$ (Fig. 66) due to scattering is readily shown to be

$$
\frac{d\mathbf{v}}{dt} \left[ \frac{\partial f(\mathbf{v}, t)}{\partial t} \right]_s = Ndw \int d\mathbf{v}' f(\mathbf{v}', t) q(\mathbf{v}', \psi) \\
-Nd\mathbf{v} \int d\mathbf{v} f(\mathbf{v}, t) q(\mathbf{v}, \psi')$$

(1)

where $q(\mathbf{v}, \psi)$ is the elastic scattering cross section per unit solid angle for a scattering angle $\psi$ between the positron and the atom. The subscript $s$ denotes that the time rate of change of $f$ is due to scattering. $\mathbf{v}'$ is the velocity that a positron must have prior to a scattering event so that after scattering through an angle $\psi$, it will be in the volume element $d\mathbf{v}$. The first term on the right hand side of equation (1) thus represents the rate at which positrons are scattered into $d\mathbf{v}$, and similarly, the second term represents the rate at which positrons are scattered out of the volume element $d\mathbf{v}$.

Since we are assuming that the thermal motions of the atoms can be neglected, the appropriate expression for describing the energy loss of a positron in an elastic collision is given by equation (6) of Chapter II. Rewriting this equation for our present purposes we obtain

$$\Delta v/v = -(m/M)(1 - \cos \psi)$$

(2)

The volume element $d\mathbf{v}$, when expressed in terms of the solid angle it subtends at the origin, is $\mathbf{v}^2 d\omega d\mathbf{v}$ and similarly, $d\mathbf{v}' = \mathbf{v}'^2 d\omega' d\mathbf{v}'$, where the
various quantities are depicted in Fig. 66. In order that all the posi-
trons in the velocity interval $dv'$ fall into the velocity interval $dv$, after scattering through an angle $\psi$, the following relationship, obtained from (2), must be satisfied.

$$dv' = (v'/v)dv$$

Using these results in (1) and dividing through by $dv$ one finds

$$\left[ \frac{\partial f(v', t)}{\partial t} \right] = Nv \int_{w'} \left[ f(v', t)\frac{v'}{v}q(v', \psi) - f(v, t)q(v, \psi) \right] dw' \quad (3)$$

At this stage it is convenient to express $f$ in terms of spherical
coordinates $\theta$, $\phi$, and $v$ where $\theta$ is the polar angle.

The velocity $v'$ appearing in (3) is very nearly equal to $v$
permitting one to make a Taylor expansion about $v$, keeping only first order terms. This result, when combined with equation (3) yields

$$\left[ \frac{\partial f(\theta, \phi, v, t)}{\partial t} \right] = Nv \int_{w'} \left[ f(\theta, \phi, v, t) - f(\theta, \phi, v, t) \right] q(v, \psi) dw'$$

$$+ \frac{N}{v^2} \frac{m}{M} \frac{\partial}{\partial v} \int_{w'} (1 - \cos \psi) q(v, \psi) v^4 f(\theta', \phi', v, t) dw' \quad (4)$$

The above result, as it stands, is prohibitively difficult to handle unless some further simplifications are introduced. Using the method of Lorentz (1952), $f$ is expanded in spherical harmonics and only the first two terms retained (Morse et al. (1935)). This expansion is valid for the case of a uniform electric field applied in the direction of the polar axis. Thus, since $f$ does not depend on the azimuthal angle $\phi$, one obtains from this expansion,

$$f(\theta, v, t) = f_o(v, t) + \cos \theta f_1(v, t) \quad (5)$$

The first term $f_o$ represents the isotropic part of the distribution while the second term represents the anisotropy introduced by the presence of the
electric field. Introducing this expansion for \( f \) into equation (4) it is readily shown that the term involving \( f_1 \) is smaller in the second integral than in the first integral by a factor of order \( m/M \). Consequently, only the term involving \( f_0 \) is retained from the second integral with the result that

\[
\left[ \frac{\partial f(\theta, v, t)}{\partial t} \right]_s = N v \int_{w^*} (\cos \theta' - \cos \theta) f_1(v, t) q(v, \psi) d\psi + \frac{N}{\sqrt{v^2 M}} \frac{m}{M} \int_{w^*} (1 - \cos \psi) q(v, \psi) \nu^4 f_0(v, t) d\psi
\]

(6)

In evaluating the above integral we adopt the method used by Holstein (1946). Letting \( \psi \) be the azimuthal angle referred to an arbitrary axis perpendicular to the vector \( \vec{v} \), the solid angle \( d\omega^* \) can be written as \( \sin \psi \, d\psi \, dv \). Also

\[
\cos \theta' = \cos \theta \cos \psi + \sin \theta \sin \psi \cos \psi
\]

Evaluating the integral we obtain

\[
\left[ \frac{\partial f(\theta, v, t)}{\partial t} \right]_s = -\cos \theta f_1(v, t) r_c(v) + \frac{1}{\sqrt{v^2 M}} \frac{m}{M} \left[ \nu^2 f_0(v, t) r_c(v) \right]
\]

(7)

where we have defined

\[
r_c(v) = N v \int_0^{\pi} 2\pi (1 - \cos \psi) \sin \psi \, q(v, \psi) \, d\psi
\]

(8)

The quantity \( r_c(v) \) is the collision rate for momentum transfer, and the integral on the right, the total elastic momentum transfer cross section (Morse et al (1935)) which we denote simple by \( q(v) \). Thus

\[
r_c(v) = N v q(v)
\]

(9)

This momentum transfer cross section is to be distinguished from the ordinary total cross section

\[
\int_0^{\pi/2} 2\pi \sin \psi \, q(v, \psi) \, d\psi
\]

(10)
These two cross sections become identical when the differential cross section \( q(v, \psi) \) is independent of the scattering angle \( \psi \). In the discussion of the elementary thermalization theory of positrons in Chapter II this distinction was overlooked for the sake of simplicity.

3. Drift Term Due to the Application of an Electric Field

We consider now the effect on the distribution function of a uniform electric field applied in the direction of the polar axis. This effect is most readily understood by analogy to the familiar continuity equation in real space. For the diffusion equation, the drift in velocity space is represented by the acceleration, \( \bar{a} \), of the positron due to the electric field. Thus

\[
\nabla_v \cdot (\bar{a} f) + \frac{\partial f}{\partial t} = 0
\]

Noting that \( \bar{a} \) does not depend on the velocity this reduces to

\[
\left( \frac{\partial f}{\partial t} \right)_t = -\bar{a} \cdot \nabla_v f
\]

This result is in accord with that given by Teutsch and Hughes (1956).

Introducing the expression (5) for \( f \) and noting that \( \bar{a} \) has a component in the Z-direction only, we obtain

\[
\frac{\partial f}{\partial v_z} = \cos \theta \frac{\partial f}{\partial v} + \frac{f_1}{v} \left[ 1 - \frac{v_z^2}{v^2} \right] \left( \frac{v_z}{v} \right)^2 \frac{\partial f_1}{\partial v}
\]

Since we are neglecting spherical harmonics of order higher than the first, \( v_z^2 \) is replaced by its average value \( v^2/3 \). Thus

\[
\left( \frac{\partial f(n, v, t)}{\partial t} \right)_t = -a \cos \theta \frac{\partial f_0(v, t)}{\partial t} - \frac{a}{3v^2} \frac{\partial}{\partial v} \left[ v^2 f_1(v, t) \right]
\]

4. Annihilation of Free Positrons and Positronium Formation

We denote by \( \sigma(v) \) and \( s(v) \), the cross sections per atom for
direct annihilation and positronium formation respectively. Due to the combination of these effects the distribution function \( f \) will change with time according to

\[
\frac{\partial f(\theta, v, t)}{\partial t} = -Nv \left[ \sigma(v) + s(v) \right] f(\theta, v, t)
= - \left[ r_a(v) + r_f(v) \right] \left[ f_o(v, t) + \cos \theta f_1(v, t) \right]
\]

(15)

where \( r_a(v) \) and \( r_f(v) \) represent the direct annihilation rate and the positronium formation rate respectively, and are defined by

\[
r_a(v) = Nv\sigma(v), \quad r_f(v) = Nvs(v)
\]

(16)

5. Simplification of the Diffusion Equation

The overall time rate of change of the distribution function due to the various processes is now given by the sum of equations (7), (14), and (15). The resulting equation can be separated into two other equations, the coefficients of \( \cos \theta \) yielding the first of these equations, and the remaining terms, the other. These equations are

\[
\frac{\partial f_1}{\partial t} = -\frac{\partial f_0}{\partial t} - f_1 r_c(v) - \left[ r_a(v) + r_f(v) \right] f_1
\]

(17)

and

\[
\frac{\partial f_o}{\partial t} = -\frac{a}{3v^2} \frac{\partial}{\partial v} (v^2 f_1) + \frac{1}{v^2} \frac{m}{M} \frac{\partial}{\partial v} \left[ v^3 f_c r_c(v) \right] - \left[ r_a(v) + r_f(v) f_0 \right]
\]

(18)

The coupled equations (17) and (18) are still difficult to solve and further simplification is desirable. Fortunately, this simplification is readily effected under the present circumstances. In Section G of Chapter II it was pointed out that the collision rate, \( r_c(v) \), in argon was of the order of \( 10^{12} \) per sec. Some elementary manipulations of equation (17) soon reveal that if the fractional change in \( f_0 \), in the time
Interval $1/r_c(v)$, is much less than one, then this equation may be inte-
grated immediately to yield

$$f_1 = -\frac{a}{r_c(v)} \frac{f_0}{v}$$

(19)

This condition is certainly satisfied since the distribution function
can change but little in a time equal to the mean time interval between
collisions.

Substituting for $f_1$ from (19) we obtain the final result for $f_0$

$$\frac{\partial f_0}{\partial t} = \frac{1}{v^2} \frac{\partial}{\partial v} \left[ \frac{a^2 v^2}{3 r_c(v)} \frac{\partial f_0}{\partial v} + \frac{m}{M} v^3 r_c(v) f_0 \right] - \left[ r_a(v) + r_f(v) \right] f_0$$

(20)

The equation for $f_1$ is of no further interest to us since an
examination of (5) reveals that the net number of positrons in this com-
ponent is zero, and hence the experimentally observable quantity - the
annihilation rate - is completely determined by $f_0$ alone. As a check on
the validity of our expansion according to equation (5), however, we
calculate $f_1$ using as $f_0$ the equilibrium distributions, equations (27)
and (28) of Chapter II. In both cases one obtains

$$f_1/f_0 = \frac{3}{2^{1/2}} \sqrt{m/M}$$

when $v$ is set equal to $u$. Since even for the case of helium this ratio is
only about $10^{-2}$, the neglect of higher order terms in the expansion is
justified.

B. Extension of the Diffusion Equation to Take into Account the Thermal

Motions of the Scattering Atoms

The differential equation for the distribution function $f_0$, derived in the previous section, is valid only for the case where the
thermal motions of the scattering atoms can be neglected. For a complete
description of the positron distribution function valid at energies comparable with the energy of the scattering atoms, an extension of the above result is obviously required. As shown in Chapman and Cowling (1952), this extension can be effected by including in equation (20) a term of the form

\[
\frac{1}{2} \frac{\partial}{\partial v} \left[ \frac{m}{M} v^2 r_c(v) \frac{kT}{mv} \frac{\partial f}{\partial v} \right]
\]

Thus

\[
\frac{\partial f}{\partial t} = \frac{1}{2} \frac{\partial}{\partial v} \left[ \frac{a^2 v^2}{3r(v)} + \frac{m v^2 r_c(v) kT}{mv} \frac{\partial f}{\partial v} + \frac{m v^2 r_c(v)}{M} \right]
- \left[ r_a(v) + r_f(v) \right] f
\]

where T is the temperature of the gas and k is Boltzmann's constant. The f subscript has been dropped since this distinction is no longer required.

C. Numerical Solution of the Diffusion Equation

In order to facilitate the programming of equation (21) for numerical solution, it was rewritten in terms of the quantities defined at the beginning of Section G, Chapter II. The positron velocity was expressed in terms of the dimensionless quantity, \( x = v/v_0 \), and the various cross sections were each defined in units of \( \pi a_o^2 \), as follows:

- \( Q(x) = \sigma(xv_0)/(\pi a_o^2) \), the elastic momentum transfer cross section.
- \( Z(x) = \sigma(xv_0)/(\pi a_o^2) \), the direct annihilation cross section per atom.
- \( S(x) = s(xv_0)/(\pi a_o^2) \), the positronium formation cross section per atom.

Introducing these quantities, the differential equation (21) becomes
\[
\frac{df}{dt} = 25.26 \frac{m}{P} \left[ \left( 1.192 \times 10^{-5} \frac{M}{m P^2} \right) \frac{1}{xQ(x)} + \frac{xQ(x)}{3} \right] \frac{d^2 f}{dx^2} \\
+ \left[ (1.192 \times 10^{-5} \frac{M}{m P^2}) \left( \frac{1}{x^2Q(x)} - \frac{1}{xQ''(x)} \frac{dQ(x)}{dx} \right) + (1 + x^2)Q(x) + x \frac{dQ(x)}{dx} \right] \frac{df}{dx} \\
+ \left[ 4xQ(x) + x^2 \frac{dQ(x)}{dx} - \frac{M}{m} \left( xZ(x) + xS(x) \right) \right] f \]
\]

(22)

Here the unit of time, \( t \), is expressed in nsec.

A program to solve the partial differential equation (22) was developed by A. Fowler of the Computing Center at the University of British Columbia, using the Crank-Nicholson method of numerical solution. In carrying out the computation, the 'time-axis' was divided into intervals of .05 nsec and the 'x-axis' into intervals of 0.25. Thus, typically, a matrix of order 2600 x 100 was involved in a calculation of a spectrum extending to 130 nsec. The machine time for such a calculation on the IBM 7040 computer was just under 13 minutes.

Since the calculated annihilation spectrum corresponds to the time differential of the total number of positrons in the distribution, a very high degree of accuracy has to be obtained for the numerical solution if the spectrum is to be determined to an accuracy of about 1\%. In practice, the error in the magnitude of \( f \) was sufficiently large, that when solutions were obtained for zero values of \( Z(x) \) and \( S(x) \), the total number of positrons would slowly increase with time. In order to overcome this difficulty a renormalization procedure was introduced into the program. This renormalization procedure simply requires that the total number of positrons at the time \( t + \Delta t \) be equal to the number at time \( t \), minus the number that have been removed in the time \( \Delta t \) by direct annihila-
lation and positronium formation.

For convenience, the cross sections were expressed in the following form:

\[
Q(x) = \frac{1}{x} (a + bx + cx^2 + dx^3) \tag{23}
\]

\[
Z(x) = \frac{1}{x^n} (\alpha + \beta x + \gamma x^2) \tag{24}
\]

where all the constants could be readily varied. The positronium formation cross section was simulated by a ramp function of arbitrary slope, changing to a constant value at a selected value of \( x \). The information for the initial velocity distribution, \( f(x,0) \), was entered into the computer by reading one hundred numbers from data cards.

The computer output consisted of the following information:

(i) The velocity distribution of the positions, \( f(x,t) \).

(ii) The direct annihilation spectrum

\[
y(t)_a = A \int_0^\infty Z(x)x^3f(x,t)dx \tag{25}
\]

where \( A \) is an arbitrary factor.

(iii) The positronium formation rate,

\[
y(t)_f = A \int_0^\infty S(x)x^3f(x,t)dx \tag{26}
\]

(iv) The spectrum from the annihilation events of orthopositronium,

\[
y(t)_o = \frac{3}{4} A e^{-\lambda T} \int_0^T \left\{ e^{T/T} \int_0^\infty S(x)x^3f(x,T)dx \right\} dT \tag{27}
\]

(v) The observed spectrum consisting of the sum of events from both direct annihilations and positronium annihilations,

\[
y(t) = y(t)_a + \frac{1}{2} y(t)_f + B y(t)_o \tag{28}
\]

(vi) The total number of direct annihilation events,

\[
N(t)_a = \int_0^t y(t)_a dt \tag{29}
\]
(vii) The total number of positronium formation events,

\[ N(t)_f = \int_0^t y(t)_f \, dt \]  

(30)

The foregoing quantities were printed for a series of preselected values of the time, \( t \). The orthopositronium annihilation spectrum represented by equation (27) results from a consideration of the delay in the annihilation of the orthopositronium atoms, which are characterized by a mean lifetime, \( \tau \). In Equation (28), representing the observed spectrum, the parapositronium component contributes the amount \( 1/\phi y(t)_f \), since these annihilations appear 'promptly' upon formation of the parapositronium atom. The orthopositronium contribution to \( y(t) \) requires a consideration of the relative detection efficiency of a 3-quantum event to that of a 2-quantum event. This relative detection efficiency is taken into account by the factor \( B \).

The fractional increase in positronium formation, \( \phi \), in an electric field is readily shown to be

\[ \phi = \frac{R + g}{g(1 + R)} \]  

(31)

where \( g \) is the fraction of positrons forming positronium at zero field and \( R = N(\infty)_f / N(\infty)_a \).

During initial testing of the program a number of consistency checks were made as follows:

(i) For an annihilation cross section inversely proportional to the positron velocity, a single exponential decay should result for this component. This result was indeed obtained for several choices of the elastic scattering cross sections.

(ii) For a direct annihilation cross section and positronium
formation cross section equal to zero, a stationary equilibrium distribution should be obtained after sufficiently long times have elapsed. This was observed for the case of an applied electric field as well as for the case of zero electric field. The analytical expressions for the distribution function for cases of a constant scattering cross section and a cross section proportional to $1/x$ are readily derived, as was shown in Section H of Chapter II. For the latter case, a comparison of the theoretical and computer calculated distribution functions is shown in Fig. 67. While the general agreement between these distributions is quite good, some further improvement in the numerical solution is desirable.
FIG. 67. Comparison of Analytical and Numerical Solutions of the Diffusion Equation. The Curves are Shown for Argon Assuming a Scattered Cross Section Inversely Proportional to the Velocity of the Positron, and Using the Value $\epsilon/P = 100$.
APPENDIX H

EXPERIMENTAL DATA FOR THE ANNIHILATION SPECTRA IN ARGON

The experimental data presented for the annihilation spectra in argon have been subjected to the linearization procedure described in Appendix F and have also had the random coincidence background subtracted. Other experimental details pertinent to these spectra are given in Table VI. The data are entered beginning with channel 100 and read from left to right across the page.
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APPENDIX I

EXPERIMENTAL STUDIES OF THE ANNIHILATION OF POSITRONS IN THE NOBLE GASES

W. R. FALK AND G. JONES

Department of Physics, University of British Columbia, Vancouver, British Columbia

Received May 5, 1964

ABSTRACT

The direct annihilation rate of positrons in He, A, and Kr has been studied. Deviation from an exponential for the initial portion of the time distribution of these events is observed and is interpreted as a velocity dependence of the direct annihilation rate. The product of the gas pressure and the width of the non-exponential portion of the time distribution has a value of 270 nsec atm and is approximately independent of the pressure. An estimate of the fraction of positrons forming positronium in argon yields a value of 0.37 ± 0.03. Using this result, a check of the statistical ratio of ortho- to para-positronium formation indicates that 69 ± 7% of the positronium is formed in the ortho state.

1. INTRODUCTION

The early experimental work by Deutsch (1953), and later work by Falk and Jones (1963) and Paul and Saint-Pierre (1963) on the direct annihilation rate of positrons in gases indicated that a single exponential characterizes the time distribution for these annihilation events. Recent measurements (described in this paper) on several of the inert gases have revealed, however, a pronounced deviation of the time distribution of the direct annihilation events from a single exponential. Tao et al. (1964) and Paul (1964) have, independently, observed this behavior in argon and argon–nitrogen mixtures. These effects were not observed previously (Falk and Jones 1963) because of poorer stability and time resolution during the course of the earlier work.

The time distributions of the direct annihilation events in argon, krypton, and an argon–helium mixture all revealed a relatively "flat" region immediately following the prompt peak. After the termination of this region a simple exponential decay is observed. An interpretation of these results in terms of a velocity dependence of the annihilation rate is presented in Section 4. The noble gases were selected for study because of the absence of low-lying excitation levels which can be excited by a positron. Tao et al. (1963) have shown that the slowing-down time for a positron in such gases is relatively long and times of the order of 100 nsec are required to slow a positron from an initial energy of 9 eV to an energy of 1 eV in argon at 1 atm pressure. A dependence of the direct annihilation rate on the positron velocity should be most readily observable in media in which the positron slowing-down time is extended over a long period, as in the noble gases.

The experimental arrangement used in these measurements permitted an estimate of the fraction of positrons forming positronium in argon and argon–NO mixtures to be made. A check of the statistical ratio of ortho- to para-positronium formation was further obtained from these time distributions.

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2. EXPERIMENTAL ARRANGEMENT

The experimental arrangement used was the standard one employed by other workers in this field (Paul and Saint-Pierre 1963; Duff and Heymann 1962). In our case, a transistorized time-to-amplitude converter (to be published), similar in principle to that of Simms (1961) was used. A Na$^{22}$ source ($\approx 8 \mu$C) deposited on 0.2 mg/cm$^2$ aluminum was placed at the center of a spherical aluminum chamber 14 inches in diameter. The 0.10-in. wall thickness enabled the chamber to withstand pressures of up to 11 atm, yet caused minimal absorption of the annihilation gamma rays. With this arrangement the number of positrons annihilating in the chamber walls was negligible for pressures as low as 7 atm in argon, and annihilations in the source backing less than 2% of the total. Since positrons annihilating in the chamber walls and source produce events which appear in the prompt coincidence peak, it is necessary to keep these to a minimum if details in this region of the time spectra are to be observed. The counters consisted of two $2 \times 2\frac{1}{2}$ in. NaI crystals mounted on RCA-6810 photomultipliers. An overall time resolution of 6 nsec was obtained using a Na$^{22}$ source in aluminum.

3. RESULTS

(a) Direct Annihilation Spectra

The annihilation time spectra for positrons in pure argon at two different pressures* are shown in Fig. 1. A single exponential can be fitted to these two curves in the region to the right of the shoulder, marked S. The deviation of the points from an exponential shape in the region below channel 50 is due to the detection of some of the longer-lived orthopositronium annihilations. The single-channel pulse-height analyzer in the 500-keV channel was set with a narrow window encompassing the 500-keV full-energy peak in order to suppress the registering of the three-quantum orthopositronium events and so exhibit the direct component more clearly.

Our interpretation of the various components of the time spectra is in general agreement with that given by Tao et al. (1964). The peak in channels 83–86 is attributed to parapositronium annihilations together with the small amounts of source and wall annihilations. The broad flat region to the left of the shoulder, as well as the single exponential to the right of S, is attributed to the direct or free annihilation of positrons. In order to check this picture and to show that the flat region of the spectrum was associated with a velocity dependence of the annihilation rate, a small quantity of CO$_2$ (\approx 2%) was added to the argon. The electric-field experiments of Marder et al. (1956) on positronium formation indicate that CO$_2$ is very effective in slowing positrons. Results for this mixture are also illustrated in Fig. 1. In this case the experimental points beyond channel 80 can be fitted by a single exponential whose mean life is in good agreement with that obtained for the exponential portion of the decay curve for pure argon at the same pressure. The analysis of these curves given in Table I indicates that the counts to the left of the

*Note added in proof: All gas pressures quoted have been normalized to 0 °C.
FIG. 1. Time distribution of positron annihilations in argon. The indicated points are the raw data with the random coincidence background subtracted.

TABLE I
Analysis of results for pure argon and for argon with a small admixture of CO₂ (Fig. 1). The direct annihilation events are taken as all the remaining events after subtraction of the ortho- and para-positronium events.

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</table>

shoulder S (but excluding those in the prompt peak) should be identified with the direct component of the positron annihilation. It has been assumed that the addition of the 2% CO₂ to the argon does not appreciably alter the fraction of the positrons forming positronium.
Similar deviations from a simple exponential decay were also observed in Kr and in He as illustrated in Fig. 2. The helium results were obtained using a mixture of He and A to provide sufficient stopping power for the positrons.

The mean life of the positrons in the exponential portion of the direct annihilation component varies inversely as the gas pressure for the pure argon spectra of Figs. 1 and 2. A comparison of our results was made with those obtained from Dirac's formula (Deutsch 1953) in which a plane-wave approximation is assumed. A value for the direct annihilation rate of 1.5 times the Dirac rate (assuming that all 18 electrons of the argon atom participate) was obtained. This result is in good agreement with the value of 1.37 reported by Tao et al. (1964) and the result of 1.43 obtained from Paul and Saint-Pierre (1963) when calculated on the basis of 18 electrons per argon atom.

Since the purity of the gases is a crucial factor in these experiments, several precautions were carefully observed. The chamber was cleaned thoroughly and baked before assembly and, prior to filling with gas, was pumped to pressures below $10^{-5}$ mm Hg. The gases A and He were obtained from standard cylinders with quoted maximum impurities of 0.02% and 0.01%, respectively.
For the case of Kr, measurements were made before and after a calcium-magnesium purifier (Ruff and Facchini 1952) was inserted into the gas system. The results obtained were the same in both cases.

(b) Positronium Formation

Accurate measurements on the fraction of positrons forming positronium in a gas have not been made, although estimates have been given (Marder et al. 1956). This measurement, while not of direct relevance to the study of the free annihilation of positrons, is of considerable interest in understanding the overall behavior of positrons in gases.

An adequate experimental check of the statistical ratio of ortho- to para-positronium formation has not been made. The present experimental arrangement with very low source absorption and negligible annihilation of positrons with the chamber walls greatly facilitates such a measurement. Only preliminary results will be given here.

Four time spectra were obtained, one with pure argon, and three others with varying amounts of NO added to the argon. Two of these curves are shown in Fig. 3. For the curve with NO added no evidence of any ortho-

Fig. 3. Time distribution of positron annihilations in argon and argon–NO mixture. The indicated points are the raw data with the random coincidence background subtracted.
positronium events is visible. NO is known to quench orthopositronium very strongly, so that the addition of even a small percentage of this gas causes nearly all the orthopositronium formed to decay by two photons with a greatly reduced lifetime. It is relatively simple to compute the fraction of positrons forming positronium for these latter spectra with NO–argon mixtures, since all the decay events are via two-quanta emission so that the detection efficiency is the same for each process.

The presence of NO in argon does, however, affect the amount of positronium formed because of the additional ore gap contributed by the NO. A plot of the percentage of positronium formation as a function of NO concentration is shown in Fig. 4. When extrapolated to zero NO concentration, it is seen that about 37% of the positrons form positronium in pure argon. From the time spectrum for pure argon shown in Fig. 3 an estimate of the statistical ratio of

![Diagram](image)

Fig. 4. Positronium formation in argon–NO mixtures.

...to para-positronium can now be made. If we let \( f \) denote the fraction of the positrons forming positronium, and \( f_q \) the fraction forming orthopositronium, then it can easily be shown that

\[
\frac{N_p}{N_d + N_o} = \frac{f(1 - q) + \epsilon}{1 - f + fq} ,
\]

where \( N_p \) is the observed number of counts in the prompt peak, and \( N_d \) and \( N_o \) are the observed direct and orthopositronium counts respectively. The factor \( \epsilon \) allows for the small amount of source annihilation and has a value
of about 0.02. The relative detection efficiency of three-quantum annihilation events to two-quantum events and the partial quenching of the orthopositronium lifetime are taken into account by the factor \( \gamma \), which has been estimated to be approximately 0.4. The result of Duff and Heymann (1962) on the quenching of orthopositronium in argon was used in the above calculation. Variations of \( \pm 0.1 \) in \( \gamma \) affect the calculated value of \( q \) by less than 2%. Putting in the appropriate values of \( N_B, N_A, \) and \( N_0 \) obtained for pure argon, and using the previously obtained value of 0.37 for \( f \), one obtains \( q = 0.69 \pm 0.07 \). This result is to be compared to the value of 0.75 expected statistically.

4. DISCUSSION

(a) Direct Annihilation

An interpretation of the annihilation process in noble gases which yields the observed results is as follows. An energetic positron emitted into a gas at a pressure of several atmospheres is slowed to an energy of tens of electron volts in a time that is short compared to the width of the "prompt" peak observed in Figs. 1 and 2. A positron which has a kinetic energy somewhere below that of \( E_{th} \), the threshold for positronium production, continues to lose energy via elastic collisions with the gas atoms and at the same time has a relatively small probability of annihilation. On reaching some energy \( E_0 \), the probability of annihilation per second increases rapidly to a constant value, which depends on the nature of the gas. This is the value characterizing the exponential decay beyond the shoulder, \( S \). The position of the shoulder \( S \) would then be determined by the time required for a positron to slow down from \( E_{th} \) to \( E_0 \).

Further evidence for this interpretation follows.

(i) In Fig. 1 the point \( S \) for argon at 6.1 atm corresponds to a delay of about 45 nsec after the prompt peak. In terms of the analysis of Tao et al. (1963) this corresponds to the time required for a positron to slow from \( E_{th} \) to an energy \( E_0 \), of the order of 1 eV. This latter analysis assumes a constant elastic scattering cross section of \( 1.5\pi a_0^2 \)—the value obtained by Marder et al. (1956).

(ii) Again from the analysis of Tao et al. (1963), the product of the gas pressure and the time required to reach the energy \( E_0 \) should be independent of the pressure. For the three pure argon spectra this product has values of 241, 273, and 294 nsec atm for pressures of 4.8, 6.1, and 9.4 atm, giving an average value for the product of about 270 nsec atm. The zero of time was taken at the center of the prompt peak. Considerable uncertainty exists in the location of the shoulder \( S \), and this may have contributed to the spread in the above values. Comparison of our results with those of Tao et al. (1964) indicates that our mean value of the pressure slowing-down product of about 270 nsec atm is three times larger than their value of 90 nsec atm. This ratio is reduced only about 10% if allowance is made for the slightly different definition of the shoulder width employed in the two cases. It is indeed difficult to see how a value larger than the correct one could be obtained. The stability of the prompt-peak position was excellent (less than 1 nsec maximum drift over...
a period of many weeks), and the effect of the finite resolution time would tend to smooth out the shoulder effect rather than increase its width. On the other hand, the effect of some polyatomic impurities could certainly reduce the shoulder width considerably. Paul (1964) indicates a value of about 300 nsec atm for the pressure slowing-down product, in approximate agreement with our result.

An annihilation probability which increases rapidly for decreasing positron velocity as suggested in the preceding discussion may be the result of an attractive interaction between the positron and the gas atom at low energies. Such an interaction for the scattering of positrons on atomic hydrogen has been suggested by Bransden (1962). A similar effect involving positrons scattered by helium is discussed by Paul and Saint-Pierre (1963) and Wallace (1964).

(b) Positronium Production

Tao et al. (1964) state that approximately 40% of the total counts are within the prompt peak of the time spectra after allowance for the annihilations with the chamber walls is made. Since wall annihilations are high for a chamber of the size employed, the difficulties involved in making an accurate allowance for these events would seem to preclude making quantitative estimates of the relative intensities of the various components. In the geometry characterizing the measurements described in this paper, however, wall effects are negligible. This permits the following conclusions to be made. From Fig. 3 for the pure argon curve we obtain the figures:

Total number of counts in the peak, \( N_p \), equals 4580,

Total number of remaining counts (direct and orthopositronium), \( N_t \), equals 24800.

Thus, 15.6% of all the observed counts fall in the prompt peak. However, \( N_p \) includes not only the parapositronium annihilations but also all the source annihilations (about 600). Furthermore, \( N_t \) includes only about one-half of the orthopositronium events (because of the single-channel analyzer setting). Clearly less than 15% of the total annihilation events in the gas fall within the prompt peak of the time spectrum.

Our results do not preclude the possibility of formation of the collision complex postulated by Tao et al. (1964). However, the value of 0.37 ± 0.03 obtained in Section 3 for the fraction of positrons forming positronium (which is in agreement with other estimates ranging from 0.3 to 0.4 (Marder et al. 1956)), together with the value of \( q = 0.69 ± 0.07 \) previously obtained in Section 3 for the fraction of positronium forming the ortho state, strongly supports the view that essentially all of the positrons either annihilate directly (that is, are observed in the direct component) or form positronium.

Further experiments are underway to establish the effect of static electric fields on the annihilation of positrons in the noble gases.

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REFERENCES

WALLACE, P. R. 1964. Private communication.
BIBLIOGRAPHY


Malloy, Mr. (1964). The Aluminum Co. of Canada Ltd., Vancouver, B. C.
Private Communication.


