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THE ANNIHILATION OF POSITRONS IN ARGON

ABSTRACT

The annihilation of positrons in Argon has been investigated as a function of Argon density and applied electric field using the technique of lifetime measurements. Lifetime spectra were analyzed using the maximum likelihood method of curve fitting. Results obtained at zero electric field yielded a linear dependence on density for the direct annihilation rate of $(5.6 \pm 0.1) \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$, with some evidence of non-linearity at densities greater than 10 amagats. The density dependence of the long-lived component of the time spectra indicated a zero density intercept of $(7.2 \pm 0.4) \times 10^6 \text{ sec}^{-1}$ in agreement with the theoretical value of the free orthopositronium annihilation rate ($7.2 \times 10^6 \text{ sec}^{-1}$). In addition, an orthopositronium quenching rate of $(0.29 \pm 0.04) \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$ was obtained from the linear dependence of the orthopositronium annihilation rate on density.

The electric field dependence of the direct annihilation rate and orthopositronium formation has been measured and is used to provide an internally consistent picture of the behaviour of positrons in

a gas under the influence of an applied electric field. Furthermore, these results have been compared with theoretical results for the direct annihilation rate obtained from one parameter representations of the effective positron-Argon atom interactions. It is shown that, while such potentials are successful in describing the low-energy elastic-scattering of electrons from noble gas atoms, they are inadequate for the case of positrons. However, consideration of the way in which the direct annihilation rate changes as a function of electric field leads to an upper limit of $15\pi a_0^2$ for the momentum-transfer cross-section for positrons in Argon at thermal energies. Such an estimate is shown to be independent of any assumption concerning the effective positron-Argon atom interaction.

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Elementary Quantum Mechanics	F. A. Kaempffer
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Cosmic Rays and High Energy Physics	J. B. Warren
Electronic Instrumentation	F. K. Bowers

PUBLICATIONS AND PAPERS

W. Falk, G. Jones and R. Orth, A Random Time Generator for Timesorter Linearity Measurements. Nucl. Inst. & Methods 33, 345 (1965).

W. R. Falk, P.H.R. Orth and G. Jones, Effect of Electric Field on Positron Lifetimes in Argon and Helium. Phys. Rev. Letters 14, 507 (1965).

G. Jones, W. R. Falk and P.H.R. Orth, Positron Annihilation in the Noble Gases, p.372, IVth International Conference on the Physics of Electronic and Atomic Collisions (abstracts of papers), Science Bookcrafters Inc., N.Y. (1965).

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Garth Jones and Robert Orth, Annihilation of Positrons in Argon Gas. Bull. Am. Phys. Soc. II, 11, 749 (1966).

THE ANNIHILATION OF POSITRONS IN ARGON

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ABSTRACT

The annihilation of positrons in Argon has been investigated as a function of Argon density and applied electric field using the technique of lifetime measurements. Lifetime spectra were analyzed using the maximum likelihood method of curve fitting. Results obtained at zero electric field yielded a linear dependence on density for the direct annihilation rate of $(5.6 \pm 0.1) \times 10^6 \text{sec}^{-1} \text{ amagat}^{-1}$, with some evidence of non-linearity at densities greater than 10 amagats. The density dependence of the long-lived component of the time spectra indicated a zero density intercept of $(7.2 \pm 0.4) \times 10^6 \text{sec}^{-1}$ in agreement with the theoretical value of the free orthopositronium annihilation rate ($7.2 \times 10^6 \text{sec}^{-1}$). In addition an orthopositronium quenching rate of $(0.29 \pm 0.04) \times 10^6 \text{sec}^{-1} \text{ amagat}^{-1}$ was obtained from the linear dependence of the orthopositronium annihilation rate on density.

The electric field dependence of the direct annihilation rate and orthopositronium formation have been measured and are used to provide an internally consistent picture of the behaviour of positrons in a gas under the influence of an applied electric field. Furthermore, these results have been compared with theoretical results for the direct annihilation rate obtained from one parameter representations of the effective positron-Argon atom interaction. It is shown that, while such potentials are successful in describing the low-energy elastic-scattering of electrons from noble gas atoms, they are inadequate for the case of positrons. However, consideration of the way in which the direct annihilation

rate changes as a function of electric field leads to an upper limit of $15\pi a_0^2$ for the momentum-transfer cross-section for positrons in Argon at thermal energies. Such an estimate is shown to be independent of any assumption concerning the effective positron-Argon atom interaction.

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1. POSITRONS AND THEIR INTERACTION WITH GAS ATOMS.

1.1. Introduction.

Of all the predictions of modern relativistic quantum mechanics, surely one of the most satisfying has been the deduction of the existence of the positron. This achievement is due to Dirac (1928), who proposed a relativistic wave equation for the electron, which, in addition to predicting such dynamical characteristics as the spin of the electron, also predicted the existence of a positively-charged anti-particle (Dirac, 1931). This positive particle, the positron, was discovered experimentally by Anderson (1932), and was found to have a mass equal to that of the electron.

The study of the interaction of slow positrons with gas atoms was first undertaken by Shearer and Deutsch (1949), who studied the slowing down of positrons in various gases. As a direct result of these experiments the existence of a positronium atom postulated by Ruark (1945) was verified. A bound system containing a positron and an electron, positronium is similar to hydrogen in many of its properties. However, it differs radically from hydrogen in that the two particles annihilate each other with the emission of two or more quanta of gamma radiation.

The annihilation of positrons with electrons forms the basis of most experiments dealing with the atomic interactions of positrons. Use is made of the annihilation radiation to study

either the relative positron-electron velocity at annihilation (Heinberg and Page, 1957; Celitans and Green, 1964), or the lifetime of the positrons in a gas, liquid or solid (Falk, 1965).

The latter technique forms the experimental basis of this work.

Under certain conditions to be made clear in the following sections, the lifetime of positrons in a noble gas is simply connected to the cross-sections for momentum transfer, annihilation and positronium formation. It is the task of quantum mechanics to give precise predictions of these cross-sections, in order that comparison with experiment can be made. The theoretical aspect of this thesis is concerned both with the calculation of momentum transfer and annihilation cross-sections for some simple potentials describing the positron-atom interaction, and with the reduction of these cross-sections in order to make comparison with experimental lifetime results. It is necessary to describe the positron-atom interaction in an approximate way, since, although the correct wave equation representing the interaction can be given, it was not possible to make a complete analysis because of the many particle aspect. Thus approximate models must be used and justified by comparison with experiment. The current situation regarding the theory of positron-atom interactions is enlarged upon in Chapter 3.

1.2. The fate of positrons in a gas.

1.2.1. Introductory remarks.

The study of positron-atom interactions has been largely confined to experiments using Na-22 or Cu-64 as positron sources.

The energy-spectrum of positrons from such sources is the continuous Fermi distribution. In the case of Na-22, the maximum positron energy is 542 keV, and the distribution is peaked at 170 keV (Macklin, et al., 1950). The method by which these high energy positrons lose energy and subsequently annihilate in a gas is qualitatively understood.

Figure 1 (Falk, 1965) illustrates the various mechanisms involved in this energy loss. In order to discuss these mechanisms, discussion is confined to the case of Argon at a density of the order of 10 amagats. (1 amagat = 4.4589×10^{-5} moles/cc. American Institute of Physics Handbook.) Argon is the gas used throughout this work and the density is such that the rate of the processes involved can be related to the time resolution, 7.4 nsec, characteristic of the instrumentation used.

1.2.2. Description of the annihilation time spectrum in terms of a positron velocity distribution.

Annihilation life time spectra of positrons are determined in the following way. The emission of a positron from a Na-22 nucleus is followed within 10^{-12} secs by a 1.28 MeV gamma ray necessary to de-excite the daughter nucleus Ne-22 to its ground state. This gamma ray provides the positron "birth" signal. The annihilation of the positron with an electron results in a total of 1.02 MeV of gamma radiation being emitted. The detection of this gamma radiation signifies the "death" of a positron. An annihilation time-spectrum displays the number of such events as

Figure 1. Annihilation mechanisms of positrons in gases.
Ps-positronium.

a function of the lifetime of the positron. Such a time spectrum will in general contain components arising from the annihilation of free positrons (direct annihilation), and of para- and ortho-positronium. For a gas in which there is no positronium formation, the time spectrum consists of direct annihilations only.

In order to discuss the lifetime of a free positron in such a gas, it is convenient to consider the experiment as one in which all the positrons detected were emitted simultaneously. Furthermore the positron density is thought of as so low that no one positron can interact with another during its lifetime, which is the case for the weak ($10\text{ }\mu\text{Ci}$) sources used here. In such a case, the swarm of positrons has some energy distribution, which is modified as a function of time by interaction of the positrons with the gas atoms.

The logarithm of the slope of a time spectrum at any time t is then inversely proportional to the velocity-dependent annihilation rate averaged over the velocity distribution appropriate to the time t . Returning now to the actual conditions under which the experimental data is gathered it can be seen that there is no difference between these two descriptions. The positron velocity distribution $y(v,t)$ consistent with either of these descriptions is then the probability that at a time t , the positron has a velocity between v and $v+dv$. For the case of a swarm of positrons, the positron velocity distribution is equally defined as the fraction of positrons at time t which have a velocity between v and $v+dv$.

The above discussion applies equally to the case where

there is positronium formation, except that positronium annihilations modify the shape of the time spectrum somewhat.

1.2.3. The influence of inelastic collisions on the slowing-down time.

At 10 amagats the positrons from the continuous energy distribution lose energy rapidly in the region where inelastic collisions with the Argon atoms are possible. Within about 0.7 nsec all the positrons have lost all but about 10 keV of their initial energy (Falk, 1965). The calculation of this time relies on experimental and theoretical values of dE/dx over the energy range from 540 keV to 10 keV (Nelms, 1956), and is probably correct to within 20%. The time involved for the remaining energy loss from 10 keV to the last inelastic level in Argon at 11.6 eV is less certain. However, a realistic upper limit to this time can be found by assuming that a minimum of 11.6 eV is lost per inelastic collision. For an average inelastic collision cross-section of $\sim \pi a_0^2$, where a_0 = Bohr radius, this gives an upper limit of $\sim 5 \times 10^{-10}$ seconds for a density of 10 amagats. Thus it is clear that, given an experimental time resolution of about 7 nsec, all annihilations that occur during this initial slowing-down period occur within the resolution of the apparatus. In addition, the positron annihilation during this time is negligible (Gerhart, et al., 1954; Kendall and Deutsch, 1954).

... the effect of elastic collisions

1.2.4. The influence of the elastic collisions on the slowing-down time.

The majority of the positrons, then, reach energies below 11.6 eV without annihilating. Below this energy the positrons can only be slowed down by elastic collisions with the Argon atoms. During this slowing-down period, the positron distribution will be depopulated by three main processes. Firstly, a positron can annihilate with an atomic electron during a direct collision with the atom, the so-called direct annihilation. Secondly, a bound Ar-e^+ complex may be formed which subsequently also annihilates. These processes will be discussed in Section 1.3. Thirdly, a positron may capture an atomic electron to form a positronium atom which may subsequently annihilate with one of two characteristic spin-dependent lifetimes. This possibility is discussed in Section 1.4. Scattering elastically only, the velocity distribution of the positrons will relax to an equilibrium distribution characterised only by the temperature T of the host gas. This equilibrium distribution will be Maxwellian except for a possible small deviation produced by the direct annihilations. These deviations occur if the annihilation rate (probability per unit time for annihilation of a positron) is velocity-dependent.

For an annihilation rate which is velocity-independent the rate of removal of positrons per unit velocity interval from the velocity distribution is also velocity-independent. It follows that for this special case the shape of the positron velocity distribution is unaffected by annihilations and is therefore Maxwellian

at equilibrium. Since the annihilation rate is directly proportional to the product of velocity of the positron (relative to the atom) and the annihilation cross-section at that velocity, it is clear that a velocity-independent annihilation rate results from an annihilation cross-section inversely proportional to the positron velocity.

For the case of an annihilation rate which increases with decreasing velocity, positrons are removed preferentially from the low-energy end of the distribution. At any instant the average positron energy will be higher than in the case of a constant or zero annihilation rate. This can only occur if the centre of gravity of the velocity distribution is shifted up in velocity compared with the Maxwellian distribution.

The time taken to reach thermal equilibrium depends on the shape of the initial velocity distribution below 11.6 eV and also on the momentum-transfer cross-section. Typically, for a momentum-transfer cross-section of the order of πa_0^2 , the relaxation time is greater than about 10 nsec (Tao, et al., 1963), which is larger than the experimental time resolution of 7 nsec.

1.2.5. Summary.

For about 10 amagats of Argon, and an experimental time resolution of about 7 nsec, positron annihilations during the initial slowing-down period to the first excitation level in Argon at 11.6 eV are not expected to have any observable lifetime. The remaining positrons can lose energy only by elastic collisions,

the positron velocity distribution being depopulated by annihilations and positronium formation. The latter can occur only when the positron has sufficient energy to make up the difference between the ionization potentials of Argon and positronium. For Argon this threshold energy is 8.9 eV, below which positronium formation is not possible. The relaxation time of the distribution to equilibrium is expected to be of the order of 10 nsec or greater. Effects due to this slow relaxation time have been previously observed and are discussed in Section 1.3. and in Chapter 2.

1.3. Direct annihilation rate and momentum-transfer cross-sections for positrons in Argon.

1.3.1. Relationship of annihilation rate to positron-electron overlap.

During the collision of a free positron with an atomic system, there is the possibility that the positron will annihilate with an atomic electron without forming positronium. Ferrell (1956) has shown that the positron annihilation rate is proportional to the gas density and to the electron density at the positron averaged over the positron position. The detailed expression is given in Chapter 3.

If the incoming positron is described by a plane wave, the integral involving the positron-electron overlap is equal to the total number, Z , of electrons comprising the atom. Because of the departure of the positron wave-function from a plane wave due to the coulomb interaction, the overlap integral is not in general

equal to Z . The resulting effective number of electrons, Z_{eff} , depends on the particular form of the positron-atom interaction assumed.

An alternate mode of annihilation would result if a bound Ar-e^+ state is possible. Annihilations from such a system would be indistinguishable from the direct annihilations by the experimental techniques employed here. The lifetime of this system will be of the order of the parapositronium lifetime (10^{-10} secs). The observed annihilation rate characteristic of this process is thus the capture rate of positrons into the bound Ar-e^+ system and is thus proportional to pressure.

No theoretical discussion of the annihilation rate so far has included this possibility. The annihilation rates calculated in this thesis are those obtained by considering only the positron-electron overlap during an elastic collision. As yet, there is no way of telling which of the two processes predominates. The calculations presented in this thesis are based on the assumption that the contribution to the observed annihilation rate from this process is negligible compared with the direct rate considered.

1.3.2. Direct annihilation rate in Argon.

Experimental results indicate (Falk, 1965; Tao, Bell, and Green, 1964; Osmon, 1965; Paul, 1964) that the average annihilation rate for Argon (about $5 \times 10^6 \text{sec}^{-1} \text{amagat}^{-1}$) implies a Z_{eff} of about 30, substantially larger than the Dirac Z_{eff} of 18.

This indicates that the positron must be attracted to the atom at some stage in order for the wave-function of the positron at the atom to be substantially larger than that appropriate to a plane wave. The required attraction could be due, at least partially, to the standard long range polarization term found necessary to fit low energy electron scattering from noble gas atoms (Mott and Massey, 1965). The polarization potential behaves asymptotically as α/R^4 , where R is the positron-atom separation and α is the polarizability of the Argon atom. A further contribution to the high Z_{eff} could arise from distortion of the electron cloud around the Argon atom by the incoming positron, this effect tending to increase the electron density at the positron position.

Two calculations directed specifically to the case of Argon have been reported (Massey, et al., 1966; Jones and Orth, 1966). Both use an empirical polarization potential with an asymptotic α/R^4 behaviour, with a simple cutoff in order that the potential remain finite at the origin. The cutoffs are also chosen such that the Hartree-Fock potential appropriate to the ground-state atom dominates in the interior of the atom. A scattering potential is taken to be the sum of the static Hartree-Fock and polarization potentials. This type of scattering potential is the simplest for which an attraction is possible, occurring when the polarization potential dominates the static Hartree-Fock potential, which is repulsive for a positron. In both calculations reported no effects due to electron distortion are considered. Although this type of potential is adequate for describing the Ramsauer effect in Argon

(Holtzmark, 1929; Kivel, 1959; Labahn and Callaway, 1966) it is shown in this thesis to be inadequate for the positron-Argon scattering case. This point is discussed further in Chapter 3.

1.3.3. Velocity dependence of the direct annihilation rate in Argon.

That the direct annihilation rate for positrons in Argon is velocity-dependent in the region between 0 eV and about 10 eV follows from the experimental observation that the direct annihilation rate is not exponential, but is characterised by a "shoulder" in the positron annihilation time spectrum (Falk and Jones, 1964; Falk, 1965; Tao, Bell and Green, 1964; Osmon, 1965; Paul, 1964). A typical time spectrum showing this feature is given in Figure 2. The shoulder is followed by a single exponential which is taken to be the direct annihilation rate for the thermalized positrons (Falk, 1965). The justification for this description of the observations is considered in Chapter 2, Section 2.1.

1.3.4. Electric field dependence of the direct annihilation rate in Argon.

The use of an applied electric field to study the velocity dependence of the positron annihilation rate was introduced by Falk, Orth and Jones (1965). This method relies on the fact that the velocity distribution of the positrons at equilibrium can be influenced by the application of a uniform dc electric field. Since the positrons may gain energy from the field, the average velocity of the distribution is thus increased. The observed annihilation rate, which is the velocity-dependent annihilation

rate averaged over all positron velocities in the distribution, is then electric field dependent. The detailed shape of the equilibrium velocity distribution is governed to first order by the magnitude of the applied electric field, and the velocity dependence of the elastic scattering momentum-transfer cross-section. In order to test a specific model of the interaction with the experimental results, it is therefore necessary to compute both the direct annihilation rate and the momentum-transfer cross-section for the model assumed.

The first measurements of the electric field dependence of the direct annihilation in Argon (Falk, 1965; Falk, Orth and Jones, 1965) showed that the annihilation rate decreased with increasing electric field, reaching a value of about half that at zero electric field when the field reached a value of about $80 \text{ V cm}^{-1} \text{ amagat}^{-1}$.

1.3.5. Direct annihilation rate of positrons in Helium.

In order to test the general features of this picture of the interaction of positrons with atoms of the noble gases, it is intended to extend such measurements to the other readily obtained noble gases (eg. He, Ne, Kr). Indeed preliminary measurements being performed at present indicate a significant shoulder effect in the annihilation time spectrum for Helium in contradiction to the earlier results of Falk, Orth and Jones (1965), and Osmon (1965). In addition, the shape of the time spectrum is found to be dependent on the presence of an electric field, much

as is observed for the case of Argon.

In fact, a complete experimental investigation of the annihilation rate in Helium is of particular significance since there appears to be considerable interest in the theoretical aspects of this problem at present (Drachman, 1966; Kestner, et al., 1965; Massey, et al., 1966).

1.4. Positronium formation and annihilation in Argon.

1.4.1. Structure of positronium.

As mentioned in Section 1.1., positronium is the bound state of a positron and electron. In the gross structure of its energy levels it differs from the hydrogen atom by a factor of two reflecting a smaller reduced mass. Thus the ionization potential of positronium is 6.78 eV instead of 13.6 eV. The transition probabilities are halved as the average positron-electron distance is twice that of the proton-electron distance. The fine structure differs considerably from that of hydrogen due to the large difference in mass between the proton and positron. An additional fine structure term is introduced by the particle-antiparticle nature of the electron and positron. Only the gross structure of the energy levels is important in this work. Details of the overall structure of positronium have been summarized by Series (1957).

1.4.2. Annihilation of positronium.

Positronium in the ground state has two spin states. Roughly speaking, orthopositronium contains the positron and electron with spins parallel, whereas parapositronium has the spins

anti-parallel. The particle-antiparticle nature of the positron-electron system causes it to be unstable against annihilation into photons. Because of the selection rules governing the decay of a spin 1 particle into photons, orthopositronium can only decay into an odd number of photons. By the same token, parapositronium can only decay into an even number of photons. The general situation regarding the number of photons for annihilation from the ground state and excited states of positronium has been summarized by Kaempffer (1965).

The lifetime of positronium depends on the total angular momentum of the state. Orthopositronium in its ground state has a calculated mean lifetime of 1.4×10^{-7} seconds. Parapositronium in the ground state decays far more rapidly, its mean lifetime being calculated to be 1.25×10^{-10} seconds (Ore and Powell, 1949). With reference to the experimental time resolution of 7×10^{-9} secs quoted in Section 1.2.5. it is clear that the lifetime of orthopositronium will be resolved, whereas that of parapositronium will not be visible. The lifetime of excited states of positronium has been calculated for the S and P states by Alekseev (1959), and are all longer than for the ground states with the same spin alignments. In particular the 2S level should have a mean lifetime of about 1×10^{-9} sec. Furthermore, it appears that this is the only excited level in positronium from which it is at all feasible to detect annihilation, since this state is metastable against optical transitions to the ground state. For all other states the optical transition rates exceed the annihilation rates by a factor of at

least 1000.

1.4.3. Positronium formation.

Formation of positronium in its ground state in a gas can occur if the positron energy is greater than or equal to the difference between the ionization potentials of the gas atom and the positronium atom. For Argon this threshold energy (E_{thr}) is 8.9 eV. The threshold energy for formation into the first excited state is 14.1 eV.

Because of the different spins of ground state ortho- and parapositronium, the ratio of the amounts formed in each of these states is expected to be the normal statistical ratio based on the relative multiplicities of the states. Thus three ortho-positronium atoms should be formed for each parapositronium atom. The amount of $n=2$ positronium formed relative to ground state positronium will be considerably less due to the expected smaller cross-section for formation (Massey and Mohr, 1954) and the competition from the inelastic collisions with the Argon atom. Any positronium atom formed in an excited state (except the 2S) should make the optical transition to the ground state before annihilation. At 10 amagats of Argon, the 2S state is more likely to suffer collisional de-excitation to the 2P state than to annihilate. Furthermore, at these densities, the collisional de-excitation of any excited state into the ground state is probably considerably more rapid than the optical transition rate of about 10^8 sec^{-1} (Wallace, 1955). This might explain to some extent the

unsuccessful attempts to detect the Lyman alpha spectral line of positronium (Brock and Streib, 1958; Bennett, et al., 1961; Duff and Heymann, 1963).

1.4.4. Quenching of positronium lifetimes.

The collisions of a positronium atom with a gas atom can also result in the annihilation of the positron with one of the atomic electrons. This "pick-off" quenching of the lifetime of the positronium atom is the most important type of quenching encountered in collisions with noble gas atoms. This extra channel for annihilation of a positronium atom results in a pressure-dependent annihilation rate. The quenching rate has been measured for orthopositronium in Argon and other noble gases by Heymann, et al., (1961) to be about $0.25 \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$ for Argon. At 10 amagats, the mean lifetime of orthopositronium is thus expected to be reduced to about 1.0×10^{-7} secs. A quenching rate of this magnitude will have a negligible effect on parapositronium lifetime.

1.4.5. Theoretical situation regarding formation, quenching and elastic scattering cross-sections.

1.4.5.1. Positronium formation.

Very little work has been done on the positronium formation cross-section in general. Cheshire (1964) has considered positronium formation by fast positrons in atomic hydrogen, using the Born and impulse approximations. The cross-sections so obtained are of the order of πa_0^2 and decrease with increasing energy, in

agreement to within an order of magnitude with those of Massey and Mohr (1954). Similar calculations using the Born approximation for the case of Helium by Massey and Moussa (1961) indicate that the cross-section is of the order of $0.1\pi a_0^2$ or less near threshold. The cross-section rises to a maximum of $0.4\pi a_0^2$ at 27 eV. No calculations have been published for the case of many-electron atoms.

1.4.5.2. Positronium quenching and elastic-scattering cross-sections.

The general problem of low energy positronium scattering from atomic Hydrogen has been investigated by Massey and Mohr (1954) using the Born approximation. In this case, quenching of ortho-positronium is achieved by direct conversion of ortho- into para-positronium made possible by the exchange of the single atomic electron. The cross-section for this type of quenching was found to be highly energy dependent. These calculations have been repeated in more detail by Fraser (1961) who has considered the elastic scattering of orthopositronium from Helium atoms (Fraser, 1962). Neglecting polarization and excitation, but including the effects of electron exchange, he has found an elastic scattering cross-section for positronium with Helium of $17.7\pi a_0^2$ at zero kinetic energy. The appropriate pick-off quenching cross-section has yet to be reported. Again no calculations exist for other many-electron atoms.

1.5. Summary of work contained in the thesis.

1.5.1. Theoretical aspects.

That a velocity dependent annihilation rate is sufficient to give rise to the observed shoulder in the time spectra has been shown by Falk (1965). One purpose of this thesis is to present calculations of such velocity dependent annihilation rates, based on a simple model of the positron-atom interaction. Elastic-scattering momentum-transfer cross-sections are also derived on the basis of these models. The appropriate velocity averaged annihilation rates at equilibrium as a function of applied electric field have also been derived in order to compare with experiment. The calculation of the velocity dependent annihilation rates, momentum transfer cross-sections, and appropriate velocity averaged annihilation rates as a function of electric field constitute an original contribution in that there are no previously published reports.

1.5.2. Experimental techniques.

Limitations in the instrumentation used by Falk prevented the measurement of the long-lived orthopositronium component at the same time as the shorter-lived direct component in annihilation time-spectra. Consequently, there was some doubt as to the accuracy of the lifetime of the long-lived component. These inaccuracies were expected to have a noticeable effect on the estimation of the short-lived (direct) component. These problems were further compounded by errors in the maximum likelihood method used in fitting

the experimental data.

The experimental results contained in this thesis have been obtained with considerably modified instrumentation, in that the time scale has been extended in order that the orthopositronium component may be measured simultaneously with the direct component. Furthermore, the random background has been reduced by a factor of ten, yielding increased statistical accuracy.

The errors inherent in the original method of curve fitting have been successfully removed and, in addition, the size and applicability of the standard deviations of the resulting parameters are discussed. The original statistical analysis of Falk involved modification of the raw data to take into account instrumental effects. In the present case, these effects are taken into account by making appropriate modifications to the function to be fitted to the raw data.

1.5.3. Positronium formation.

There is some doubt as to the internal consistency of the discussion in Falk's thesis which relates to the electric field dependence of the positronium formation rate and to the effect of this positronium formation rate on the direct annihilation rate. This arises from the fact that the measurements of positronium formation as a function of electric field due to Marder, et al., (1956) were used for that discussion. The present work contains such measurements for the same gas samples as were used in the lifetime experiments presented here. The results differ consider-

ably from those of Marder, et al. There is, therefore, reasonable doubt that the Marder, et al. values were relevant to the Falk experiments.

1.5.4. Orthopositronium annihilation rates.

The pressure dependent quenching rate of orthopositronium in Argon is discussed in this thesis in some detail. These measurements are considerably more accurate than any previously reported, a result of increased statistical accuracy and improved curve-fitting techniques.

2. EXPERIMENTAL INVESTIGATION OF POSITRON LIFETIMES IN ARGON.

2.1. Introduction.

Recent investigations of the lifetime of positrons in Argon have shown that the free positron annihilation rate cannot be described by a single exponential (Tao, Bell and Green, 1964; Paul, 1964; Osmon, 1965; Falk and Jones, 1964). Time spectra of the annihilation gamma rays show clear evidence of a shoulder followed by a single exponential whose lifetime is presumed to arise from the direct annihilation at thermal velocities. It has been shown that the shoulder is removed, and the lifetime of the exponential changed, on the application of a moderate static electric field (Falk, Orth and Jones, 1965). Typically, a field of about $80 \text{ V cm}^{-1} \text{ amagat}^{-1}$ is sufficient to decrease the direct lifetime by a factor of two. In view of the importance of these results as the only available experimental test of the validity of models describing the positron-Argon atom interaction, a further series of these measurements has been made with improved instrumentation, and to a greater degree of statistical accuracy. These measurements are used to test the validity of several empirical potentials describing the effective positron-Argon interaction (Chapter 3).

Positrons emitted by a radioactive source into a gas lose most of their energy by inelastic collisions with the gas atoms. Once positrons in Argon have their kinetic energy decreased to

11.6 eV, the lowest excitation energy of Argon, they can only lose energy by elastic collisions. In addition, positronium can be formed until the threshold energy for positronium formation, 8.9 eV, is reached. Those positrons which terminate their rapid slowing-down at kinetic energies below 11.6 eV, can be described by a particular initial time-dependent velocity-distribution function (Falk, Orth and Jones, 1965; Falk, 1965).

Once the positrons thermalize, the shape of the distribution function becomes time independent (essentially Maxwellian) and the depopulation by annihilation is characterised by a single exponential. A single exponential will also occur, regardless of the shape of the positron distribution function, if the annihilation rate is velocity independent (Chapter 3, Section 3.4.). The observed shoulder in the time spectrum from Argon, followed by a single exponential, all superimposed on the orthopositronium decay, is therefore a clear indication that here the direct annihilation rate of positrons is velocity dependent.

That the direct annihilation rate changes on the application of electric field further confirms this point of view (Falk, Orth and Jones, 1965). An applied electric field increases the average energy of the positrons, the equilibrium distribution being to first order a function of the positron-Argon elastic-scattering momentum-transfer cross-section, and electric field. The observed annihilation rate is thus changed since the velocity-dependent annihilation rate is averaged over a different equilibrium velocity distribution. The experimental results contained herein

are measurements of the annihilation rates for this equilibrium distribution.

Measurements of the ratio of three photon to two photon annihilation in Argon have also been made by monitoring the "valley" and "peak" count rates of the 0.51 MeV gamma-ray spectra (Marder, et al., 1956). This ratio is directly related to the fraction of positrons forming positronium in Argon.

2.2. Experimental method.

2.2.1. Lifetime measurements.

A 10 μ Ci Na-22 source deposited on 30 μ inch aluminum foil served as the positron source. The instrumentation for the recording of the time spectra is primarily that used previously by Falk (1965)(see also Falk and Jones, 1964; Falk, Orth and Jones, 1965). To improve reliability, the limiters and slow coincidence sections were replaced by transistorized equivalents. Details of these modifications are given in the Appendix. In order to improve the statistical accuracy of the measurements, the random coincidence background was reduced (relative to the measurements of Falk) by a factor of ten by reducing the positron source strength by the same factor. In order, however, to maintain the same overall true-coincidence count rate, detectors of significantly greater efficiency and solid angle were required. To this end the gamma ray detectors used by Falk, consisting of 2 in. x 2 $\frac{1}{2}$ in. diameter NaI(Tl) crystals mounted on R.C.A. 6810 photomultipliers were replaced by 4 in. x 3 in. diameter NaI(Tl) crystals mounted on R.C.A. 7046

photomultipliers.

In addition, use of a 256 channel analyzer (ND101) made it possible to extend the time range of the measurements to include the whole orthopositronium decay, thus facilitating the analysis of data. The gas chamber used is the same as that used previously in this laboratory (Falk, 1965; Falk, Orth and Jones, 1965). The gas was purified as before by continuous recirculation over a hot CaMg eutectic mixture (Colli and Fachini, 1952). Analysis of the gas by The Matheson Co., Inc. N.J. indicates that the main impurity present was N_2 at about one part in 10^4 . The results of the Matheson analysis on both the bottle gas and chamber gas (after purification) are shown in Table I.

TABLE I.
Gas Purity Analysis.

<u>Impurity</u> <u>gas</u>	<u>Bottle</u> <u>gas</u>	<u>Chamber</u> <u>gas</u>
N_2	190 ppm	119 ppm
O_2	5 ppm	3 ppm
CO_2	< 4 ppm	< 4 ppm
H_2	23 ppm	32 ppm
He	153 ppm	< 100 ppm

The effectiveness of the purifier in at least maintaining the purity level of the Argon is quite apparent.

A block diagram of the electronic instrumentation used is shown in Figure 3. A typical run extended over a period of two

Figure 3. Block diagram of electronics used in lifetime measurements.

days in which time some 2×10^7 1.28 MeV nuclear gamma rays were counted in the photo peak. The number of these gamma rays counted was used as normalization from run to run in order to avoid difficulties caused by the source decay and the single channel analyzer drifts. Each single channel analyzer setting was obtained by using the two independent outputs of the appropriate linear amplifier. The single channel analyzer associated with the annihilation gamma rays was usually set at the photopeak of the 0.51 MeV gamma ray. However, many runs were taken with the same channel width but with the baseline set so as to include only the region between the photopeak and the Compton edge, the so-called valley region. At this setting the ratio of three photon to two photon events was enhanced relative to the usual setting, since the three photon count rate in the valley region is comparable to that in the 0.51 MeV peak region (Ore and Powell, 1949), while the two photon count rate is sharply diminished. This setting allowed much greater accuracy for the orthopositronium lifetime. The energy spectrum of the 0.51 MeV gamma radiation together with the two settings of the single channel analyzers are shown in Figure 4. The overall time resolution for both S.C.A. settings was 2.7 channels, or 7.4 nsecs (full width at half maximum), measured using a Na-22 source in aluminum (see Appendix).

A representative time spectrum is shown in Figure 5. The peak is due to annihilation of positrons in the walls of the chamber and in the source holder, of parapositronium formed during the initial slowing-down period, and of positrons annihilating in

Figure 4. Energy spectrum of 0.51 MeV gamma rays showing S.C.A. settings. V - valley position; P - peak position. A_{1,2} - direct-enhanced setting; C_{1,2} - ortho-enhanced setting; A₂B - valley-to-peak ratio setting.

the gas within about 7 nsecs of emission from the source. The end of the shoulder is marked by a relatively short-lived exponential component, the direct component. The orthopositronium annihilations give rise to the long-lived exponential. Omitted from the figure is the flat random background region which occupies that portion of the kicksorter up to the prompt peak. The spectrum in this region was obtained by recording events where a 0.51 MeV gamma-ray is detected before a 1.28 MeV gamma ray. The counts per channel in this part of the time spectrum, then, correspond to the random background counts per channel. The spectrum in Figure 5 is shown with this random coincidence background subtracted.

2.2.2. Valley-to-peak ratio measurements.

The energy spectrum of the annihilation gamma rays as a function of the electric field was obtained using one 4 in. x 3 in. NaI crystal assembly and the 256 channel kicksorter. A single channel analyzer was used to gate the kicksorter, and was set so that only pulses between the Compton edge for 0.51 MeV gamma rays and the high energy side of the 0.51 MeV photopeak would be analyzed by the kicksorter (Figure 4). Each run lasted approximately 45 minutes in which some 2×10^4 and 8×10^4 counts per channel were obtained in the valley and peak positions respectively.

Figure 5. Representative time spectrum of positron annihilation in Argon. P is in amagats, E/P is in $V \text{ cm}^{-1} \text{ amagat}^{-1}$; I - prompt peak; II - shoulder region; III - direct component; IV - orthopositronium component.

2.3. The exponential portions of the time spectra.

The appearance of a single exponential superimposed on the orthopositronium component in the time spectra is evidence that the velocity distribution of the positrons has become "static" (see Chapter 1, Section 1.3. and Chapter 3, Section 3.4.). That is, the time dependence of the velocity distribution is described by a single exponential. The time spectrum under these conditions is composed of the following components:

2.3.1. The direct or free annihilations.

The population of free positrons at any time t is given by

$$\frac{dN(t)}{dt} = -[\lambda_f + \lambda_d] N(t) \quad (1)$$

where λ_f is the velocity-averaged positronium formation rate, and λ_d is the velocity-averaged direct annihilation rate. Assuming that the direct annihilations are all by two photon decay whose efficiency for detection is ϵ_2 , the observed direct annihilation rate is:

$$R_d(t) = \epsilon_2 \lambda_d N(0) e^{-(\lambda_f + \lambda_d)t} \text{ sec}^{-1} \quad (2)$$

2.3.2. Orthopositronium annihilations.

The orthopositronium population is given by

$$\frac{dN_o(t)}{dt} = -\bar{\lambda}_o N_o(t) + \frac{3}{4} \lambda_f N(t) \quad (3)$$

where $\bar{\lambda}_o$ is the sum of the free orthopositronium decay rate λ_o and the orthopositronium quenching rate λ_q . It is assumed that the fraction of positronium atoms which are formed as orthopositronium is the statistical ratio 3:4. The population at any time

t is then

$$N_O(t) = [N_O(0) + \frac{3}{4} \lambda_f \frac{N(0)}{\lambda_d + \lambda_f - \bar{\lambda}_O}] e^{-\bar{\lambda}_O t} - \frac{3}{4} \frac{\lambda_f N(0)}{\lambda_f + \lambda_d - \bar{\lambda}_O} e^{-(\lambda_d + \lambda_f)t} \quad (4)$$

where $N_O(0)$ is the number of orthopositronium atoms present at $t=0$. The observed orthopositronium decay rate, corresponding to $\bar{\lambda}_O N_O(t)$ is:

$$R_O(t) = \lambda_O \epsilon_3 N_O(t) \quad (5)$$

where ϵ_3 is the three photon detection efficiency. It is apparent that this contains two terms, one, $R_O^I(t)$, representing the decay of the orthopositronium atoms present at time $t=0$, the other, $R_O^F(t)$, representing annihilation from the "delayed" positronium formation. Thus

$$R_O^I(t) = \lambda_O \epsilon_3 N_O(0) e^{-\bar{\lambda}_O t} \quad (6)$$

$$R_O^F(t) = \lambda_O \epsilon_3 \frac{3}{4} \cdot \frac{\lambda_f}{\lambda_d + \lambda_f - \bar{\lambda}_O} N(0) [e^{-\bar{\lambda}_O t} - e^{-(\lambda_d + \lambda_f)t}] \quad (7)$$

It should be noted that if the positronium formation rate λ_f becomes appreciable under the influence of the electric field, and if $\lambda_d + \lambda_f \gg \bar{\lambda}_O$, the observed orthopositronium spectrum will exhibit a characteristic exponential growth, followed by a slow exponential decay (of lifetime $1/\bar{\lambda}_O$).

2.3.3. Parapositronium annihilations.

Since parapositronium has a mean life ($\sim 10^{-10}$ sec) considerably shorter than the experimental time resolution available (7.4 nsec) its contribution to the spectrum is governed entirely by the parapositronium population present at any time t. Thus the decay rate is given by

$$R_P(t) = \epsilon_2 [\lambda_q N_O(t) + \frac{\lambda_f}{4} N(t)] \quad (8)$$

The effect of orthopositronium quenching is taken into account here noting that this is equivalent to parapositronium formation at a rate given by the quenching rate λ_q . The separate formation of parapositronium from free positrons is also taken into account. This component has the same detection efficiency as the direct annihilations since the annihilation is by two photons only (Yang, 1950).

2.3.4. The observed spectrum in the exponential region.

The total observed spectrum is the sum of these three components, viz.,

$$R(t) = I_1 e^{-(\lambda_d + \lambda_f)t} + I_2 e^{-\bar{\lambda}_o t} \quad (9)$$

where

$$I_1 = \left[\epsilon_2 \frac{\lambda_f}{4} + \epsilon_2 \lambda_d - \epsilon_3 \lambda_o \frac{3}{4} \frac{\lambda_f (1 + \epsilon_2 \lambda_q / \epsilon_3 \lambda_o)}{\lambda_d + \lambda_f - \bar{\lambda}_o} \right] N(0) \quad (10)$$

and

$$I_2 = \left[\epsilon_3 \lambda_o + \epsilon_2 \lambda_q \right] \left[N_o(0) + \frac{3}{4} \lambda_f \frac{N(0)}{\lambda_d + \lambda_f - \bar{\lambda}_o} \right] \quad (11)$$

It is clear that although there may be a growth in the orthopositronium component the observed spectrum is always a simple sum of the two exponentials unless I_1 happens to be a negative. Such a situation has not yet been observed.

2.4. The valley-to-peak ratio.

The energy spectrum of three-photon annihilation is continuous, with a maximum at 0.51 MeV (Ore and Powell, 1949). In the annihilation spectrum of positrons in Argon, therefore, the ratio of counts in the valley region (between the 0.51 MeV peak and Compton edge) to the counts in the 0.51 MeV peak increases as

a function of positronium formation.

Consider a 0.51 MeV gamma-ray spectrum obtained with no three-photon events. The valley-to-peak ratio, R_0 , will be less than that in a spectrum where three-photon events were also counted. If, in a spectrum of the latter type, the peak and valley count rates are C_p , C_v , respectively, then C_3 , where

$$C_3 = C_v - (C_p - R_3 C_3) R_0 \quad (12)$$

will be the count rate due to the three-photon events only, in the valley region. The coefficient of R_0 is the count rate C_p in the 0.51 MeV photopeak with the contribution due to three-photon events in this region, $R_3 C_3$, subtracted. In this case R_3 represents the relative probability of counting a gamma ray from a three-photon annihilation in the 0.51 MeV peak to counting such a gamma ray at the valley position. From a knowledge of the energy spectrum of three-photon annihilations (Ore and Powell, 1949), and the relative efficiencies of the crystal for counting gamma rays whose energies correspond to the peak and valley regions respectively, R_3 can be determined. Equation (12) can be more usefully rewritten in the form:

$$C_3 = (C_v - C_p R_0) / (1 - R_3 R_0). \quad (13)$$

In the case of positrons annihilating in Argon under the influence of an applied electric field, where it is possible to change only the relative number of three-photon orthopositronium and two-photon decays, C_3 will be directly proportional to the fraction of positrons forming orthopositronium. Thus

$$f = k C_3 \quad (14)$$

where f is the fraction of positrons forming positronium and k is

a proportionality constant which is independent of electric field, if the fraction of orthopositronium atoms which are quenched is independent of electric field. In practice the peak count rate has to be reduced by an additional factor of $1-W$, where W represents the fraction of positrons which annihilate in the walls of the gas chamber. Thus finally

$$f = k [C_v - C_p (1 - W) R_0] / [1 - R_3 R_0] \quad (15)$$

The proportionality constant k is determined in Section 2.6.3. from a knowledge of the fraction of positrons forming positronium at zero field (Falk and Jones, 1964), the fraction $(1-W)$ of positrons stopped in the gas (Falk, 1965), and from the peak (C_p) and valley (C_v) count rates at zero applied electric field. The expression (15) is similar to that deduced by Marder, et al., (1956), but is simpler in that no magnetic field quenching is present in this case.

2.5. Analysis of results.

2.5.1. Analysis of time spectra.

2.5.1.1. Maximum likelihood theory.

In order to fit the experimental data to a sum of two exponentials (as expected according to the discussion of Section 2.3.), a computer programme was devised utilizing maximum-likelihood theory (Orear, 1958). For finite channel widths, w_k , and a constant random background B per unit channel, the theoretical spectrum shape is

$$y_k = \int_{w_0}^{w_0+w_k} [I_1 \exp(-t/\tau_1) + I_2 \exp(-t/\tau_2)] dt + w_k B \quad (16)$$

where w_0 is the sum of the channel widths up to the start of channel k. Since the counts in individual channels are Poisson distributed, the probability of observing N_k counts in channel k is

$$P_k = (y_k)^{N_k} \exp(-y_k) / N_k! \quad (17)$$

while for the whole exponential region the joint probability is the likelihood function

$$L = \prod_k P_k \quad (18)$$

The aim is to determine the four parameters, I_1 , I_2 , τ_1 , τ_2 , for which this probability L is a maximum. It is more convenient to deal with the logarithmic probability

$$W = \sum_k \ln P_k = \sum_k N_k \ln y_k - y_k - \ln(N_k!) \quad (18a)$$

The programme which maximizes this probability follows the same lines as that of Falk (1965). The values of the parameters for which W is a maximum is given by the set of four equations

$$\frac{\partial W}{\partial I_1} = 0; \quad \frac{\partial W}{\partial I_2} = 0; \quad \frac{\partial W}{\partial \tau_1} = 0; \quad \frac{\partial W}{\partial \tau_2} = 0. \quad (19)$$

2.5.1.2. Iterative solution of the maximum likelihood problem.

Since the equations (19) are extremely nonlinear, the method followed was an iterative procedure involving some initial estimate of the parameters and a first order Taylor's series expansion of each of the partial derivatives (19) about these initial estimates. If the four parameters are denoted a_1 , a_2 , a_3 , a_4 , respectively, then the four simultaneous equations above reduce to a set of four simultaneous linear equations represented

by

$$CA = V. \quad (20)$$

The 4 x 4 matrix C contains the elements

$$C_{ij} = \sum_k \left[\left(\frac{N_k}{y_k} - 1 \right) \frac{\partial^2 y_k}{\partial a_i \partial a_j} - \frac{N_k}{y_k^2} \cdot \frac{\partial y_k}{\partial a_i} \cdot \frac{\partial y_k}{\partial a_j} \right] \quad (21)$$

The vectors A and V are respectively

$$A_i = \Delta a_i \quad (22)$$

and

$$V_i = \sum_k \left(\frac{N_k}{y_k} - 1 \right) \frac{\partial y_k}{\partial a_i} \quad (23)$$

Solution of the four simultaneous equations yields the increments Δa_i which are to be added to the initial estimates a_i . The new a_i so obtained are then inserted in the set of equations (20) and the process repeated. Convergence for the case of two exponentials generally requires less than six iterations, once the initial estimates are sufficiently good that none of the parameters become negative. Should the latter occur it was found convenient to fix one of the parameters, usually the long lifetime. In this way, the problem is reduced to a problem involving solution of three equations in three unknowns. Once convergence under this three parameter variation was achieved, the four parameter iterations, using the best values obtained from the three parameter solution as the initial estimate, usually converged.

2.5.1.3. Initial estimates of the four parameters.

The initial estimates were made using a very fast iterative procedure based on the least squares method. The spectrum is divided into three regions. The first region is the range over

which the short lifetime predominates and the second the range over which the long lifetime is most important. Between these is the third region, where contributions from each of the components are comparable. The iterative procedure involves making a straight line fit to the logarithm of the channel counts in the second region. This fit is extrapolated into the first region, and its contribution to the first region subtracted out. The logarithm of the resulting curve in the first region is then fitted to a straight line. This line is extrapolated into the second region, and its contribution subtracted from the second region. The logarithm of the remainder in the second region is once more fitted to a straight line and the whole procedure repeated.

2.5.1.4. Estimation of channel widths w_k and random background B.

The relative channel widths, w_k , have been measured using the random time generator (see Section 2.6.6.2.). The results of the measurement are shown in the Appendix. Estimation of the random background counts, B per unit channel, follows the procedure used by Falk (1965) and involves the measurement of the random coincidence rate directly from the lifetime spectra (Section 2.2.1.).

2.5.1.5. Estimation of variances.

Once the most probable set of values: a_1^* , a_2^* , a_3^* , a_4^* , have been obtained, an estimate of the statistical uncertainty in each of these values is desired. It has been shown (Orear, 1958) that a matrix of the type

$$H_{ij} = - \frac{\partial^2 W}{\partial a_i \partial a_j} \quad (24)$$

when inverted, yields the so-called error matrix

$$(H^{-1})_{ij} = (a_i - a_i^*) (a_j - a_j^*) \quad (25)$$

where a_k^* is the most probable value of the parameter a_k .

If it is assumed that the likelihood function L is Gaussian with respect to each of the parameters a_i , and that the parameters are independent of each other, then

$$L \propto \prod_i \exp \left[- \frac{1}{2} \left(\frac{a_i^* - a_i}{\sigma_i} \right)^2 \right] \quad (26)$$

where σ_i^2 is the variance of the Gaussian. It follows that

$$W = \sum_i - \frac{1}{2} \left(\frac{a_i^* - a_i}{\sigma_i} \right)^2 + \text{constant} \quad (27)$$

and

$$\frac{\partial^2 W}{\partial a_i^2} = - 1/\sigma_i^2$$

whence

$$\sigma_i^2 = - \left(\frac{\partial^2 W}{\partial a_i^2} \right)^{-1} \quad (28)$$

Thus the diagonal elements of H are

$$H_{ii} = - \frac{\partial^2 W}{\partial a_i^2} \quad (29)$$

In this case, since H_{ii} is a diagonal matrix,

$$(H^{-1})_{ii} = (H_{ii})^{-1}$$

so

$$\sigma_i^2 = (H_{ii})^{-1}$$

yield the variance of the parameters if L is Gaussian in shape.

If H is not diagonal, inversion of the entire matrix H

allows the correlation between different parameters to be taken into account in the estimation of the variances (Orear, 1958). Should any of the off-diagonal elements be negligible compared with the main diagonal elements, then the relevant parameters have an appropriately small degree of correlation.

In order to check the assumption that the likelihood function describing typical experimental results is near to Gaussian in shape, the following analysis was performed. Each of the parameters in turn was set at one and then two standard deviations on either side of the best value obtained using the maximum likelihood programme. The remaining parameters were then varied to maximize the likelihood once more. The resulting likelihood relative to the best maximum likelihood was then plotted as a function of the parameter concerned. Figure 6 indicates that the shape of the likelihood function is indeed approximately Gaussian for each of the four parameters.

2.5.1.6. Goodness of fit.

In order to discuss the goodness - or otherwise - of the fit, the usual chi-square test was made on each spectrum. However, since the chi-square test is defined in terms of the normalized probability of getting a worse fit for quantities distributed according to Gaussian statistics (Orear, 1958; Mathews and Walker, 1965) it is not sufficient to make a straight channel-by-channel computation in the present case. This arises because of the low number of counts, usually thirty to forty, in the tail of the time

Figure 6. Dependence of the likelihood function on I_1 , I_2 , τ_1 , τ_2 . The values of these parameters which give the maximum likelihood L^* correspond to the point a^* ; $\sigma_{a^*}^2$ is the variance of a^* as calculated from Equation 25. Values of the Gaussian curve $\exp[-(\frac{a-a^*}{\sigma_{a^*}})^2]$ are denoted Gaussian in the legend.

spectrum (Figure 5). Although the variance of these Poisson-distributed counts N is N , the best fit is not obtained by minimizing the mean square deviation about the mean since the Poisson distribution is skew about the mean for counts below about 100. In fact, counts lower than the mean are more probable. However, if the counts are summed over a certain number of channels, such that the total number of counts is above about 100, then the chi-square test becomes applicable. Table II shows the results of chi-square tests on all the spectra presented. In order to reinforce the point concerning the non-applicability of the chi-square test for non-Gaussian distributed counts, Table II further contains results for the channel-by-channel chi-square test.

2.5.2. Analysis of experimentally-determined annihilation rates.

In general the annihilation rates of positrons in a gas are dependent on the gas density. In order to fit the observed annihilation rates, direct or orthopositronium decays, to a particular polynomial dependence on density, the least squares method was employed. The justification for using the least squares method lies solely in the fact that the likelihood functions for the typical time spectra reported here have been demonstrated to be nearly Gaussian in shape (Section 2.5.1.5.). That they are so shows that the probability distribution governing the parameters I_1 , I_2 , τ_1 , τ_2 , is nearly Gaussian. Thus, in order to analyze the lifetimes, it is consistent to use the maximum likelihood method assuming Gaussian statistics, which gives rise to the least squares method of curve fitting (Orear, 1958). Furthermore, the standard

TABLE II.

RESULTS OF CHI-SQUARE TEST ON LIFETIME SPECTRA.

Q is to be interpreted as the normalized (to 1) probability of getting a worse fit should the experiment be repeated.

Q_B is the result of the channel-by-channel computation, while Q_A results from the alternative integral method outlined in Section 2.5.1.6.

<u>Q_A</u>	<u>Q_B</u>
0.30	0.37
0.50	0.15
0.32	0.005
0.91	0.59
0.94	0.14
0.70	0.004
0.46	0.12
0.91	0.0003
0.90	0.12
0.64	0.34
0.46	0.002
0.92	0.45

TABLE II (continued)

RESULTS OF THE CHI-SQUARE TEST ON LIFETIME SPECTRA.

<u>Q_A</u>	<u>Q_B</u>
0.75	0.39
0.22	0.003
0.77	0.06
0.58	0.06
0.24	0.11
0.79	0.000007
0.76	0.006
0.42	0.004
0.63	0.18
0.01	$<10^{-8}$
0.93	0.03
0.25	0.02
0.56	0.45
0.97	0.73
0.34	0.004

chi-square test is relevant in this case (Section 2.5.1.6.).

The polynomial parameters were obtained using a computer solution for the set of analytic equations pertinent to the least squares problem for a function linear in these parameters (Orear, 1958; Rose, 1953). The function which is linear in the parameters a_i (to be determined) is

$$y(x) = \sum_{i=1}^M a_i f_i(x) \quad (30)$$

where the $f_i(x)$ are any functions of x only. In a situation where p experimental values $N(x_j) \pm \sigma_j$ have been obtained as a function of the p data points x_j , the least squares solutions for the a_i are

$$a_i^* = \sum_{k,j=1}^M \sum_{j=1}^p \frac{N(x_j) f_i(x_j)}{\sigma_j^2} (H^{-1})_{ki} \quad (31)$$

where

$$H_{ij} = \sum_{k=1}^p \frac{f_i(x_k) f_j(x_k)}{\sigma_k^2} \quad (32)$$

The error matrix is given as usual by

$$(H^{-1})_{ij} = (a_i - a_i^*) (a_j - a_j^*) \quad (33)$$

The chi-square test was also performed on each fit.

2.6. Experimental results.

2.6.1. Criteria for presentation of data.

The data presented here represent the results of runs made with four different Argon gas samples. All the spectra were fitted to two exponentials by the maximum-likelihood technique (Section 2.5.1.). The results presented here satisfied the following conditions:

(a) Convergence was obtained varying all four parameters simultaneously in the maximum-likelihood programme.

(b) No high voltage breakdowns occurred during the actual run in question. It was found that the occurrence of such breakdowns could lead to significant deviations in the results. These deviations were ascribed to the effect of contaminating gases liberated by the breakdown prior to the absorption of such contaminants by the purifier.

(c) The results of all the chi-square tests indicated that the probability of getting a worse fit was greater than 0.1 (10%). Out of a total of 33 runs, 6 were rejected on the basis of these criteria. None was rejected on the basis of the chi-square test alone, since a poor result here signified that the assumption of two exponentials was poor for the case considered. This fault could normally be remedied by altering the channel number of the time spectra at which analysis was started. The chi-square test in this case, thus serves as a useful test for determining a good starting point for the analyses.

2.6.2. Direct annihilation rate: zero electric field results.

2.6.2.1. Results of fitting the annihilation rate to functions of the Argon density.

Figure 7 shows the dependence of the direct annihilation rate at zero electric field on Argon density. Table III shows the results of fitting the results shown in Figure 7 to various simple functions of the density. The results, Q , of a chi-square

Figure 7. Direct annihilation rate in Argon at zero electric field as a function of density. The straight line represents $\lambda_a = 5.6 P \times 10^6 \text{ sec}^{-1}$.

calculation are also given. The value of Q is to be interpreted as being the probability of getting a worse fit should the series of experiments be repeated. Unless otherwise stated, the fits were made to all the points shown in Figure 7.

On the basis of the chi-square test alone it would appear that form VI is the best fit to the experimental data. However, there is no reason to suppose that the observed direct annihilation rate is different from zero at zero Argon density, unless a model involving an Argon-positron complex is invoked. According to fit VI such a system would have to have a half-life of about 42×10^{-9} secs, which is considerably longer than that which would be expected (10^{-9} sec), because of the higher electron density at the positron position compared with the parapositronium atom (see Chapter 1, Section 1.4.2.). On the basis of this argument fits V and VI can be rejected. Further examination of Table III shows that below 10 amagats a linear fit IV to the data is adequate. Inclusion of the data obtained at higher densities results in a significantly worse linear fit I. Some improvement is obtained for fits containing a non-linear dependence on density as well as the linear term (fits II and III). It is apparent that a value for a_1 between 5.5 and $5.7 \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$ describes well the linear dependence on Argon density of the direct annihilation rate. Such a value for a_1 is in keeping with the observation that the linear term in fits I to IV is somewhat independent of the detailed nature of the fit. Regarding the non-linearity at higher densities, the tendency is for the annihilation rate to be reduced relative to

TABLE III.

DEPENDENCE OF λ_a ON P.

Summary of the results of fitting the curve in Figure 7.

Type of fit	Results				Q
	$a_0/10^6$	$a_1/10^6$	$a_2/10^6$	$a_3/10^6$	
	10^6 sec^{-1}	sec^{-1}	$\text{sec}^{-1} \text{ amagat}^{-1}$	$\text{sec}^{-1} \text{ amagat}^{-2}$	$\text{sec}^{-1} \text{ amagat}^{-3}$
I. $\lambda_a = a_1 P$		5.42 ± 0.05			0.02
II. $\lambda_a = a_1 P + a_2 P^2$		5.71 ± 0.12	-0.027 ± 0.011		0.08
III. $\lambda_a = a_1 P + a_3 P^3$		5.59 ± 0.08		0.0013 ± 0.0004	0.13
IV. $\lambda_a = a_1 P$		5.53 ± 0.06			0.16
$P < 10 \text{ amagats}$					
V. $\lambda_a = a_0 + a_1 P$	11.44 ± 0.94	5.28 ± 0.11			0.02
VI. $\lambda_a = a_0 + a_2 P^2$	23.85 ± 0.52		0.250 ± 0.006		0.21

Note: Unless otherwise stated, the fits were
made to all the points in Figure 7.

the linear fit acceptable at lower densities. The detailed nature of this non-linearity remains undetermined by these experiments, as examination of Q for fits II and III shows.

2.6.2.2. Discussion of the fits to the data.

The estimated purity of the Argon gas has been given in Table I, Section 2.2.1. The small deviation from linearity of the annihilation rate as a function of density might be due in some way to the Nitrogen present. However, as the direct annihilation rate in Nitrogen is expected to the first order to vary linearly with density, it is difficult to see how the presence of Nitrogen or any other impurity gas could affect the linearity of the direct annihilation rate.

It is far more reasonable to suppose that the non-linearity at high densities arises from the interaction of the positron with more than one Argon atom at a time. The possibility of such an effect has been raised by Tao, Bell and Green (1964)(see also Kivel, 1959). At 15 amagats, the average interatomic distance is about 10^{-7} cm, which is of the same order as the de Broglie wavelength of a thermalized positron. It is also possible that interatomic distances of this magnitude could result in some screening of the field of the positron at the scattering atom, and thus reduce the magnitude of the very important attractive polarization potential. Clearly, this problem can only be resolved by further experiments at high pressures, and by calculations that take into account the presence of more than one scattering atom.

From the results given here it seems that the deviation from linearity of the dependence of the annihilation rate on density is less than 10% at about 17 amagats.

2.6.2.3. Comparison of the linear term with previous results.

The magnitude of the direct annihilation rate per unit amagat is compared in Table IV with the values obtained by other workers. The value resulting from the present work was obtained from the fits to the data as indicated in Section 2.6.2.1. The error associated with the value reflects the statistical errors and also the uncertainty in the exact non-linear behaviour of the annihilation rate. The systematic errors (Sect. 2.6.6.3) associated with this measurement are of the order of 1%. This estimate is not included in the value given in Table IV.

TABLE IV.

Published values of the Direct Annihilation Rate.

Direct annihilation rate $10^6 \text{sec}^{-1} \text{amagat}^{-1}$	Author
5.90 $\pm$ 0.23	Falk (1965)
4.96	Tao, Bell and Green (1964)
5.18	Duff and Heymann (1962)
3.04	Osmon (1965)
5.78	Paul (1964)
5.6 $\pm$ 0.1	Present work

The values presented for other workers differ from the published

values where necessary by the factor $\frac{298}{273}$. This is the factor by which results obtained at room temperature (25°C) have to be multiplied in order to express them in units of $\text{sec}^{-1}\text{amagat}^{-1}$.

The lack of agreement between all the results published thus far arises mainly from the presence of impurities in the gas, and inadequate analysis of results. For example, the result given by Osmon (1965) is significantly smaller than the other values probably because the long-lived orthopositronium component was completely ignored in the lifetime analysis. If such a long-lived component is not taken into account, it can happen that its contribution will raise the tail of the short-lived component relative to the rest of the spectrum. Subsequent analysis as a single exponential then results in an increased lifetime for this short-lived component, and hence in a reduced annihilation rate. The Argon used by Tao, Bell and Green (1964) has subsequently been shown (Tao and Bell, 1966) to contain significant impurities.

The agreement between different workers will only improve when reproducible results are obtained with gases containing less than a total of 1 ppm of impurity. In this sense the results quoted in Table IV should be assumed to be those for impure Argon.

The annihilation rate of $(5.6 \pm 0.1) \times 10^6 \text{sec}^{-1}\text{amagat}^{-1}$ presented here is that appropriate to Argon containing impurities to the extent given in Table I. Since the main impurity is Nitrogen, and since the direct annihilation rate in Nitrogen is about $5.5 \times 10^6 \text{sec}^{-1}\text{amagat}^{-1}$ (Falk, 1965), it can hopefully be assumed that the annihilation rate given here is very little different from

the annihilation rate in pure Argon.

2.6.3. Direct annihilation rate and the valley-to-peak ratio:
electric field results.

Figure 8 illustrates the effect of applied electric field per unit amagat on the direct annihilation rate. For small E/P the annihilation rate begins to decrease rapidly reaching a fairly constant value of about $2.8 \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$ at $90 \text{ V cm}^{-1} \text{ amagat}^{-1}$. A comparison of the current results with those of Falk (1965) indicates good agreement.

The measurements of the valley-to-peak ratio (indicating as discussed (Section 2.4.) the increased formation of positronium) as a function of E/P are plotted on the same graph for comparison. These differ from the measurements of Marder, et al., (1956) in that the rate of rise of the valley-to-peak ratio with increasing applied electric field is considerably less in the present case. Investigation of the shape of the curve as a function of Nitrogen contamination of up to 1% of the total density indicated that the rate of rise can increase somewhat with this gas as an impurity. However, the results of Marder, et al. were not reproduced, probably because some other unknown impurity was present in that Argon. That there was a tendency for their curve to rise more steeply as a function of time is consistent with the evolution of some impurity.

It is clear from Figure 8 that as E/P is increased, there is an increase in the three-photon component at the expense of the

Figure 8. Direct annihilation rate in Argon as a function of E/P. The valley-to-peak ratio measurements indicate the increased orthopositronium formation. The curves are the best lines drawn through all the points.

two-photon component. Thus it is reasonable to suppose that some of the flattening off of the λ_a vs E/P curve arises from the increased positronium formation. In this case, positronium production thus contributes an extra channel by which the overall equilibrium distribution of free positrons can decay. As positronium formation from the equilibrium distribution increases with applied electric field, so does that contribution to the direct annihilation rate λ_f (see Section 2.3.4. Equation 9) which results from the velocity-averaged positronium formation rate. Figure 9 shows the fraction of positrons forming positronium as calculated from Equation 15, Section 2.4. The fraction W of positrons annihilating in the walls of the chamber was calculated from data derived from the dE/dx data for positrons in Argon (Falk, 1965). The constant k was determined assuming that 37% of the positrons stopped in the gas form positronium at zero field (Falk and Jones, 1964). It is quite apparent that for the highest electric fields used a majority of the positrons form positronium.

Table V shows the dependence of the overall number of counts in the long-lived component as a function of electric field. Two sets of data are given corresponding to spectra obtained with the 0.51 MeV single channel analyzer set at the peak ("direct-enhanced") and valley ("ortho-enhanced") positions respectively (see Section 2.2.1.). The product $I_2\tau_2$ is found by integrating the long-lived exponential $I_2\exp(-t/\tau_2)$ (see Section 2.3.4.) from $t=0$ to $t=\infty$, and was calculated using the appropriate values for I_2 and τ_2 found from maximum-likelihood fits to the lifetime spectra.

Figure 9. Fraction of positrons forming positronium as a function of E/P . The error bars take into account only the statistical errors in C_v and C_p .

In the case of the data presented in Table V the zero of time has been arbitrarily defined as channel 89 of the kicksorter, at which point the maximum-likelihood fits were begun.

It is not possible to draw any conclusions from Table V as to the electric field dependence of the total number of ortho-positronium annihilations recorded by the timesorter. The reason for this is that the statistical uncertainty in $I_2\tau_2$ is at least as large as the size of the effect searched for (see Figures 8 and 9), even at the highest fields used.

To summarize, the rapid decrease of annihilation rate as a function of applied electric field shows that the velocity-dependent annihilation rate decreases as the positron velocity increases. The explicit velocity dependence cannot be found from the experimental data unless the velocity-dependent momentum-transfer cross-section is known. Some implications of the shape of annihilation rate dependence on electric field are discussed in Chapter 3, Section 3.5.

2.6.4. The shoulder in the time spectra.

2.6.4.1. Width of the shoulder.

In the case of zero applied electric field, all the time spectra displayed the usual shoulder structure (Tao, Bell and Green, 1964; Falk and Jones, 1964; Osmon, 1965; Paul, 1964) (see for example Figure 5). Since the form of the time spectrum of the direct component following the shoulder is well fitted by a single

TABLE V.

DEPENDENCE OF $I_2\tau_2$ ON ELECTRIC FIELD.

Ortho-enhanced		Direct-enhanced	
$I_2\tau_2$ 10^4 counts	E/P $V\text{ cm}^{-1}\text{ amagat}^{-1}$	$I_2\tau_2$ 10^4 counts	E/P $V\text{ cm}^{-1}\text{ amagat}^{-1}$
6.0 ± 0.3	0	4.7 ± 0.3	0
5.8 ± 0.3	14.4	4.4 ± 0.4	14.0
5.6 ± 0.4	34.7	4.9 ± 0.5	34.7
6.3 ± 0.5	50.7	5.1 ± 0.7	48.9
6.1 ± 0.8	70.7	4.5 ± 0.8	70.7
6.5 ± 1.1	125.1		

Note: The results given above are for Argon at an average density of 9.0 amagats. The terms "ortho-enhanced" and "direct-enhanced" are defined in Section 2.6.3. The errors quoted were obtained by simply compounding the errors in I_2 and τ_2 that were found from the maximum likelihood fits. Each spectrum was normalized to the same total number of 1.28 MeV counts as indicated in Section 2.2.1.

exponential, the end of the shoulder signifies either a thermalized positron velocity distribution, or that the annihilation rate becomes velocity independent at low velocities or some combination of these two. The application of a small electric field, however, reduces the annihilation rate significantly without perturbing the shoulder markedly. This is shown in Figure 10. Since the applied electric field increases the average positron energy at equilibrium, the direct lifetime corresponds to a velocity-dependent annihilation rate averaged over velocities higher than thermal. Therefore, it seems consistent to interpret the single exponential after the shoulder in terms of a thermalized positron distribution. The time width of the shoulder, then, is the time taken for a positron to thermalize from energies corresponding to E_{thr} (8.9 eV). It has already been shown (Chapter 1, Section 1.2.3.) that the time taken for the positron to reach such energies (about 10 eV) is less than the time resolution of the apparatus used.

The shoulder width is inversely proportional to density. The width-density product was measured to be about 340 nsec-amagat, in agreement with previous results (Falk and Jones, 1964; Paul, 1964). A narrower shoulder width would indicate a more rapid slowing down of the positrons, which would occur through the agency of foreign gas atoms. Thus the presence of foreign molecules with low-lying excitation levels would be expected to narrow the shoulder significantly, simply by increasing the energy loss per collision and thereby the rate of thermalization.

A positron scattered by the host atoms, with momentum-

Figure 10. Time spectrum for positrons in Argon at small E/P. P is in amagats, E/P is in $V \text{ cm}^{-1} \text{ amagat}^{-1}$. The continuous curve is the maximum likelihood fit to the points.

transfer cross-sections of the order of πa_0^2 , will suffer about 10^{11} - 10^{12} collisions sec^{-1} at 10 amagats. Thus in 30 nsec the positron suffers 10^3 - 10^4 collisions. An impurity concentration as low as a few parts in 10^4 could be expected to have a significant effect on the shape of the shoulder. Throughout these experiments the shoulder width remained essentially constant showing that here there was no significant change in gas composition from run to run, either due to evolution of impurities from the walls, or due to the different gas samples used.

2.6.4.2. The logarithmic slope of the shoulder.

A second feature of the shoulder is its reasonably constant logarithmic slope. Furthermore it appears that the shape of the time spectrum in the region of the shoulder is somewhat independent of the shape of the orthopositronium component in that region. This is demonstrated by comparing the two time spectra in Figure 11 which were obtained with the 0.51 MeV single channel analyzer set at the peak (direct-enhanced) and valley (ortho-enhanced) positions in turn (see Section 2.2.1.). In the region of the shoulder there is very little difference in the time spectra, despite the fact that the relative intensities of the direct and orthopositronium components differ in the two cases. Such a situation could arise if the orthopositronium component underlying the shoulder has a logarithmic slope little different from the logarithmic slope of the direct annihilation contribution to the shoulder. This indeed seems to be the case for Argon at

Figure 11. Comparison of direct- and ortho-enhanced time spectra obtained at $P=4.9$ amagats, $E/P=0$ V cm^{-1} amagat $^{-1}$. The continuous curves are the maximum likelihood fits to the points. A - direct-enhanced; C - ortho-enhanced.

a density of 4.9 amagats (Figure 11). For higher densities these logarithmic slopes should differ to a greater extent, because of the different pressure dependence, and hence the shoulder in the direct-enhanced time spectrum should be somewhat different from that in the ortho-enhanced time spectrum. Figure 12 shows such a pair of spectra obtained at 9.3 amagats. Because the shoulder is much narrower in this case, it is clearly difficult to draw any conclusions here concerning the previous argument.

If it is assumed, however, that all the positronium is formed (for zero electric field) at the time corresponding to the prompt peak, it is possible to extrapolate the orthopositronium component in the region of the two exponentials back into the region of the shoulder. Subtraction of the orthopositronium component in the shoulder region in this fashion and fitting the resulting curve graphically to a single exponential yields an average annihilation rate in the shoulder region of about $1.5 \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$ for the two spectra in Figure 11. For the two spectra in Figure 12 the result is $\sim 1.0 \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$. This agrees fairly well with the figure of $1.2 \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$ given by Osmon (1965) (this value has been corrected as indicated in Section 2.6.2.3.). The size of this "shoulder annihilation rate", in relation to the direct annihilation rates observed as a function of electric field, is indicated in Figure 8.

A reasonably constant logarithmic slope of the direct annihilation contribution to the shoulder cannot be interpreted simply in terms of a changing annihilation rate as a function of

Figure 12. Comparison of direct- and ortho-enhanced time spectra obtained at $P=9.3$ amagats, $E/P=0$ V·cm⁻¹ amagat⁻¹. The continuous curves are the maximum likelihood fits to the points.

time. In order that a constant logarithmic slope appear in the direct component of the time spectra, either the positron velocity distribution must be at equilibrium, or the direct annihilation rate must be essentially velocity independent over the appropriate range of the shoulder. This is discussed in detail in Chapter 3, Section 3.4. It has already been pointed out that the single exponential following the shoulder corresponds to annihilation from the equilibrium velocity distribution. Thus it is clear that the constant logarithmic slope in the shoulder region can only arise from a reasonably velocity independent annihilation rate at velocities somewhat higher than thermal.

Therefore, it appears that in a velocity range appropriate to the time span defined by the shoulder, the direct annihilation rate is approximately velocity independent, subject to the assumption that all the positronium formation occurs at the time corresponding to the prompt peak. The small shoulder annihilation rate should be taken as evidence that the velocity dependent direct annihilation rate can become as small as $\sim 1.5 \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$. Furthermore, since the shoulder has a constant logarithmic slope over the greater part of its extent, it seems likely that the velocity-dependent direct annihilation rate is constant over some of the energy range between 8.9 eV (the positronium formation threshold) and 1/40 eV (thermal energy).

2.6.4.3. Effect of the electric field on the shoulder.

It has been suggested in the previous subsection that

the velocity averaged annihilation rate corresponding to the time interval occupied by the shoulder is due to a somewhat velocity independent annihilation rate at velocities higher than thermal. Furthermore, this shoulder annihilation rate has been shown to be considerably less than the annihilation rate at thermal velocities. Reference to Figure 8 shows that the application of an electric field reduces the direct annihilation rate, but that this does not fall below the shoulder annihilation rate. This is so presumably because of the onset of increased positronium formation as the electric field is applied. However, it is evident that the shoulder disappears as the electric field is increased (Fig. 13) principally because the direct annihilation rate averaged over the velocity distribution at high E/P tends towards the shoulder annihilation rate.

Lack of knowledge of the initial velocity distribution characterizing the velocity distribution of positrons with energies below 11.6 eV (see Chapter 1, Section 1.2.4.) prevents a more detailed discussion of the way in which the shoulder changes as a function of electric field. It can only be stated that the observations are entirely consistent with the view of a velocity-dependent annihilation rate which decreases as a function of increasing velocity.

2.6.5. Orthopositronium annihilation rate.

2.6.5.1. Fitting of experimental data.

Figure 14 shows the Argon-density dependence of the annihilation rate appropriate to the long-lived component of the time spectra. Only those results are presented which satisfied the criteria discussed in Section 2.6.1. and are thus for the same spectra as are the direct annihilation rates discussed in Section 2.6.2.

The results of fitting the data in Figure 14 to simple functions of the density are shown in Table VI. The meaning of Q is the same as in Section 2.6.2.1. Unless indicated, the fits were made to all the experimental values in Figure 14. Of the 27 points presented, 11 were obtained with an applied electric field. The functions to which the data have been fitted can be divided into two categories. Either the parameter a_0 is determined by the data (fits I to VII) or it is (fits VIII to X) the theoretical free orthopositronium annihilation rate ($7.2 \times 10^6 \text{ sec}^{-1}$; see Chapter 1, Section 1.4.2.).

The relatively large scatter of the results in Figure 14 is reflected in the poor fits obtained in Table VI. However, some systematic trend is apparent. Fit I, which contains no density dependent terms, is clearly unacceptable. For densities less than 10 amagats, with $E/P=0$, a good linear fit II is obtained. Including the electric field results, the fit VI is obtained. The parameters a_0 and a_1 are relatively unchanged but the fit is judged worse by

Figure 14. Orthopositronium annihilation rate in Argon as a function of density. The straight line represents the function $\lambda_0 = (7.2 + 0.29 P) \times 10^6 \text{ sec}^{-1}$.

the chi-square test. A similar situation holds for densities less than 20 amagats where the corresponding fits are III and VII.

As discussed previously, the long-lived component is expected to be due to orthopositronium annihilation, while the change in annihilation rate as a function of gas density is associated with the quenching of the free orthopositronium lifetime (Chapter 1, Section 1.4.4.). The parameter a_0 , then, is the free orthopositronium annihilation rate as measured by these experiments. In view of the theoretical prediction of this lifetime (Chapter 1, Section 1.4.2.) and the associated annihilation rate ($7.2 \times 10^6 \text{ sec}^{-1}$), fit V has little physical justification. The value of a_0 for this fit, which contains only a quadratic dependence on density, is considerably different from the theoretical orthopositronium annihilation rate, if compared with the a_0 from the linear fits to the data. A similar argument holds for fit IV where a linear term is included. However, in view of the reasonable Q , and the fact that the value for a_0 is significantly closer to the theoretical orthopositronium annihilation rate, the possibility of a quadratic density dependence in addition to the linear term can not be ruled out by the set of data presented.

In the fits VIII to X the parameter a_0 is given the value of $7.2 \times 10^6 \text{ sec}^{-1}$, the theoretical orthopositronium lifetime. As expected the fits are considerably worse than those obtained where a_0 was not fixed. This is so since the value for a_0 from any of the latter yields the best fit to the data, and these values for a_0 were not equal to the $7.2 \times 10^6 \text{ sec}^{-1}$. The

TABLE VI.

DEPENDENCE OF $\bar{\lambda}_0$ ON P.

Summary of the results of fitting the curve in Figure 14.

	Type of fit	Results			Q
		$a_0/10^6$	$a_1/10^6$	$a_2/10^6$	
	10^6 sec^{-1}	sec^{-1}	$\text{sec}^{-1} \text{ amagat}^{-1}$	$\text{sec}^{-1} \text{ amagat}^{-2}$	
I.	$\bar{\lambda}_0 = a_0$	10.57 ± 0.05			$< 10^{-6}$
II.	$\bar{\lambda}_0 = a_0 + a_1 P$ E/P = 0, P < 10 amagats	6.83 ± 0.50	0.339 ± 0.059		0.88
III.	$\bar{\lambda}_0 = a_0 + a_1 P$ E/P = 0	7.55 ± 0.18	0.240 ± 0.013		0.16
IV.	$\bar{\lambda}_0 = a_0 + a_1 P + a_2 P^2$ E/P = 0	7.97 ± 0.59	0.164 ± 0.102	0.0030 ± 0.0041	0.15
V.	$\bar{\lambda}_0 = a_0 + a_2 P^2$ E/P = 0	8.90 ± 0.11		0.0095 ± 0.0005	0.10

TABLE VI (continued)

DEPENDENCE OF $\bar{\lambda}_0$ ON P.

	Type of fit	Results			Q
		$a_0/10^6$	$a_1/10^6$	$a_2/10^6$	
	10^6 sec^{-1}	sec^{-1}	$\text{sec}^{-1} \text{ amagat}^{-1}$	$\text{sec}^{-1} \text{ amagat}^{-2}$	
VI.	$\bar{\lambda}_0 = a_0 + a_1 P$ $P < 10 \text{ amagats}$	7.07 ± 0.48	0.305 ± 0.056		0.02
VII.	$\bar{\lambda}_0 = a_0 + a_1 P$	7.54 ± 0.15	0.242 ± 0.011		0.004
VIII.	$\bar{\lambda}_0 = 7.2 + a_1 P$ $P < 10 \text{ amagats}$		0.294 ± 0.008		0.017
IX.	$\bar{\lambda}_0 = 7.2 + a_1 P$		0.267 ± 0.004		0.0004
X.	$\bar{\lambda}_0 = 7.2 + a_1 P + a_2 P^2$		0.303 ± 0.016	-0.0024 ± 0.0010	0.0012

Note: Unless otherwise stated, the fits were made to all the points in Figure 14. The electric field E/P is in units of $\text{V cm}^{-1} \text{ amagat}^{-1}$.

values for the parameter a_1 in fits VIII to X differ little from the a_1 obtained in the fits where a_0 was allowed to vary.

2.6.5.2. Discussion of the linear density dependence of the orthopositronium annihilation rate.

As mentioned earlier, the annihilation rate at zero gas density is expected to be the free orthopositronium annihilation rate. This annihilation rate increases by pick-off quenching as the density increases (see Chapter 1, Section 1.4.4.). The quenching cross-section will in general be velocity dependent, and the velocity-dependent quenching rate proportional to the gas density. The velocity-averaged quenching rate λ_q is thus also density dependent. On the basis of the discussion presented here, the density dependence of the quenching rate would be expected to be linear.

The large scatter of the experimental points is interpreted to reflect slight differences in the type and concentration of impurities in the Argon gas from run to run. On the average the quenched orthopositronium atom exists for about 100 nsec in 10 amagats of Argon. This is considerably longer than the width of the shoulder observed in the same time spectra (Section 2.6.4.). In addition, the elastic-scattering cross-section for orthopositronium is expected to be less than that for free positrons where the long-range attraction is proportional to $1/R^4$ (Chapter 1, Section 1.3.2.; Chapter 3, Section 3.1.) as compared with the long-range $1/R^6$ van der Waals attraction expected for the positron-

ium case. For an elastic scattering cross-section of the order of πa_0^2 the orthopositronium atom would make of the order of 10^{11} collisions/sec at 10 amagats. Thus the orthopositronium atom makes of the order of 10^4 collisions before annihilating. Should one of these collisions involve an impurity atom with a large quenching cross-section, the orthopositronium lifetime will be considerably shortened. It thus appears that the orthopositronium annihilation rate is at least as sensitive to impurities as the shoulder width in the Argon time spectra. The level of Nitrogen contamination found in many of the Argon samples used in these experiments (Table I) is probably high enough to explain these discrepancies.

2.6.5.3. Influence of the electric field.

Although there is no physical basis for expecting any electric field dependence in the orthopositronium annihilation rate, the data has been checked for such an effect. An examination of the data in Table VI shows no evidence for any electric field dependence. The increased scatter of the annihilation data, when the electric field results are included (reflected in the smaller value for Q) is most probably due to the effect of foreign gases liberated by undetected high-voltage breakdowns during a run. It is expected that foreign gases added to the Argon in this manner would not be immediately removed by the purifier, and could thus affect the measured orthopositronium annihilation rate as discussed in Section 2.6.5.2.

2.6.5.4. Summary of the orthopositronium results.

From Table VI, Figure 14, and the above discussion it is clear that some density dependence of the orthopositronium annihilation rate is necessary. A statistical analysis of the data indicates that a linear dependence on density is sufficient if the electric field results are excluded. Inclusion of the electric field results worsens the fit but does not change the value of the parameters significantly.

Comparison of the fits in Table VI which contain a linear density dependence and a zero density intercept a_0 , indicates a value for a_0 between 6.8 and $7.6 \times 10^6 \text{ sec}^{-1}$, if the results of fit IV are neglected (Section 2.6.5.1.). Such a range for a_0 takes into account the systematic error introduced by the electric field as discussed in Section 2.6.5.3. On this basis a free orthopositronium annihilation rate of $(7.2 \pm 0.4) \times 10^6 \text{ sec}^{-1}$ is given by these experiments, in good agreement with the theoretical prediction of $7.2 \times 10^6 \text{ sec}^{-1}$.

The values for a_1 in Table VI all lie between 0.24 and $0.34 \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}$ if once again fit IV is neglected. Furthermore, the values for a_1 in fits VIII to X lie within these limits. Finally, this range of values for a_1 also allows for the systematic error introduced by the electric field. The linear quenching rate λ_q given as a result of these experiments is then

$$\lambda_q = (0.29 \pm 0.05) \times 10^6 \text{ sec}^{-1} \text{ amagat}^{-1}.$$

It should be emphasized that the quenching rate has been obtained with about 100 ppm of Nitrogen present (Table I) and would be

expected to decrease somewhat for Argon containing less than 1 ppm of any impurity.

Table VII compares the present results with previously published values. Both of the quadratic fits (Celitans, Tao and Green, 1964; Celitans and Green, 1964) were obtained from data where the shoulder width was about 90 nsec-amagats, which is at least a factor of three smaller than the currently accepted value. This smaller shoulder width is a result of a relatively large impurity concentration (Tao and Bell, 1965), and following the arguments in Section 2.6.5.2., the orthopositronium lifetimes were probably affected to a considerable extent. The results of Heymann et al.(1961) were obtained from analysis of 0.51 MeV gamma-ray spectra, and agree well with the values presented in this thesis which were obtained from lifetime measurements.

2.6.6. Discussion of errors not related to counting statistics.

All the standard deviations quoted thus far, except where it is explicitly stated otherwise, are based on the effect of counting statistics only. It is necessary also to discuss the role of errors introduced by instabilities in the electronic instrumentation, by inaccuracies associated with the measurement of the integral and differential linearities of the timesorter, the applied electric field and the gas density, and by changes in the gas composition from run to run.

TABLE VII.

SUMMARY OF PUBLISHED RESULTS FOR ORTHOPOSITRONIUM QUENCHING IN ARGON.

Type of density dependence	Results			Reference
	$a_0/10^6$	$a_1/10^6$	$a_2/10^6$	
10^6 sec^{-1}	sec^{-1}	$\text{sec}^{-1} \text{ amagat}^{-1}$	$\text{sec}^{-1} \text{ amagat}^{-2}$	
$\bar{\lambda}_o = 7.0 + a_2 P^2$			0.017 ± 0.002	Celitans, Tao and Green (1964).
$\bar{\lambda}_o = 7.0 + a_2 P^2$			0.015 ± 0.002	Celitans and Green (1964).
$\bar{\lambda}_o = 7.0 + a_1 P$		0.277 ± 0.005		Heymann, et al. (1961).
$\bar{\lambda}_o = a_0 + a_1 P$	7.2 ± 0.4	0.29 ± 0.04		Present work.

Note: The results due to other workers have been corrected by the factor 298/273 (see Section 2.6.2.3.) on the assumption that the atmosphere units given by them are at 25°C. The value of the free orthopositronium annihilation rate assumed by the other workers is not in accordance with the calculated values (Ore and Powell, 1949; Alekseev, 1959).

2.6.6.1. Effect of instabilities in the electronic instrumentation.

The discussion of the effect of electronic instability can be divided into two sections. In this subsection the effects of changes brought about in the prompt resolution and in the prompt peak position are considered. The following subsection deals with the uncertainties in the time calibration of the timesorter. Figure 20 (Appendix), shows the effect on the time resolution of making a large change in the 0.51 MeV single channel analyzer setting. One curve was obtained with the single channel analyzer set at the 0.51 MeV photopeak, the other with the single channel analyzer set at the valley position (see Section 2.2.1. and Fig. 5). The resolution is somewhat different for the two settings. Table VIII shows the results of lifetime measurements for some representative pairs of time spectra where the main difference was the single channel analyzer setting. Taking note of the standard deviations in the results, there is little evidence for a marked trend. To first order then, changes in the electronics which modify the time resolution to the extent indicated in Figure 20, have no observable effect on the lifetime measurements. The effect is expected to be significant only when the lifetimes are of the order of, or less than, the prompt time resolution.

During the course of the experiments the prompt peak (see Figure 6) of the time spectra was observed to shift a maximum of ± 1 channel (2.7 nsec). A shift in the prompt peak of this magnitude during a run will have no effect on the lifetime of the exponentials but does modify the intensities. In fact, any non-

TABLE VIII.

DEPENDENCE OF LIFETIME RESULTS ON S.C.A. SETTING.

<u>Direct-enhanced</u>			<u>Ortho-enhanced</u>		
τ_1	τ_2	P	τ_1	τ_2	P
nsec	nsec	amagats	nsec	nsec	amagats
18.3 ± 1.7	97.8 ± 2.6	9.3	19.4 ± 0.7	101.2 ± 2.1	9.3
37.8 ± 1.7	90.3 ± 4.9	9.0	38.7 ± 3.4	101.0 ± 5.0	9.0
27.9 ± 0.8	95.0 ± 4.0	9.0	29.3 ± 1.5	105.5 ± 3.3	9.0
22.0 ± 0.5	107.0 ± 4.0	9.0	24.2 ± 1.0	108.5 ± 2.4	9.0
20.7 ± 0.5	104.3 ± 3.0	8.9	18.6 ± 1.0	100.5 ± 2.0	8.8
34.3 ± 1.4	90.3 ± 4.3	9.0	33.1 ± 2.4	98.8 ± 3.4	8.7
13.9 ± 0.4	95.8 ± 2.0	13.5	14.2 ± 0.6	95.2 ± 1.4	13.5
35.2 ± 1.5	112.9 ± 8.9	5.3	35.6 ± 3.8	114.8 ± 5.9	5.3

exponential portion of the time spectrum such as the shoulder region is modified by a shift in the prompt peak.

Other instabilities in the electronics are expected to have an effect considerably less than the accuracy to which the integral and differential linearities of the timesorter have been measured.

2.6.6.2. The integral and differential linearities of the timesorter.

The integral linearity of the timesorter was measured using the method described by Jones and Falk (1965). The overall error associated with the measurement of the average time width per channel given by the integral linearity is estimated to be less than 1%, arising mainly from the uncertainty in reading the time between pulses from the pulser. The integral linearity measurement was repeated during the course of the experiment in order to check that there were no significant changes in the average time width per channel. The second measurement, separated from the first by two months was within 1% of the first measurement (see Appendix).

The differential linearity (or relative channel width) is measured by the relative number of counts in each channel of the timesorter, when the input to the timesorter is a source of pulse pairs separated by time intervals of a random length (Falk, Jones and Orth, 1965). The relative channel widths are directly proportional to the relative number of counts in each channel. The accuracy of the differential linearity measurement

is thus governed by the counting statistics, and since about 1000 counts per channel were recorded, the relative channel widths are accurate to about 3%. The integral and differential linearities of the timesorter are shown in Figure 22 (Appendix).

It is clear that the use of the differential linearity in the fitting of the experimental data (see Section 2.5.1.) already constitutes a first order correction to the estimation of the lifetimes. Any uncertainty in the relative channel widths must then be considered as affecting this correction to the lifetime measurements (when compared with the uncertainty in the integral linearity). From this point of view it is reasonable to suppose that the total uncertainty in the lifetimes due to the calibration of the timesorter is of the order of 1%, this figure representing the maximum systematic error due to the integral linearity measurement.

2.6.6.3. Systematic error in the annihilation rates.

Taking into account the above discussion, it is clear that the systematic error in the measurement of the annihilation rates is of the order of 1%, and arises from the uncertainty in the absolute time calibration of the timesorter.

2.6.6.4. Applied electric field.

Since much of the analysis of the experiments relies on the assumption of a spatially uniform electric field, it is necessary to discuss, not only the uncertainty in the magnitude of this electric field, but also the degree to which this electric

field is uniform within the chamber.

The voltage applied to the electric field rings was found by measuring the current flowing through a selected 500 M Ω resistor connected in parallel with the electric field rings. The value of the resistor was measured to 1% at 20.0 kV and 0.5% at 3 kV, and was found to have a negligible voltage coefficient. The Avometer used to measure the current through the resistor was accurate to 1% for a full scale deflection corresponding to the highest fields used. The distance between the ground plate and the high voltage ring has been previously reported by Falk (1965) and is known to less than 1%. The overall uncertainty in the magnitude of the electric field taking into account these factors is of the order of 3% for all electric fields measured.

The spatial uniformity of the electric field was investigated by Falk (1965) using a low-voltage two-dimensional analogue of the grid structure. The results indicated that the non-uniformities were confined to the region immediately surrounding the electric field rings, and occupied about 8% of the volume enclosed by these rings. The effect of these non-uniformities will be less marked at high Argon densities, because of the reduced positron range, compared with lower densities. The points in Figure 9 have been obtained for a variety of densities, and lie on a continuous curve, depending on the E/P. In view of this, it is considered that the small non-uniformities in the electric field are unimportant.

2.6.6.5. Measurement of gas density.

The density of the gas was found using the perfect gas law. Deviations from this law as expressed by the van der Waals equations are negligible compared with the uncertainty in the pressure measurement. This uncertainty is associated with the calibration of the high pressure gauge and is estimated to be 2% (Falk, 1965). The measurement of the absolute temperature of the gas was done using a mercury thermometer with the bulb placed against the chamber wall. There is a possibility that the temperature of the gas was higher than that of the chamber walls, due to the heating action of the purifier. However, there was no evidence that the temperature of the chamber walls increased when the purifier was turned on, indicating that most of the gas is in thermal equilibrium with the chamber walls.

The average absolute temperature of the gas was known in this way to within 1% during a run. The uncertainty in the density measurement is thus of the order of 3%, taking into account uncertainties in both the pressure and temperature measurements.

2.6.6.6. Uncertainty in E/P.

In the light of the discussions in the previous two subsections, the uncertainty in the measurement of E/P is of the order of 6% which is obtained by compounding the uncertainties in E and P.

2.6.6.7. Gas composition.

In experiments of the type reported here, it is desirable to be fairly certain that the gas composition remained constant over the length of time that the experiments were performed. The effectiveness of the Ca-Mg eutectic purifier in maintaining the gas composition over a period of time has been discussed in Section 2.2.1. The possibility that small changes in impurity concentration have affected the lifetime results is discussed in Section 2.6.2.3. and Section 2.6.5.2.

3. THEORETICAL CONSIDERATIONS OF THE POSITRON-ARGON ATOM INTERACTION.

3.1. Introduction.

The subject of low-energy positron-atom interactions has received considerable attention recently (Massey, et al., 1966; Drachman, 1966) due to the development of experimental techniques which make comparison between theory and experiment possible (Falk, Orth and Jones, 1965; Paul, 1964). The overall non-relativistic time-independent Hamiltonian H for a system of electrons bound to a nucleus, and an unbound positron is given by

$$H = \frac{\hbar^2}{2m_N} \nabla_N^2 - \frac{\hbar^2}{2m_e} \sum_i \nabla_i^2 - \frac{\hbar^2}{2m_e} \nabla_p^2 - \sum_i \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_N|} + \sum_i \sum_{j \neq i} \frac{1}{2} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} + \frac{Ze^2}{|\mathbf{r}_p - \mathbf{r}_N|} - \sum_i \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_p|} \quad (34a)$$

where the subscript N denotes the nucleus, p denotes the positron and summation over i refers to the Z electrons in the atom (see Mott and Massey, 1965, p. 287). The wave equation describing the complete system is given by

$$H\psi = E\psi. \quad (34b)$$

E is the total energy of the system.

The solution to a system such as (34a) and (34b) is beyond the scope of the present day techniques, and rather drastic simplifications have to be made in order that the problem become at all tractable.

The simplest approximation to this many-particle problem involves the use of an effective two-body interaction. In the field of the low energy electron scattering from noble gas atoms, such a simplification has met with considerable success (Holtsmark, 1929; Kivel, 1959; Labahn and Callaway, 1966). In this model the incoming electron scatters from the unperturbed atom, the interaction potential being that due to the average electronic charge distribution in the atom.

The long range electric polarization of the atom by the incoming electron, expected from first order perturbation theory (Crown and Russek, 1965), is taken into account by the addition, to the atomic potential, of an attractive term which behaves as α/R^4 for large electron-atom separations R . The parameter α is chosen to be the classical electric polarizability of the atom. Early work by Holtsmark (1929) showed that a two-body interaction consisting of the two potentials indicated above was able to reproduce the Ramsauer effect in Argon and other noble gases in some detail. Much of the recent work in the scattering of electrons from noble gases has confirmed this point of view (Kivel, 1959; Labahn and Callaway, 1966).

3.1.1. Discussion of the differences between low-energy positron and electron scattering.

In a recent review of the subject of approximate polarization potentials for the electron-Helium interaction (Labahn and Callaway, 1966), the overall lack of sensitivity of the velocity-

dependence of the total scattering cross-sections to details of the effective scattering potential is clearly demonstrated. Good agreement with experiment is obtained for a variety of potentials as long as they exhibit a $1/R^4$ dependence for large R , even though they differ substantially at the edge of the Helium atom. This chapter contains the results of calculations which, when compared with the experimental results of Chapter 2, indicate that the case of positron-Argon scattering is far less ambiguous.

It is possible in such a calculation to compute not only the scattering cross-section as a function of energy but also the positron-electron annihilation rates. The latter depend solely on the overlap of positron and electron density in the atom. An experiment which depends on both the scattering cross-sections and annihilation rates as a function of velocity should thus impose more stringent conditions on the choice of potential than experiments which depend on one alone.

Such a situation is provided in the annihilation of positrons elastically scattering in Argon gas under the influence of an applied dc electric field. The positron velocity distribution is then determined to first order by the momentum-transfer cross-section and electric field. The observed annihilation rate is given by the velocity-dependent annihilation rate averaged over this velocity distribution. Thus, in general, the annihilation rate is a function of the applied electric field.

Further, the scattering of positrons from atoms differs significantly from that for electrons in that the positron is

distinguishable and is thus not prevented by the Pauli principle from having significant wave function overlap with the electrons of the atom. For this reason, short-range positron-electron correlation effects may be expected to play a more important role than in the corresponding case for electron scattering. The annihilation rate, in particular, should reflect the detailed extent to which both the atomic and positron wave functions are distorted by the interaction. Finally, it may be expected that both long-range polarization of the atom and short-range correlation of the positron-electron wave functions will play an important part in the effective interaction.

As mentioned in Section 3.1., the problem of positron-atom scattering is complicated by the many particle aspect. It has been pointed out that a very simple two-body approximation has been particularly useful in the electron scattering case. It is desirable, then, to know the extent to which such two-body approximations apply to the case of positron scattering. Furthermore, it is of interest to establish whether the ambiguity which arises in electron scattering occurs for positrons. Accordingly, the theoretical results of this Chapter were obtained using the empirical polarization potentials of the type used in electron scattering. These take into account the long-range polarization in terms of a potential whose asymptotic behaviour is $1/R^4$ (see also Moussa, 1959). Some cut-off parameter is always used in order that the polarization potential V_p remain finite at the origin. The effective interaction V is taken to be the sum of V_p and the potential V_H characterizing the Hartree-Fock self consistent field for Argon in the ground

state (Hartree and Hartree, 1936). The results indicate quite clearly that these simple two-body potentials are inadequate to account for the experimental results.

3.1.2. Outline of procedure.

In order to compute the effect of the electric field on the annihilation rate of positrons in a gas both the velocity dependent annihilation rate $\nu_a(v)$ and the momentum-transfer cross-section $\sigma_d(v)$ have to be known. For a specific model of the positron-atom interaction, these can be found by obtaining the usual partial wave solution to the appropriate Schrodinger equation. It should be emphasized that the analysis of this Section neglects any contribution to the annihilation rate from radiative capture of a positron into a bound Ar-e^+ system. Neither the existence nor probability of formation of such bound states has been discussed in any quantitative way in the literature. Should it exist, it is expected that the lifetime would be of the order of the parapositronium lifetime (10^{-10} sec). If the capture rate is comparable, then, to the direct annihilation rate due to the positron-electron overlap during elastic scattering, competition from this channel could represent a significant contribution to the overall observed direct annihilation rate.

The velocity distribution of positrons under the influence of an electric field in a gas at temperature T is then determined using the $\sigma_d(v)$ and $\nu_a(v)$ discussed above. The differential equation describing this situation is similar to the Wilkins

equation used to describe thermal neutron diffusion where neutron capture is important (Sobrino and Clark, 1961), except that, in this case, there is an additional term due to the applied electric field. In addition, the positrons in the gas approximate a Lorentz gas to a very high degree by virtue of their small mass and low density. The resulting differential equation has been reported previously in preliminary work done on this problem (Falk, 1965; Falk, Orth and Jones, 1965).

Once the velocity distribution is obtained for a particular $\sigma_d(v)$, $v_a(v)$ and electric field, the direct annihilation rate λ_a is found by averaging the velocity-dependent annihilation rate over the entire velocity distribution.

3.2. The two-body Schrodinger equation and its solution.

3.2.1. The Schrodinger equation.

Because of the spherical symmetry of the effective positron-Argon atom interaction assumed, the problem is reduced to solving the radial part of the relevant Schrodinger equation. This is, in atomic units (Wu and Ohmura, 1962)

$$\left[\frac{d^2}{dr^2} + k^2 - \frac{l(l+1)}{r^2} - 2V(r) \right] x_l = 0 \quad (35)$$

$$\text{where } V(r) = \frac{Z_p(r)}{r} - V_p \quad (35a)$$

is the effective interaction potential at a distance r from the origin, and k^2 is the kinetic energy of the incident positron (in Rydberg units)

The term $Z_p(r)$ takes into account the screening of the

nucleus by the atomic electrons. Thus, for Argon

$$Z_p(0) = 18 a_0^{-1},$$

and $Z_p(\infty) = 0 a_0^{-1}$, where a_0 is the Bohr radius.

For all the potentials used here, the Hartree-Fock part of the interaction ($Z_p(r)/r$), ascribed to the unperturbed Argon atom, is due to Hartree and Hartree (1936).

3.2.2. Calculation of phase shifts and wave functions.

The solution of (35) was undertaken using the Runge-Kutta method for solving differential equations. The method of solution consisted of integrating the differential equation numerically from a point near the origin until the asymptotic form for x_ℓ , which is known analytically, is obtained. In order to begin the integration, the slope of x_ℓ , dx_ℓ/dr , was set to some arbitrary initial value at a small distance r away from the origin. For non-zero ℓ , and $kr \ll \ell$, the corresponding initial value of x_ℓ obtained by solving

$$\left[\frac{d^2}{dr^2} - \frac{\ell(\ell+1)}{r^2} \right] x_\ell = 0 \quad (36)$$

is given by

$$x_\ell = \frac{r}{\ell+1} \frac{dx_\ell}{dr}. \quad (37)$$

This ensures that x_ℓ has the correct relationship to its slope near the origin.

For $\ell=0$, and $rZ_p(r) \ll 1$, the initial value of x_0 is given by

$$x_0 = r \frac{dx_0}{dr} \quad (38)$$

where x_0 is the solution to

$$\left[\frac{d^2}{dr^2} - \frac{2Z_p(r)}{r} \right] x_0 = 0 \quad (39)$$

in the region where $Z_p(r) \approx Z$. Expressions (37) and (38) can be combined to give

$$x_\ell = \frac{r}{\ell+1} \frac{dx_\ell}{dr} \quad (40)$$

for $kr \ll \ell$ if $\ell > 0$, and $rZ_p(r) \ll 1$ if $\ell = 0$.

The absolute magnitude of x_ℓ was determined by matching the numerical solution for x_ℓ to the appropriate asymptotic expression for x_ℓ . The value of the asymptotic x_ℓ was found by normalizing the incoming positron flux to 1 positron $\text{cm}^{-2} \text{sec}^{-1}$ in the usual manner (Wu and Ohmura, 1962).

3.2.2.1. Asymptotic solution for $k \neq 0$.

For k unequal to zero, the asymptotic solution for x_ℓ in the region where the $V(r)$ term is negligible compared with the others in (35) is

$$x_\ell(r) = kr [A_\ell j_\ell(kr) - B_\ell n_\ell(kr)] \quad (41)$$

where $j_\ell(kr)$, $n_\ell(kr)$ are spherical Bessel functions of the first and second kind. For still larger r where, in addition, the $\frac{\ell(\ell+1)}{r^2}$ term is negligible compared with k^2 , this becomes (Wu and Ohmura, 1962)

$$x_\ell(r) = C_\ell \sin \left(kr - \frac{\ell\pi}{2} + \delta_\ell \right) \quad (42)$$

where

$$\tan \delta_\ell = \frac{B_\ell}{A_\ell} \quad (43)$$

and

$$C_l^2 = A_l^2 + B_l^2. \quad (44)$$

Thus it is unnecessary to compute the wave function up to values of r for which it becomes sinusoidal, since the phase shift and normalization constant can be calculated when the wave function satisfies (41). For any r satisfying the asymptotic requirement ($V(r)$ is negligible), the constants A_l , B_l can be calculated from x_l and $\frac{dx_l}{dr}$. Differentiating (41) with respect to r gives

$$\frac{dx_l}{dr} = (l+1) x_l/r - k r [A_l j_{l+1}(kr) - B_l n_{l+1}(kr)] \quad (45)$$

where use has been made of the relation

$$\frac{l}{x} f_l(x) - \frac{d}{dx} f_l(x) = f_{l+1}(x)$$

where f_l is any spherical Bessel function. Solving (41) and (45) for A_l , and B_l yields

$$\begin{aligned} A_l &= x_l [(l+1) n_l(kr) - k r n_{l+1}(kr)] - r \frac{dx_l}{dr} n_l(kr) \\ B_l &= x_l [(l+1) j_l(kr) - k r j_{l+1}(kr)] - r \frac{dx_l}{dr} j_l(kr) \end{aligned} \quad (46)$$

In practice the ratio B_l/A_l was continuously monitored as the differential equation (35) was solved. Once the ratio B_l/A_l converged, the calculation was terminated, and the phase shift δ_l and normalization constant C_l found using equations (43) and (44) respectively. The phase shifts were then used to calculate the total momentum-transfer cross-sections σ_d (in units of πa_0^2) from (Bowe, 1960)

$$\sigma_d = \frac{4}{k^2} \sum_{l=0}^{\infty} (l+1) \sin^2(\delta_l - \delta_{l+1}) \quad (47)$$

In practice, all the phase shifts for partial waves up to and

including $\ell=5$ were calculated, the higher order phase shifts being negligible.

3.2.2.2. Asymptotic solution for $k=0$.

For the case of very small incident velocity, and large r (that is, k^2 and $V(r) \ll \ell(\ell+1)/r^2$), the Schrodinger equation (35) simplifies to

$$\left[\frac{d^2}{dr^2} - \frac{\ell(\ell+1)}{r^2} \right] x_\ell = 0 \quad (48)$$

This free particle equation has as its general solution

$$x_\ell = C_1 r^{\ell+1} + C_2 / r^\ell \quad (49)$$

However, it is only necessary to compute the wave function for $\ell=0$ for this special case as all the other phase shifts are small in comparison with δ_0 . This occurs only because all the potentials used here fall off more rapidly than $1/r^3$ (Landau and Lifshitz, 1965; p.500). Therefore, x_0 is appropriately normalized if at large r

$$x_0 = r + C_2 \quad (50)$$

The constant C_1 has been set to unity since asymptotically the wave function $R = x/r$ has unity amplitude.

The momentum-transfer cross-section is now identical to the total elastic scattering cross-section, the latter being expressed by (Landau and Lifshitz, 1965; p.500)

$$\sigma = 4(C_2/C_1)^2 \quad (51)$$

If the wave function is normalized (that is, $C_1=1$), this reduces to

$$\sigma = 4C_2^2 \quad (52)$$

For smaller values of r , for which the α/r^4 term in $V(r)$ is significant and the k^2 term still negligible, the solution to the Schrodinger equation for $\ell=0$

$$\left[\frac{d^2}{dr^2} + \frac{2\alpha}{r^4} \right] x_0 = 0 \quad (53)$$

is given by (Landau and Lifshitz, 1965; p.504)

$$x_0(r) = C_2 j_0\left(\frac{\gamma}{r}\right) - C_1 n_0\left(\frac{\gamma}{r}\right) \quad (54)$$

where

$$\gamma = \sqrt{2\alpha}$$

For large r , this expression for $x_0(r)$ tends to the asymptotic form given in equation (50), if in addition (54) is normalized by putting $C_1=1$. The constants C_1 and C_2 can also be expressed in terms of x_0 , dx_0/dr (see Section 3.2.2.1.) and this yields

$$\begin{aligned} C_1 &= \gamma \frac{dx_0(r)}{dr} n_0\left(\frac{\gamma}{r}\right) - \frac{\gamma^2}{r^2} x_0(r) n_1\left(\frac{\gamma}{r}\right) \\ C_2 &= \frac{dx_0(r)}{dr} j_0\left(\frac{\gamma}{r}\right) - \frac{\gamma}{r^2} x_0(r) j_1\left(\frac{\gamma}{r}\right) \end{aligned} \quad (55)$$

The wave function for very small incident velocity was thus found by integrating (35) (for $k=0$, and $\ell=0$) numerically until x_0 was of the form given by (54). This was checked by continuously monitoring the ratio C_2/C_1 as given by (55). Once the ratio converged, the calculation was terminated and the momentum-transfer cross-section was found using (51). The wave function was also normalized by setting $C_1=1$, in order that the appropriate Z_{eff} could be calculated.

3.3. Calculation of Z_{eff} .

As mentioned in Chapter 1, Section 1.3.1. the direct annihilation rate of positrons in a gas is proportional to the gas density, and to the electron density $|\psi^-|^2$ at the positron averaged over the positron position. This annihilation rate, is given by (Ferrell, 1956)

$$v_a = \pi r_o^2 c n_s \int d^3x |\psi^-|^2 |\psi^+|^2 \quad (56)$$

where r_o is the classical electron radius, and n_s is the gas density in atoms cm^{-3} , and c is the velocity of light.

Equation (56) may be rewritten in the form

$$v_a(k) = \pi r_o^2 c n_s Z_{\text{eff}}(k) \quad (57)$$

where Z_{eff} is the value of the integral in (56). For a plane wave representation of the positron wave ψ^+ the value of Z_{eff} is just the atomic number Z of the atom. When the positron-atom interaction is taken into account, ψ^+ must be found by solving the relevant Schrodinger equation (see Section 3.2.). In terms of the x_ℓ of Section 3.2., ψ^+ is given by (for $k \neq 0$)

$$\psi^+ = \sum_{\ell=0}^{\infty} \frac{x_\ell}{kr} P_\ell(\cos \theta) \quad (58)$$

where the F_ℓ are Legendre polynomials. Performing the integration over angles in the expression for Z_{eff} then shows that each partial wave contributes to the total Z_{eff} , the ℓ^{th} partial wave contribution being

$$(Z_{\text{eff}})_\ell = \int_0^\infty |\psi^-|^2 (2\ell+1) \frac{x_\ell^2(kr)}{k^2 r^2} dr \quad (59)$$

The electron density $|\psi^-|^2$ in all the calculations presented here

is that appropriate to the Argon atom in the ground state as calculated by Hartree and Hartree (1936).

The numerical calculation of $(Z_{\text{eff}})_k$ involves an overlap integral for which Simpson's rule for numerical integration was found sufficiently accurate. The integration was carried out to $r=7a_0$ at which point the electron density $|\psi^-|^2$ is less than 10^{-5} of its maximum value within the atom.

For $k=0$, the Z_{eff} was calculated from

$$Z_{\text{eff}} = \int_0^\infty |\psi^-|^2 \frac{x_0^2(r)}{r^2} dr \quad (60)$$

where $x_0(r)$ was calculated as indicated in Section 3.2.2.2.

3.4. The positron velocity distribution.

3.4.1. The modified Wilkins equation.

The differential equation describing the velocity distribution of positrons in a gas in the energy range where only elastic collisions can take place has been shown to be (Falk, 1965; Falk, Orth and Jones, 1965)

$$\begin{aligned} \frac{\partial f(v,t)}{\partial t} = & \frac{1}{v^2} \frac{\partial}{\partial v} \left[\left\{ \frac{a^2 v^2}{3v_d(v)} + \frac{\mu v^2 v_d(v) K T}{m} \right\} \frac{\partial f(v,t)}{\partial v} + \mu v^3 v_d(v) f(v,t) \right] \\ & - [v_a(v) + v_f(v)] f(v,t) \end{aligned} \quad (61)$$

where: $f(v,t)$ is the probability density in velocity space at time t ;

$a = eE/m$ is the acceleration of the positron due to the electric field;

e is the positron charge;

E is the applied electric field;

m is the positron mass;

$$v_d(v) = n_s \sigma_d(v) v;$$

σ_d is the momentum-transfer cross-section for positron-gas atom collisions;

n_s is the density of scattering atoms;

$$v_a(v) = \pi r_o^2 c n_s Z_{\text{eff}}(v); \text{ (see Equation 57)}$$

r_o is the classical electron radius;

c is the velocity of light;

$$v_f(v) = n_s \sigma_f(v) v;$$

$\sigma_f(v)$ is the cross-section for positronium formation;

$$\mu = m/M;$$

M is the mass of scattering atoms;

T is the temperature of the host gas in $^{\circ}\text{K}$;

K is Boltzmann's constant;

The velocity v is related to wave number k by $v = k\alpha c$, where α is the fine structure constant. The function $y(v, t) = v^2 f(v, t)$ is the velocity distribution of positrons per unit velocity interval. By analogy with the Maxwellian and Druyvesteyn distributions and from physical considerations, it is expected that the boundary conditions for $y(v, t)$ are

$$\begin{aligned} y(0, t) &= 0 \\ y(\infty, t) &= 0 \end{aligned} \tag{62}$$

for all physically meaningful momentum-transfer, annihilation and formation cross-sections.

3.4.2. General computer solution of the differential equation.

The equation (61) can be transformed into

$$\frac{\partial y(v,t)}{\partial t} = \frac{\partial}{\partial v} \left[\left\{ \frac{a^2}{3v_d} + \frac{\mu v_d KT}{m} \right\} \frac{\partial y(v,t)}{\partial v} + \left\{ \mu v v_d - \frac{2a^2}{3v_d v} - \frac{2\mu v_d KT}{mv} \right\} y(v,t) \right] - [v_a(v) + v_f(v)] y(v,t) \quad (63)$$

If some estimates can be made as to the initial positron velocity distribution $y(v,0)$, equation (63) can be solved by standard numerical techniques. The function

$$\lambda(t) = \int_0^\infty y(v,t) v_a(v) dv / \int_0^\infty y(v,t) dv \quad (64)$$

yields the velocity-averaged direct annihilation rate as a function of time and is the quantity which is compared with the experimentally-determined time-dependent direct annihilation rate. Preliminary results of this type of calculation have been reported (Falk, Orth and Jones, 1965). However, the difficulty in estimating a realistic $y(v,0)$ makes it advisable to examine the equilibrium solutions to (63) which have no implicit time dependence.

3.4.3. The case of no implicit time dependence.

Consider the case where the applied electric field is zero and where the positron annihilation rate is negligible. Then it is known that the initial velocity distribution of positrons will relax after a time t' to a Maxwellian distribution appropriate to the temperature T of the host gas. Since the annihilations are negligible it can be assumed that they do not appreciably affect the velocity distribution. Then the velocity-averaged annihilation rate at the time t' is given by $\lambda(t')$ using (64). Because the shape of the velocity distribution remains Maxwellian after t' ,

the average annihilation rate is a constant thereafter. The probability density of positrons, however, is dependent on time, and can be seen to decrease exponentially with a rate given by $\lambda(t')$.

It can be shown that neither the applied electric field nor the annihilation rate have to be negligible for the exponential behaviour to be retained. It is only necessary to require that after some time the velocity distribution has an explicit dependence on time. Thus the shape of the velocity distribution is unchanged for later times. Putting

$$y(v,t) = T(t)Y(v) \quad (65)$$

separates equation (63) into

$$\frac{d}{dv} \left[\left\{ \frac{a^2}{3v_d} + \frac{\mu v_d K T}{m} \right\} \frac{dY}{dv} + \left\{ \mu v v_d - \frac{2a^2}{3v_d v} - \frac{2\mu v_d K T}{m v} \right\} Y \right] = [v_a + v_f - \lambda] Y \quad (66)$$

and

$$\frac{dT}{dt} = -\lambda T \quad (67)$$

The solution to (65) and (67), $y(v,t) = Y(v)T(0)e^{-\lambda t}$ is clearly exponential in t . The value of λ is found by integration of (66) once with respect to v over all velocities. Then

$$\left[\left\{ \frac{a^2}{3v_d} + \frac{\mu v_d K T}{m} \right\} \frac{dY}{dv} + \left\{ \mu v v_d - \frac{2a^2}{3v_d v} - \frac{2\mu v_d K T}{m v} \right\} Y \right]_0^\infty = \int_0^\infty (v_a + v_f - \lambda) Y dv \quad (68)$$

For potentials of the type used here, the momentum-transfer cross-section tends to a constant value as the incident velocity tends to zero (Landau and Lifshitz, 1965; p.500). Furthermore, the probability density of positrons in velocity space, $f(v,t)$ must be finite for $v=0$. It follows that $Y(v)$ must tend to zero at

least as rapidly as v^2 near $v=0$.

For high velocities, it is expected the momentum-transfer cross-section remains finite. In addition $Y(v)$ must tend to zero more rapidly than v^{-3} at high velocities in order that the overall energy associated with the distribution $Y(v)$, remain finite.

Making use of the limits established above, it is clear that the left hand side of (68) vanishes, both at $v=0$ and $v=\infty$. Hence

$$\lambda = \int_0^\infty [v_a(v) + v_f(v)]Y(v)dv / \int_0^\infty Y(v)dv \quad (69)$$

Thus when the positron velocity distribution becomes "static", the resulting annihilation rate is a constant, and the positron population decays exponentially. This exponential decay will also be exhibited if the annihilation cross-section is proportional to $1/v$. The annihilation rate v_a is then independent of velocity, and from (64)

$$\lambda(t) = v_a \quad (70)$$

3.4.4. Solution of the time-independent equation.

For a given set of cross-sections, the solutions to (66) and (69) can be readily obtained using a digital computer.

Consider the integral of (66) from $v'=0$ to $v'=v$. Then

$$\left\{ \frac{a^2}{3v_d} + \frac{\mu v_d kT}{m} \right\} \frac{dY}{dv} + \left\{ \mu v v_d - \frac{2a^2}{3v_d v} - \frac{2\mu v_d kT}{mv} \right\} Y = \int_0^v (v_a + v_f - \lambda) Y dv \quad (71)$$

since the integral of the left hand side of (66) vanishes at $v=0$ (see Section 3.4.3.). If the annihilation rate is small compared with the average scattering rate, then its effect on the velocity

distribution will simply be a perturbation. In Argon this is a good approximation since the scattering rate is about 10^{12}sec^{-1} at 10 amagats for a scattering cross-section of the order of πa_0^2 while the annihilation rate is experimentally about $5 \times 10^6 \text{sec}^{-1}$ for the same density (see Table IV).

Thus a first order solution to Y is obtained by solving (71) with the right hand side set equal to zero. This yields

$$Y_0 = C v^2 \exp \left[- \int_0^v \mu v_d dv / \left(\frac{a^2}{3v_d} + \frac{\mu v_d K T}{m} \right) \right] \quad (72)$$

where C is determined by normalizing the integral of Y_0 over all velocities to unity. The first order solution to λ is given by

$$\lambda_0 = \int_0^\infty (v_a + v_f) Y dv \quad (73)$$

Substitution of Y_0 , λ_0 into the right hand side of (71) and solving yields the second approximation to Y .

$$Y_1(v) = Y_0(v) \left[\gamma + \int_0^v f_2(v') / Y_0(v') dv' \right] = \gamma Y_0(v) + \Delta Y_1(v) \quad (74)$$

where

$$f_2(v) = \int_0^v (v_a + v_f - \lambda_0) Y_0(v') dv' / \left(\frac{a^2}{3v_d} + \frac{\mu v_d K T}{m} \right) \quad (75)$$

Since the $\lim_{v \rightarrow 0} Y_1(v)/Y_0(v)$ might be different from unity the constant γ is included, and can be calculated by numerical integration of (74) over all velocities, and requiring that integrals of $Y_1(v)$ and $Y_0(v)$ both be normalized to unity. Thus any approximation Y_j is thus obtained in terms of Y_{j-1} using (74).

The iterative procedure outlined has been successfully used to solve equation (71). The initial estimate Y_0 is made using 8-point Gaussian integration, and subsequent integrations were made using Simpson's rule and linear interpolation where

necessary. In general, the procedure converged within five iterations for all cross-sections used.

The effect of the annihilation perturbation was to decrease the $Y(v)$ relative to the original $Y_0(v)$ for those velocities where the annihilation rates are large. Thus, for the monotonically decreasing annihilation rates considered, the velocity distribution was uniformly shifted to a higher velocity.

In addition, for each velocity distribution that was calculated, the average positron velocity $\bar{v}$ given by

$$\bar{v} = \int_0^{\infty} Y(v) v dv \quad (76)$$

was found.

3.5. Results.

Preliminary results based on the procedure of the preceding Sections have been reported (Jones, Falk and Orth, 1965; Jones and Orth, 1966 (a); Jones and Orth, 1966 (b)). In one case (Jones and Orth, 1966 (a)), however, the published values of the velocity-averaged direct annihilation rate, λ_a , are in error to the extent that elastic-scattering cross-sections, rather than momentum-transfer cross-sections were used in the velocity distribution calculations, where only the Y_0 were calculated.

3.5.1. Discussion of the potentials used.

Three representations of the positron-Argon interaction were studied. All exhibited a $1/R^4$ behaviour at large distances, but differed in the size and type of the cut-off employed and consequently in the detailed shape of the positron-Argon interaction

in the neighbourhood of the atom. The total interaction considered was the sum of the Hartree potential for Argon (Hartree and Hartree, 1936), V_H , and the empirical polarization potential, V_p .

Three types of V_p were studied:

- A. $V_p^A = -5.5/(r^2 + r_o^2)^2$; $r_o^2 = 2.5 a_o^2$,
- B. $V_p^B = -5.5/(r^2 + r_o^2)^2$; $r_o^2 = 0.62 a_o^2$,
- C. $V_p^C = -5.5r^{-4}(1-\exp(r/r_o)^8)$; $r_o^8 = 14 a_o^8$.

In each case, the numerator is just the electric polarizability of Argon in atomic units (Holtsmark, 1929). The form of the potentials V_p^A , V_p^B has been used in reproducing the observed Ramsauer effect (Kivel, 1959) in Argon, and is known in the literature as the Buckingham semi-empirical form (Labahn and Callaway, 1966) and sometimes as the Holtsmark potential (Massey, et al., 1966).

3.5.2. Comparison with experiment.

In V_p^A , $r_o^2 = 2.5 a_o^2$ was selected for the cut-off, since this was the value found to fit low-energy electron scattering in Argon (Kivel, 1959). A similar effective interaction was employed by Massey, et al. (1966) in calculations of the momentum-transfer cross-section and Z_{eff} for positrons in Argon, and the cut-off parameter r_o^2 chosen was also the value appropriate to electron scattering.

Figures 15 and 16 show respectively the velocity dependence of the Z_{eff} and momentum-transfer cross-sections resulting from the solution of the Schrodinger equation. The values obtained for

Figure 15. Theoretical results for Z_{eff} as a function of positron wave number k .

Figure 16. Theoretical results for the momentum-transfer cross-section for positrons in Argon as a function of k .

V_p^A are in agreement with those reported by Massey, et al. (1966). Agreement with results obtained independently is necessary in calculations of the type presented here, providing an important check on the numerical techniques employed.

The results of the velocity-distribution calculations (Section 3.4.) are presented in Figure 17. Comparison with the dependence of the annihilation rate on the applied electric field obtained experimentally shows that the calculated annihilation rate $\lambda(A)$ is far too small. Furthermore, the theoretical curve does not vary sufficiently rapidly with applied electric field.

In order to reproduce at least one aspect of the experimental results, the cut-off parameter r_0^2 in V_p^B was chosen to be $0.62 a_0^2$ so that the calculated annihilation rate at zero electric field would approximate the experimental result. Reduction of the cut-off parameter by such a large amount results in an increased positron attraction, and this is reflected in the greatly increased momentum-transfer cross-section (Figure 16). In addition, the resulting increase in the positron wave-function inside the atom is indicated directly by the larger Z_{eff} (Figure 15).

Although, by this means, the magnitude of the theoretical annihilation rate is made almost equal to the experimental value at zero field, the dependence of the annihilation rate on applied electric field is too weak at low electric fields (Figure 17). An important feature for the curve for V_p^B in Figure 17 is the "break" which occurs at about $70 \text{ V cm}^{-1} \text{ amagat}^{-1}$. For this value of E/P , the mean positron wave number as calculated from Equation

Figure 17. Comparison of theoretical and experimental annihilation rates as a function of E/P.

(76), Section 3.4.4. is $0.23 a_0^{-1}$. From Figure 16, this corresponds to a momentum-transfer cross-section of about $10 \pi a_0^2$. The significance of this cross-section at the break will be discussed shortly.

The potential described by V_p^C is also a one parameter potential, but follows more closely the form of the polarization potential expected for incident electrons from a physical argument (Lenander, 1966; Crown and Russek, 1965). This potential rises to a maximum near the surface of the atom and decreases rapidly inside the atom. For positrons, however, the effect of electron-positron correlations should result in an enhanced effective potential within the atom as compared with the case of incident electrons. This interaction may be expected to be more important for low velocity positrons, thus giving rise to a velocity-dependent effective potential. This type of velocity-dependent potential is not considered here, however.

The size of the cut-off parameter $r_0^8 = 14 a_0^8$ has been chosen so that the Z_{eff} at zero velocity coincides with the Z_{eff} at $k = 0 a_0^{-1}$ calculated using V_p^B (Figure 15). Again the momentum-transfer cross-section is very large at $k = 0 a_0^{-1}$ (Figure 16), and falls off rapidly to a fairly constant value. The dependence of annihilation rate on electric field (Figure 17) bears no resemblance to the experimental results. The position of the break mentioned earlier is less well defined for this case, but can be taken to be in the region of $120 V cm^{-1} amagat^{-1}$. The average positron wave number corresponding to this field as

calculated from equation (76), Section 3.4.4. is $0.23 a_0^{-1}$. At this wave number the momentum-transfer cross-section is once more of the order of $10 \pi a_0^2$.

3.5.3. Discussion of the break in the dependence of annihilation rate on electric field.

The size of the momentum-transfer cross-section at the break appears to be independent of the detailed shapes of the momentum-transfer cross-section and Z_{eff} , but depends simply on the monotonic decrease of both the momentum-transfer cross-section and Z_{eff} . This fact can be demonstrated with the aid of equation (66), Section 3.4.3.

At low electric fields, the form of the positron velocity distribution is essentially independent of the electric field due to the fact that the energy gain term, $a^2/3v_d$ is swamped by the elastic scattering term, $\mu v_d KT/m$. Once the electric field is large enough for $a^2/3v_d$ to be comparable to $\mu v_d KT/m$, however, a transition occurs where the shape of the velocity distribution becomes determined primarily by the value of the field term. Once the electric field is increased beyond this point, the average positron velocity increases more rapidly with electric field, provided that there is no accompanying increase in the momentum-transfer cross-section. It is evident that a transition of this type will lead to the break in the electric-field dependence of velocity-averaged annihilation rate, if the Z_{eff} is a monotonically decreasing function of velocity.

In order, then, that the velocity distribution be mainly determined by the electric field term, the momentum-transfer cross-section at the average wave number k appropriate to the velocity distribution must approximately satisfy

$$\sigma_d < \frac{E}{20P} \bar{k}^{-1} \pi a_0^2 \quad (77)$$

in a host gas at 298°K. When the magnitude of the electric field ($E/P = 70 \text{ V cm}^{-1} \text{ amagat}^{-1}$) and average wave number ($\bar{k} = 0.23 \text{ a}_0^{-1}$) appropriate to the break in curve B, Figure 17 are substituted into (77), the inequality is $\sigma_d < 15\pi a_0^2$. This is in agreement with the discussion in Section 3.5.2. where the value for σ_d at the average wave number corresponding to the break in curve B was found from a detailed calculation.

3.5.4. Discussion of the experimental dependence of annihilation rate on electric field.

It is possible to discuss the experimental results in Figure 17 in the light of the previous Section. Compared with the theoretical annihilation rates, the experimental values decrease rapidly at considerably smaller electric fields. Furthermore, there is evidence that the break starts to occur at E/P values less than $15 \text{ V cm}^{-1} \text{ amagat}^{-1}$. The average positron velocity at the break must be of the order of thermal velocities ($\bar{k} \sim 0.05 \text{ a}_0^{-1}$) or slightly larger. Now for $\bar{k} = 0.05 \text{ a}_0^{-1}$ and $E/P = 15 \text{ V cm}^{-1} \text{ amagat}^{-1}$, the expression (77) yields $\sigma_d < 15\pi a_0^2$, while larger $\bar{k}$ give rise to smaller upper limits for σ_d . Thus it is reasonable to suppose that the momentum-transfer cross-section for positrons in Argon

is less than about $15\pi a_0^2$ at thermal energies.

It is interesting to observe that the estimation of this upper limit for the low-energy momentum-transfer cross-section is independent of any assumptions regarding the nature of the positron-atom interaction, and is also independent of a possible contribution to the observed annihilation rate from radiative capture processes.

The momentum-transfer cross-section cannot rise appreciably at higher velocities in view of the increased positronium formation as a function of electric field (Figure 9; see also Figure 8). For example, the amount of increased positronium formation at $100 \text{ V cm}^{-1} \text{ amagat}^{-1}$ indicates that an appreciable fraction of the equilibrium distribution is at the threshold velocity for positronium formation; that is, at $k \simeq 0.8 a_0^{-1}$. Further evidence for this is to be found in the flattening off of the direct annihilation rate at high electric fields, which has been attributed to increased positronium formation (Chapter 2, Section 2.6.3.). For the electric field to dominate in this region (as indicated, in fact, by the rapid dependence of positronium formation on electric field in this region) requires that $\sigma_d < 5\pi a_0^2$.

3.6. Conclusions.

3.6.1. Summary of experimental results.

The direct component of positron annihilation in Argon has been measured as a function of density. The annihilation rate appears to obey a linear dependence up to about 10 amagats,

with some indication of a non-linearity at higher densities.

The orthopositronium quenching rate has also been determined as a function of density. Again the results are consistent with a linear dependence up to about 17 amagats. The direct annihilation rate and relative orthopositronium production were both measured as a function of applied electric field. These measurements have been used to provide an internally consistent picture of the behaviour of the positrons in Argon under the influence of an electric field.

3.6.2. Theoretical conclusions.

The electric field results have been used as a test of the current models of interactions of incident electrons and positrons with multi-electron atoms. It has been shown here that the simple one-parameter approximations for the effective atom-electron interaction, which have been found to yield a reasonable fit to the low-energy elastic scattering data, are inadequate for the case of positron-Argon scattering. The sharp initial drop in the observed direct annihilation rate as a function of applied electric field is not reproduced using the momentum-transfer cross-sections and annihilation rates derived using these potentials. Furthermore, from the general arguments proposed in Section 3.5.4., it appears that the momentum-transfer cross-section in Argon is less than $15 \pi a_0^2$ at thermal energies.

In order to reproduce such a small momentum-transfer cross-section, while requiring a relatively large annihilation

rate, it seems that certain effects neglected in the simple model considered here should be taken into account. These effects include the positron-electron correlation, especially in the neighbourhood of the atom, and deformation of the Argon atom by the scattering positron. The possibility of radiative capture of the positron from the continuum into a bound Argon-positron system should also be expected to contribute to the observed direct annihilation rate.

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APPENDIX.

MODIFICATIONS TO THE FAST-SLOW COINCIDENCE CIRCUITRY USED BY FALK (1965).

A block diagram of the fast-slow coincidence system is shown in Chapter 2, Figure 3.

1. Photomultiplier circuitry.

The new 4in. x 3in. diameter NaI(Tl) crystals required replacement of the 2in. diameter RCA 6810 photomultiplier tube used previously with the larger 4in. diameter RCA 7046. The modified circuit for the dynode chain supplying the dynode potentials is shown in Figure 18. Since fast risetimes were required at the anode the effect of inductance was minimized from stages 6 to 11 by use of heavy, short copper strip as leads. Elsewhere, leads were kept as short as possible. The photomultiplier high voltage was supplied by the commercial Precision Power Source, Model 122 B, manufactured by Calibration Standards Inc.

2. Timing Pulse Generator.

Both limiters used by Falk have been replaced with transistorized equivalents. The circuit is shown in Figure 19, and is similar to a type of circuit, employing tunnel diodes, that is used for fast coincidence work (Schwarzschild, 1963).

Operation of the circuit is as follows: the arrival of

an electron pulse at the photomultiplier collector (Figure 18) causes current to be conducted through the tunnel diode TD. Sufficient current through the TD causes the tunnel diode to trigger. The threshold current required for triggering is set by the $10k\Omega$ potentiometer. Once the TD has triggered, the voltage at the base of transistor T drops by about 0.2 V. The transistor is rapidly driven to saturation and a positive voltage pulse of about 2.5 V appears at the 100Ω resistor. Half of the 25 mA produced is driven through the 100Ω cable into the input of the timesorter. The maximum width of the output pulse is defined by the time taken for the TD to return to its original state. This time is set by the LR network across the TD.

The threshold level of the TD affects the resolution of the timesorter to some degree. For each gamma-ray channel, the threshold was set so as to minimize the full-width at half-maximum of the prompt peak obtained with Na-22 in aluminum. An advantage of this threshold method is that noise pulses do not trigger the input to the timesorter.

The prompt resolution obtained with these timing pulse generators is shown in Figure 20. Two curves are presented, one corresponding to those annihilation gamma rays which yield pulses within the 0.51 MeV full-energy peak. The second was obtained for the same energy interval but including only those pulses which occurred in the "valley" region (Chapter 1, Section 2.2.1.). The prompt resolution had been previously optimized for the first case. There is a small difference in the resolution

due mainly to time slewing, that is, dependence of the triggering time on the amplitude of the photomultiplier collector pulse.

3. Amplifiers and Single Channel Analyzers.

The amplifiers and single channel analyzers used by Falk have been replaced with commercial transistorized units (Cosmic Amplifier, Model 901A and Single Channel Analyzer, Model 901 S.C.A.). The amplifiers produce bipolar pulses which are fed into the zero cross-over single channel analyzers. Each single channel analyzer has both a negative and a positive output. The positive output was used to trigger the coincidence gate (Chapter 2, Figure 3) described by Falk. The negative pulses were counted by a scaler (Nuclear Supplies, Model SA-250). Each scaler was used to monitor the total number of counts admitted by the single channel analyzers during a run. The number of counts in the 1.28 MeV gamma channel was used for normalization from run to run. As the typical number of counts encountered in a run were of the order of $10^7 - 10^8$, and the scalers were capable of scaling only to 10^6 , a mechanical register of counting capacity 10^4 was connected to each scaler count overflow. The circuit used for this purpose is illustrated in Figure 21.

4. Pile-up Rejectors.

The use of the Cosmic amplifiers made it possible to feed the outputs of the amplifiers directly into the pile-up rejectors, described in Falk's thesis (1965), without further amplification. The current amplifier portion of the detectors

developed by Falk was therefore not used.

5. ND 101 Kicksorter.

The 100 channel kicksorter (Computing Devices of Canada, Model AEP 2230) previously used was replaced with a 256 channel analyzer (Nuclear Data, Model 101). The difference in the input requirements to the kicksorter required slight modification of the output stages of the timesorter which in no way affected the operation of the instrument.

6. Differential and integral linearity of the timesorter.

The differential and integral linearity of the overall system were obtained routinely as reported previously (Jones and Falk, 1965; Falk, Jones and Orth, 1965), and are shown in Figure 22. Two measurements of the integral linearity were made, separated by an interval of 2 months. Results of the first measurement are shown in Figure 22. A least squares fit to this straight line yielded an average slope of 2.74 ± 0.01 nsec/channel, while the second measurement yielded 2.73 ± 0.01 nsec/channel. The integral linearity measurement was used to define the average time width per channel (that is, the time calibration of the instrument), while the differential linearity was used as a measure of the actual individual widths. Both sets of data were used for the evaluation of the lifetimes (Chapter 2, Section 2.5.).

Figure 19. Timing pulse generator circuit.

Figure 20. Prompt resolution of the electronic system. The spectra were obtained for Na-22 in Al.

Figure 21. Circuit for driving electro-mechanical register.

Figure 22. Integral and differential linearity of the timesorter.