THE FIRST EXCITED STATE OF POSITRONIUM

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ABSTRACT

THE FIRST EXCITED STATE OF POSITRONIUM

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In order to measure the Lamb shift and other radiative corrections to the fine structure of positronium it is necessary to induce microwave transitions among the \( n = 2 \) group of states. The theoretical aspects of such an experiment have been investigated, including calculations of the Zeeman-Stark perturbed energy levels and their associated radiative widths. An experimental attempt to induce the 5 e.v. transition from the \( 1^3S_1 \) to the \( 2^3P_J \) states has been completed. A change in the counting rate of \((0.2 \pm 0.09)\%\) in the "valley" of the pulse height spectrum of the annihilation radiation during excitation is in agreement with estimates of the expected effect.

Thesis Supervisor: Martin Deutsch
Title: Professor of Physics
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PART I. ATOMIC STRUCTURE OF POSITRONIUM

1. Introduction

With the experimental discovery of the electron-positron atom (1), named positronium by Ruark (2) in 1945, an atomic system became available for the investigation of bound Dirac particles under the influence of pure electromagnetic forces. The system of two charged Dirac particles and their associated electromagnetic fields is described by the Bethe-Salpeter two-body relativistic equation, from which the discrete energy spectrum may be computed to any desired accuracy.

The gross energy structure of positronium is the same as that of an electron in the static Coulomb field of one positive electronic charge (i.e., hydrogen) save that the reduced mass of the electron $\mu = m/2$ yields a "positronium Bohr radius" $a = \frac{\hbar^2}{\mu e^2} = 1.06 \times 10^{-8}$ cm and thus a discrete energy spectrum $E_n = \frac{e^2}{2an^2} = 6.78 \ (1/n^2) \ eV$. The interparticle distance is twice that of hydrogen, and all the energy levels are reduced from the corresponding hydrogen levels by a factor of two.

Unlike hydrogen, however, the particle anti-particle nature of the electron and positron permit the dematerialization of the positronium atom, with the rest energy of the system transferred to the free radiation field. From the simultaneous conservation of energy and momentum it follows that the

References (in parentheses) are listed on page 90.
annihilation of a free positronium atom must occur with the emission of two or more quanta.

The fine structure interactions of positronium differ from hydrogen in that the "nuclear" magnetic moment is the positron moment (one Bohr magneton) which results in the spin-spin and the spin-other orbit energies being as large as the usual spin-orbit fine structure. The velocities of the electron and the positron are the same in the bound state so there is an orbit-orbit interaction which is again as large as the spin-orbit term. Because of the large effect of the "nuclear" (positron) spin the bound states are characterized as singlet or triplet. The possibility of transient or virtual annihilation and re-creation of the pair, with a single photon as the intermediate state, gives rise to a "virtual annihilation force" which must be spin dependent. This follows from the integral spin property of the Maxwell field so that, as angular momentum is conserved in intermediate states, the initial and final electron-positron pairs must have spin one: i.e., the triplet state.

The contributions of these interactions have been computed by Firenne (3), Berestetski (4), and Ferrell (5). Ferrell has the most recent error-free calculations of the fine structure, to order $mc^2 \alpha^4 \approx \alpha^2 \left( e^2 / 2a \right)$, with a discussion of earlier work.

Since momentum conservation requires, in the center of
mass coordinate system, that the two photons from the two-
quantum decay of positronium be emitted with equal energy in
opposite directions, and as the angular momentum associated
with this radiation field is zero, it is clear that this mode
of annihilation occurs only from the singlet state. Triplet
positronium can annihilate only by the emission of three or
more photons. Annihilation occurs only from $S$-states, in
which there is an overlap of the electron positron wave func-
tions. As parity is conserved during the annihilation process
there is no interference between the two decay modes.

An estimate of the lifetime of the $n^1S_0$ positronium states
may be made using the approximate cross section for two-quantum
annihilation in flight at low relative velocity, discussed in
(6): $\sigma = nr^2_0 c/v$ where $r_0 = e^2/mc^2$ is the "classical elec-
tron radius" and $v$ is the relative velocity. The mean annihi-
lation rate for singlet states $0.804 \times 10^{-10}/n^3 \text{ sec}^{-1}$ corre-
sponds to a mean life $\tau = n^3 \times 1.25 \times 10^{-10} \text{ sec}$. Quantum
electrodynamics yields the mean life for three-quantum decay
for $n S_1$ states: $\tau = n^3 \times 1.4 \times 10^{-7} \text{ sec}$. The probability
of occurrence of higher modes of decay (i.e., 4-quantum annihi-
lation) is much lower than for the two and three quantum pro-
cesses so that substantially all positronium annihilates by
one of these two modes.

Corrections to the $mc^2a^4$ fine structure terms arise from
a more complete treatment of the retarded interactions between
the positron and the electron, from the Lamb shift, and from
the polarization of the vacuum. In addition, there is a term arising from the virtual two-quantum annihilation of the \( n^1S_0 \) states yielding an "annihilation force" smaller than that previously considered. Many of these terms are angular momentum dependent and thus do not contribute to the ground state level shifts.

Weinstein (13) has completed an accurate experimental measurement of the ground state splitting (1 \( 3S_1 \) - 1 \( 1S_0 \) energy difference) and theory and experiment are in excellent agreement. About half of the observed splitting (which is about 2 x \( 10^5 \) \( M_0/\text{sec} \)) arises from the non-classical spin-spin interaction, the remainder from the triplet virtual annihilation force. The contribution of higher order corrections (\( mc^2\alpha^5 \)) was quantitatively determined by this measurement.

Fulton and Martin (14) have calculated the \( mc^2\alpha^5 \) corrections to the \( n = 2 \) group of states using the methods of Schwinger. It is desirable to check these results experimentally in view of the additional terms which do not enter into the ground state splitting. In this present paper theory and experiments preliminary to actual fine structure determinations are discussed.

In order to measure one or more of the level splittings of the \( n = 2 \) group of states, it is first necessary to induce the (Lyman) transition to one of these states with a reasonably high probability, in view of small numbers of positronium atoms available. (During the experiments reported here an observ-
able atom was present in the apparatus \( \sim 1/10,000 \) of the time.)

Part I summarizes the theory as discussed in Ferrell (5) which is pertinent to this excitation experiment and to a contemplated fine structure determination. Radiation theory is applied to the excitation and de-excitation processes to allow estimates of expected effect of the excitation experiment.

Parts II, III, and IV discuss the structure and conclusions of the excitation experiment.

2. Zeeman Effect

For the purpose of experimental measurements positronium is usually formed in a magnetic field. The field allows the formation to occur along a collimated beam, allows the "sweeping" of a microwave transition, and is responsible for the singlet-triplet mixing which yields observable changes in the two- to three-quantum annihilation ratio following induced transitions of almost any kind. The Zeeman and Stark effects as discussed in Ferrell (5) are summarized in this and the following section. More general derivations of the matrix elements are displayed in the Appendices, from which elements machine computation has yielded energy level diagrams.

Because of the unique situation in positronium of opposite charges of equal mass which for \( L \neq 0 \) revolve at equal distance from their center of mass, there is no orbital magnetic moment in any state of the atom. Thus, there is no
coupling of an external magnetic field with the orbital angular momentum. The Zeeman interaction energy must therefore involve only the spin coordinates of the atom (4). The perturbing operator in the Hamiltonian is

\[ H_z = \frac{e}{2m} \vec{\mathcal{H}} \cdot (\vec{\sigma}_1 - \vec{\sigma}_2) \]

where \( \vec{\sigma}_1, \vec{\sigma}_2 \) are the Pauli spin matrix vectors of the positron and electron. This is an odd operator which converts singlets to triplets and vice versa. The operator therefore has no diagonal entries between unperturbed states and there is no first-order Zeeman effect in positronium. \( H_z \) does not commute with \( S \) but, as it is independent of spatial coordinates, it commutes with \( L \). It does not commute with \( \vec{J} = \vec{L} + \vec{S} \).

If \( \vec{\mathcal{H}} = \mathcal{H} \vec{z}_o \), then \( H_z = (e/2m) \mathcal{H} (\vec{\sigma}_1 - \vec{\sigma}_2) \) and \( H_z \) commutes with \( S_z L_z J_z \). The only non-zero entries in the matrix representation of this operator will be those between states of the same \( L \) and \( J_z \) and opposite spin.

For example, in the \( S \) states of positronium (the \( n^1S_0 \) and \( n^3S_1 \)) the triplet states of \( J_z = M = \pm 1 \) will be unperturbed. \( H_z \) will connect \( n^1S_0 \) and \( n^3S_1 \) (\( M = 0 \)). Because of the orthogonality of the spatial functions, \( \mathcal{H} \) will not couple these to any other states. By solving the resulting 2 x 2 secular equation, we find that the singlet and triplet states "repel," and that for small \( \mathcal{H} \), the effect is quadratic in the field (This, it should be noted, is the effect of the linear Zeeman operator calculated in second order, and is not properly
called the quadratic Zeeman effect, a term reserved for the first-order calculation of the operator \( [(e/c)A]^2 \) which contains the field variable quadratically.

The selection rules for non-vanishing entries in the matrix representation of \( H_z \) are \( n = n', L = L', M = M', S \neq S' \). It is interesting to observe that the largest submatrix of the semi-infinite \( H_z \) matrix is \( 4 \times 4 \) and that, in a representation in which \( H_0 \) (the unperturbed Hamiltonian) is diagonal, the diagonalization of any submatrix of \( H = H_0 + H_z \) yields an exact solution of the Zeeman effect, and avoids the use of perturbation theory in finding the new eigenvalues.

In the notation of Dirac (7), the non-zero matrix elements of \( H_z \) are \( (e \frac{\mathcal{H}}{2 m} ) \langle S \ L \ J \ M \mid \sigma_{z_1} - \sigma_{z_2} \mid S' \ L \ J' \ M \rangle \) and the explicit form of these is calculated in Appendix I. The result is

\[
\frac{e \frac{\mathcal{H}}{2 m} }{2 m} \langle S \ L \ J \ M \mid \sigma_{z_1} - \sigma_{z_2} \mid S' \ L \ J' \ M \rangle = \frac{e \frac{\mathcal{H}}{m} }{m} \langle J'M \mid 1 \ L \ O \ M \rangle
\]

where we have set \( S = 0 \) implying \( S' = 1 \), and \( \langle J'M \mid j_1 j_2 m_1 m_2 \rangle \) is the Clebsch-Gordan coefficient, as defined by Condon and Shortley (8) or Blatt and Weisskopf (9). This is the matrix entry connecting a triplet state \( J' M L \) with the singlet state \( L M \).

From this expression we note the vanishing of the matrix entry connecting \( ^3P_1 \ (M = 0) \) and \( ^1P_1 \ (M = 0) \), for the Clebsch-Gordon coefficient in this case becomes \( \langle 10 \mid 1100 \rangle \), which is
zero as a consequence of the general rule that \( (a0 | bc00) = 0 \) if \( a + b + c \) is odd. There is thus no restriction against the crossing of these levels, as they do in the \( n = 2 \) states. For a non-zero element the levels could not cross by the Wigner-von Neumann theorem. This physically unexplained crossing occurs also in hydrogen (10).

The \( 2 \times 2 \) and \( 3 \times 3 \) secular determinants which arise from the submatrices within the \( n = 1 \) and \( n = 2 \) groups of states, respectively, can be easily solved numerically. The eigenvalues reflect the quadratic dependence on \( H \) for low fields characteristic of second-order perturbation theory, but become linear in \( H \) for large field strengths when Back-Goudsmit spin decoupling is essentially complete.

The pure Zeeman pattern for \( H < 5000 \) gauss has been evaluated by machining computation for the \( n = 2 \) group of states. The calculations are discussed in Appendix V.

3. **Stark Effect**

The orbital interaction of positronium with an external static magnetic field vanishes identically when the atom is at rest. An expansion of this interaction, with a canonical transformation to relative and center-of-mass coordinates has a residual term which couples these coordinates when the atomic center of mass has a momentum \( \vec{P} \). This is discussed in Ferrell (5) where it is shown that this energy is of the form
\[ H_s = \frac{e}{2mc} \vec{r} \cdot (\vec{F} \times \vec{H}) = e\vec{r} \cdot \vec{E}_t \]

where \( \vec{r} \) is the relative coordinate and \( \vec{E}_t \) is the transformed magnetic field appearing as an electric field in the frame of the positronium atom.

\[ \vec{E}_t \approx \frac{\vec{v}}{c} \times \vec{H} \] correctly describes the transformed field to order \( v/c \) where \( \vec{v} \) is the atomic velocity (cf. Heitler (11), p. 12).

The operator \( e\vec{r} \cdot \vec{E}_t \) is the usual Stark effect operator and is discussed in Schiff (12). This is an odd operator and thus has no expectation value between unperturbed positronium states. Similar to the Zeeman effect, the level shifts are quadratic in \( \vec{E}_t \) (and thus \( \vec{H} \)) for small fields. The selection rules for the non-vanishing of the entries in the matrix representation of \( H_s \) are \( \Delta L = 1, \Delta S = 0, \Delta M = \pm 1 \), no rule on \( \Delta n \).

The importance of considering this operator arises from the presumably high probability that positronium formation in gases leaves the positronium atoms with velocities somewhat higher than correspond to thermal energies. An upper bound for the mean translational energy is about 1 eV. From line broadening data in the ground state level splitting experiment, Weinstein (13) has estimated this energy to be 0.5 eV with an uncertainty of 0.5 eV. For a magnetic field of 2000 gauss, assuming thermal velocity perpendicular to the field, the magnitude of the off-diagonal elements of \( H_s \) for
n = 2 states, amounts to about 1000 Mc/sec or about an order of magnitude smaller than the Zeeman entries. For an energy of 1 ev the Stark and Zeeman entries are of the same order of magnitude. In any contemplated microwave transition between levels of n = 2, the Stark effect represents a line broadening of a rather serious nature rather than a level shift. This follows from the random orientation of the atomic momentum vector with respect to the external field plus the broadening due to the distribution of $|\vec{P}|^2$. A microwave transition experiment (such as was performed for n = 1) which involves a transition between Zeeman split levels of n = 2 which are degenerate at zero field is impossible if the positronium energy is near 1 ev as the line broadening is of the same magnitude as the transition energy. This effect is of no importance for n = 1 transitions as the nearest levels of opposite parity are several volts away and the Stark mixing (considering the energy denominators) is extremely small.

The matrix representation of $H_s$ does not yield an exact solution of the Stark effect upon the n = 2 group of levels following diagonalization of the n = 2 submatrix of $H_o + H_s$. In this case there is no selection rule $n = n'$ as exists for the Zeeman effect. The effect of mixing in levels of different n is unimportant, however, and diagonalization of the n = 2 submatrix represents an excellent approximation to the Stark effect.

In practice, the Zeeman effect must occur if Stark effect
is present. In the numerical diagonalization procedure applied to $H = H_0 + H_z + H_\text{g}$, it was possible to find both eigenvalues and eigenvectors of this operator for a range of the independent parameters $\vec{v}$ and $\vec{J}$, in terms of the eigenfunctions of $H_0$ (these of a representation $n L S J M$), using as the diagonal entries of $H$ the eigenvalues of $H_0$ correct to order $m c^2 a^5$ as calculated by Fulton and Martin (14). Although there are sixteen $n = 2$ states, the $^3P_2 (M = \pm 2)$ have no off-diagonal elements so that the smallest submatrix (for $H_\text{g} \neq 0$) is $14 \times 14$.

The matrix elements of $H_\text{g}$ for $n = 2$ are

\[
(n L S J M | H_\text{g} | n' L' S' J' M') = (n L S J M | H_\text{g} | OS'S'M')
\]

\[
= (J, M | Slm; M - M') (-1)^{J - S - L} \delta_{SS'} \frac{BG}{\sqrt{4\pi}} (\delta_{M - M'} l - \delta_{M - M'} 1)
\]

where $G = \frac{e^2}{r^2} \sqrt{\frac{2m}{3}}$

$(J, M | Slm; M - M')$ is a Clebsch-Gordan coefficient,

$R$ is the radial matrix element and we have specialized to $n = 2$ in setting $L' = 0$, $J' = S'$, from which $L = 1$. The derivation of this expression is in Appendix II.

4. Excitation of the $n = 2$ States

In order to induce the $1^3S_1 \rightarrow 2^3P_J$ transition in positronium (the first Lyman line) it is necessary to supply an energy of 5.06 ev (corresponding to a light quantum of wave-
length 2430.20 A.u.) to the atom in its rest reference frame. In principle there are several methods of communicating this energy to the atom: most of them, for one reason or another, are unsuited to experimental application.

Excitation by electron impact requires a high current density (from an electron gun or the plasma of a gas discharge) to achieve a reasonable transition rate but has been used with success for the equivalent transition (although to a $2S_{\frac{1}{2}}$ state) in hydrogen (29). In positronium the probability of electron exchange with a triplet-singlet transition would lead to almost complete quenching of three-quantum decay plus an increased annihilation-in-flight rate. Even if this were not the case the optimum electron bombarding energy would be expected to be several volts above threshold (in laboratory coordinates) and the average kinetic energy transmitted to the excited atom would result in an excessive Stark effect.

The low currents available from present sources of charged particles other than electrons entirely remove the possibility of excitation by protons, etc.

Although excitation by electron impact would be expected to populate all the triplet $n = 2$ levels the use of electromagnetic excitation would induce only the $1\ \hat{S}_{\frac{1}{2}} \rightarrow 2\ \hat{P}_{J}$ (El) transitions with reasonable probability. For contemplated microwave transitions between the $n = 2$ levels this is an ad-

*Correct to 0.02 A.U. as calculated from level shifts to order $\hbar c^2\alpha^2$ from Ferrell (5).
vantage for it leaves the $2^3S_1$ and $2^1S_0$ levels unpopulated and makes the detection of such microwave transitions much easier, as discussed in (II, 3).

For a positronium atom at rest the induced transition rate in the presence of an e.m. field can be calculated from the expectation value of the electric dipole operator between the states in question (12). With a continuous e.m. spectrum of $W$ (watts/cm$^2$/A.U.) at the Lyman absorption wavelength (2430.2 A.U.) of positronium, assuming irradiation for a time $\tau$ equal to the mean life of the $1^3S_1$ states ($1.4 \times 10^{-7}$ sec) a unit excitation probability is found for $W = 144$ (watts/cm$^2$/A.U.). It should be pointed out that this result neglects induced emission from the $n = 2$ state, which would lead, for $W$ very large, to a maximum excitation probability of $\frac{1}{2}$. The result is useful, however, for in the case of irradiation of intensity $w \ll W$, the quotient $w/W$ is the excitation probability for the time $\tau = 1.4 \times 10^{-7}$ sec.

The El transition operator yields matrix elements from an S state to a state of definite $M_L$. In the fine structure states of positronium $S L J M$ are good quantum numbers; $M_L$ is not. In the absence of perturbing external fields the states are degenerate with respect to $M$ and it is not apparent therefore, due to possible interference effects, that the transition probabilities to each final $M$ state are equal, and that the transition probabilities from each initial state are equal.
These facts are proved in Appendix III together with an evaluation of the square of the transition matrix element.

If a continuous spectrum is to be used for excitation it would, for a typical high pressure arc, extend over several thousand Angstroms. A rough estimate of the power requirements for an excitation probability of 0.1 would be
\[ P = 14 \times 3000 \approx 40,000 \text{ watts/cm}^2 \] which is unreasonably high.

Excitation by a single spectral line requires very much lower power, provided the wavelength is sufficiently close to the absorption wavelength. A search of the emission spectra of the elements (30) shows that tin yields the most suitable line, with a wavelength of 2429.5 A.U., 0.7 A.U. below the Lyman resonance in positronium, and this line was therefore chosen to provide exciting radiation for the present experiment. If there is to be an excitation by such means it is clear that either the tin emission line or the Lyman absorption line must be sufficiently broad so that some overlap exists. A simple calculation shows that the natural widths of either line or collision broadening of the tin line cannot provide the necessary overlap.

Doppler broadening of the absorption line can provide sufficient overlap, and there is experimental expectation that the atomic velocity of the positronium after formation is large enough to supply the required broadening. To calculate the excitation probability requires a modification of the usual theory which deals with a sharp absorption line and a broad ex-
citing line. The present case is the inverse of this.

The excitation probability $P$ for a narrow absorption line of resonant wavelength $\lambda_o$, irradiated by light of intensity distribution $I(\lambda)$ \((\text{watts/cm}^2/\text{A.U.})\) for a time $\tau$ is

$$P = a \cdot I(\lambda_o) \tau$$  \hspace{1cm} (1)

where $a$ is a constant depending on the details of the transition. Cf. Schiff (12).

If now the absorption line is broad, with a distribution $D(\lambda)$ per unit wavelength and $X$ watts/cm$^2$ is the power flow such that $I(\lambda)$ has the form

$$I(\lambda) = \frac{X}{\Delta \lambda} \quad \text{for} \quad \lambda' < \lambda < \lambda' + \Delta \lambda$$

$$= 0 \quad \quad \quad \quad \quad |\lambda - \lambda'| > \Delta \lambda$$

and $D(\lambda')$ is approximately constant over $\Delta \lambda$, then $D(\lambda') \Delta \lambda$ is the probability of finding the atom with a Doppler shifted absorption wavelength $\lambda'$ in range $\Delta \lambda$.

Equation (1) becomes

$$P = a \tau D(\lambda') \Delta \lambda \frac{X}{\Delta \lambda} = aD(\lambda') X \tau$$  \hspace{1cm} (2)

The above treatment does not depend on the exact width of the exciting line, provided it is small. One can ask whether the result is indeed independent of $\Delta \lambda$, i.e., is the treatment valid in the limit $\Delta \lambda \to 0$?

If the positronium were observed in a vacuum then the Doppler broadened absorption line would consist of an aver-
age over an ensemble of atoms, each atom having a discrete absorption wavelength \( \lambda \). Only those having absorption wavelength in range \( \Delta \lambda \) about \( \lambda \) would be excited. For these the excitation probability is, from Eq. (1):

\[
P^\prime = a \frac{X}{\Delta \lambda} \cdot \tau \tag{3}
\]

The excitation probability as given by Eq. (3) is the probability that the atom has been excited during the time \( \tau \). The excited state has a mean life \( t \ll \tau \) for spontaneous radiation \( (t \simeq 10^{-9} \text{ sec} \) for the \( 2^3 P_j \) levels) so if induced emission is not to compete with induced excitation (i.e., occupation probability for the \( \sum_j 2^3 P_j \) states very small) then the excitation probability during this time \( t \) must be small compared with unity:

\[
P^\prime = a \frac{X}{\Delta \lambda} \cdot t \ll 1 \tag{4}
\]

It was stated above that

\[
a \times 144 \text{ (watts/cm}^2/\text{A.U.}) \cdot \tau \simeq 1 \tag{5}
\]

and from Eq. (4) and (5) we find the condition on

\[
\Delta \lambda \gg \frac{X}{144} \frac{t}{\tau} \text{ (Angstroms)} \tag{6}
\]

and experimentally \( X \) is about 6 watts/cm\(^2\) so

\[
\Delta \lambda \gg \frac{6}{144} \frac{10^{-9}}{10^{-7}} \simeq 4 \times 10^{-4} \text{ (A.U.)} \tag{7}
\]

Measurements of the width of the tin line from the elec-
trodeless lamps at 2429.5 A.U. show that it does indeed satisfy this criteria, (III,4), but it is possible to operate a low temperature, low pressure discharge such that (7) is not satisfied.

If the positronium is formed and observed in a gas the collision rate \((\text{cm}^{-2} \text{sec}^{-1})\) may be large compared with \(t\). The rate of scattering in momentum space then allows each atom to "sweep" its own absorption line; it is no longer necessary to consider an average over an ensemble of atoms. Each atom spends, on the average, a time \(D(\lambda')\Delta\tau (\ll \tau)\) in the wavelength region for excitation, but it does this in many smaller intervals \(l/C \ll t\). For this case condition (6) is too stringent and may be replaced by

\[
\Delta\lambda \gg \frac{X}{144} \cdot \frac{1}{C\tau} \quad \text{(A.U.)} \tag{8}
\]

For \(X \approx 6 \text{ watts/cm}^2\) (8) is expected to be satisfied for pressures of the order of \(1/100\) atmospheres; no serious limitation.

If the positronium absorption resonance occurs at a circular frequency \(\omega_o\), then the Doppler broadened line has the shape (cf. Heitler (11))

\[
D(\omega) = \frac{a}{\pi} \frac{1}{\omega_o^2} \left(\omega - \omega_o\right)^2 \tag{9}
\]

where \(a\) depends on the positronium center-of-mass energy \(E\)

\[
a^2 = \frac{M_0}{E} \frac{\omega_o^2}{\omega_o^2} E \tag{10}
\]

and we wish to evaluate Eq. (9) at the exciting frequency
\[ \omega' = \omega_0. \] Letting \( W = \omega_0 - \omega' \) be a constant, (9) becomes

\[
D(\omega') = \sqrt{\frac{M_0^2}{2 \pi E}} \cdot \frac{1}{\omega_0} \cdot e^{-\frac{W^2 M_0^2}{2 \omega_0^2 E}}
\]

(11)

which has a maximum for a mean energy \( E_m \)

\[
E_m = M_0^2 \frac{(\omega_0 - \omega')^2}{\omega_0^2}
\]

(12)

\[ = 0.08 \text{ e.volts} \]

but this is due only to the component of velocity parallel or antiparallel to the radiation field propagation, i.e.,

\[ E_m = \frac{P_x^2}{2 m} \] if the light propagates in the x-direction.

The mean energy \( \overline{E} = 3 E_m = 0.24 \text{ e.v.} \), which, by a fortunate circumstance, is very close to the estimated energy (13).

Evaluating Eq. (11), Using Eq. (12), and substituting into Eq. (2), using Eq. (5), the probability of excitation during time \( \tau = 1.4 \times 10^{-7} \text{ sec} \) is

\[
P = \frac{X}{144} \cdot \frac{0.24}{\Delta \lambda}
\]

(13)

where \( \Delta \lambda = 2 \pi c \frac{W}{\omega_0^2} = 0.7 \text{ A.U.} \)

which for \( X \approx 6 \text{ watts/cm}^2 \) gives \( P \approx 0.015. \)

5. De-excitation of the \( n = 2 \) States

Of the modes of de-excitation of the 2P states the most important is the spontaneous radiation of the first line of the positronium Lyman series. These states have a mean life \( \tau = 3.2 \times 10^{-9} \text{ sec} \), corresponding to a level width of 49.5 Mc/sec.
As there is no annihilation from P states the optical decay rate is the same for singlet and triplet. There is no effect also from the LS coupling, as the transition is spin independent so the decay rate is independent of J and M.

In the presence of a magnetic field the Zeeman perturbed states are singlet-triplet mixtures of good orbital angular momentum. A perturbed 2P state will be a mixture of singlet P and two triplet states of different J. The 1P and 3P components decay to different ground states so the radiations are necessarily incoherent, but the 3P decay to the same state. The possibility of interference between these states would lead to an alteration in the transition probability for the perturbed state. (By interference is meant the contribution to the square of the transition matrix element of the perturbed states, from non-zero cross-product terms of the unperturbed states.) It can be argued from a physical standpoint that as the transition is spin independent, mixing 3P states of different J should lead to zero cross-terms as the spatial wave functions are unaltered. The spin functions are not orthogonal, however, so it is not obvious that the physical argument is correct and that they do vanish. The proof of this is in Appendix IV. The general result is: the radiation width for optical decay of any perturbed P state is the same as that of all the components and is independent of the degree of mixing.

The Stark effect mixes S and P states - components which
decay to different states and thus do not interfere. The radiation width for optical decay of a Stark perturbed state depends linearly on the amount of admixed P state. Annihilation occurs directly from the S states and the annihilation processes are entirely incoherent. It is therefore possible to calculate the branching ratios for various modes of decay for a Zeeman-Stark perturbed state by the following method.

If the unperturbed states $\psi_n$, $n = 1, \ldots, N$ have each associated a radiation width $\gamma_n$ corresponding to the $i$th mode of decay, then the perturbed states $\phi_m$ may be written

$$\phi_m = \sum_n a_{mn} \psi_n$$

where $a_{mn}$ ($m, n = 1, \ldots, N$) are the eigenvectors. The partial width $\Gamma_m^j$ for decay into the $j$th channel is then

$$\Gamma_m^j = \sum_n \gamma_n^j \left| a_{mn} \right|^2$$

where the sum is over the squares of $a_{mn}$ summed over $n$ (no cross product terms). The total width $\Gamma_m$ is simply

$$\Gamma_m = \sum_j \Gamma_m^j$$

and the equations represent entirely incoherent processes as they should.

It is possible to calculate the branching ratios for 3-quantum and 2-quantum decay of a positronium atom initially in some perturbed $n = 2$ state by summing all the singlet partial
widths and, separately, the triplet partial widths. The sum of the singlet widths contain the components annihilating directly from the 2'So and also the components which annihilate from the 1'So following optical emission and, similarly, for the triplet widths. These sums have been calculated numerically by Whirlwind computer for all the perturbed states for which energy eigenvalues were found. The information is particularly useful if microwave transitions are to be induced between n = 2 levels for it allows direct calculation of the 3-quantum quenching expected following the transition.

As in hydrogen the 2S states can decay by optical emission of two quanta. The predominant term arises from the second-order calculation of the effect of the operator $\frac{1}{\hbar} \vec{A} \cdot \vec{P}$ rather than from the first order calculation of $\frac{1}{\hbar^2} (A)^2$. This has been evaluated by Breit and Teller (34) for hydrogen; for positronium the mean life of the 2S states for this transition is about 0.3 second, which is long compared with the $10^{-6}$ second lifetime of the $3S_1$ states for annihilation.

The 2P $\rightarrow$ 1S transition occurs spontaneously as a consequence of the zero point oscillations of the Maxwell field coupled to the charge currents in the positronium atom of Heitler (11). An external field can induce the transition – in particular, the transient fields seen by the positronium during a collision with a gas atom. The probability of such collision induced de-excitation is expected to be negligible,
for the field seen by the positronium which causes such de-excitation is the field with a frequency component equal to that radiated. The quanta (real or virtual) which induce the transition have an energy equal to the transition energy. A Fourier analysis of the transient fields at the positronium during collision, in analogy to the Weisacker-Williams method, will not yield many virtual quanta of 5.06 e.v. unless the energy of relative motion of the two atoms is of order 5 e.v. or more; experimentally the energy is expected to be only 1/10 of this.

At lower gas pressures, where the positronium collision rate is not over an order of magnitude greater than the spontaneous optical emission rate, it is expected that collision-induced de-excitation will be effectively zero.

Collision-induced transitions between the n = 2 group of states does increase the width of these states. Such transitions, which would introduce discontinuous changes in phase of the emitted Lyman line, with resultant broadening, are the mechanism of the familiar collision broadening, which increases the width of an emission line by roughly 10 Mc/sec per millimeter (Hg) pressure.

These induced transitions are important in connection with determinations of the n = 2 fine structure and are discussed in (II, 4).

* Reference to Part II, Section 4 of this paper.
Energy Level Diagram #1

Pure Zeeman Pattern

$E_{\text{eff}} = 0$
Energy Level Diagram #2

Zeeman - Stark Pattern

$E_{\text{eff}} = 0.025$ e. v.
Energy Level Diagram #3

Zeeman - Stark Pattern

$E_{\text{eff}} = 0.25$ e. v.
Energy Level Diagram #4

Zeeman - Stark Pattern

$E_{\text{eff}} = 0.50$ e.v.
PART II. THE EXPERIMENT—(THEORETICAL ASPECTS).

1. Introduction

In an experimental attempt to induce the Lyman transition in positronium in such a manner that a subsequent modification will allow direct measurements of one or more of the $n = 2$ level splittings it is necessary to consider carefully the formation of positronium in gases and estimate the effect of the many mechanisms which influence the detection of induced excitation.

In Section 1 of this part, positronium formation is discussed and it is shown that either the choice of gas or a static electric field will substantially prevent re-acceleration of positrons whose energy is below the threshold for positronium formation. This condition must be assured to prevent the observed counting rates from depending on leakage r.f. fields from the lamp excitation system. Collision-induced transitions are also discussed in regard to spin and angular momentum changes.

In Section 2 the methods of positronium excitation are reviewed, and the experimental choice of microwave excitation of tin justified, with a discussion of pulsed operation and the limitations this avoids.

Section 3 considers the detection of any induced excitation effect through the mechanism of Zeeman spin mixing and resultant 3-quantum annihilation quenching.
Section 4 discusses the theoretical basis for measurements of the n = 2 fine structure, with particular regard to the higher order angular momentum dependent level shifts. The effect of collision-induced transitions, radiative and Stark widths of perturbed levels and their effects on the detection of several possible r.f.-induced transitions are treated, and the influence these have had on the present experiment.

2. Formation of Positronium in Gases

A quantitative understanding of the formation of positronium through the slowing-down and stopping of positrons in gases is not yet available but the known qualitative picture is of interest in the present investigation. Three-fourths of the positrons can form the long-lived triplet bound state (from the statistical factor) and it is thought that this figure is closely approached under some conditions. The slowing-down of positrons occurs primarily by inelastic collision, ionizing or exciting the gas atoms. In a noble gas, inelastic collisions are impossible when the positron energy falls below the energy necessary to excite the first excited state of the gas, and energy loss is then by elastic thermalizing collisions. Electron capture into the bound state by the positron is still possible in this region. When the positron energy falls below the gas ionization energy (less the positronium binding energy), positronium formation is energetically impossible and annihilation occurs with bound electrons. If a static or time-varying electric field is present, these positrons may be re-
accelerated above the threshold for positronium formation. Such a situation could occur in the present experiment due to leakage of the intense r.f. fields used to excite the discharge lamp. If the rate of positronium formation depended on the field present, detection of the excited state would be difficult.

To make the rate of positronium formation independent of the leakage r.f. field, a gas can be used which prevents re-acceleration, or a d.c. re-accelerating field can be applied which saturates the positronium formation. Such a gas is propylene, which has the high density of low-lying states characteristic of many molecules. Inelastic collisions are possible at all positron energy: 10 percent of propylene in 1 atmosphere of argon almost entirely inhibits d.c. re-acceleration (15). R.f. fields are somewhat less effective than d.c. in re-acceleration, when the oscillation amplitude of the positrons is smaller than their mean free path, a condition which prevails at microwave frequencies and gas pressures below one atmosphere. In argon, d.c. fields greater than 500 volts/cm re-accelerate virtually all the positrons whose energies fall below the threshold for positronium formation (6). Largely due to the increased stopping power, there is more positronium formed at 1 atmosphere pressure in propylene than at 1 atmosphere in argon with complete positron re-acceleration (15).

Singlet-triplet transitions can occur during a collision
with a gas atom having an internal magnetic field arising from a paramagnetic moment. An estimate shows this effect to be small so that it need not be considered in a choice of gas.

Electron exchange collisions resulting in singlet-triplet transitions are possible if the new spin state of the atom differs in energy from the initial state by an amount small compared to thermal energy. This occurs in the electronegative halogens, in nitric oxide, and to some extent in oxygen. Small quantities of these gases partially or entirely inhibit triplet positronium formation and are used mainly to determine background counting rates.

If the positronium is in an excited state, there is a high probability that the orbital angular momentum will change following a collision with a gas molecule (17). This is not wholly a quantum effect (it occurs classically in the collision of a rigid rotor and an infinite wall) and leads to the equal population of all orbital angular momentum states of approximately equal energy (compared with thermal energy). The effects of such collision are discussed in (II, 4).

3. Sources of 2430 Å Radiation

The tin spectrum yields a line at 2429.5 Å (0.7 A.u. below the Lyman absorption of positronium) from a transition $6s5p (J = 3, \text{ odd}) \rightarrow (5p)^2 3P_2$. This is not a resonance line (the ground state is $3P_0$); therefore self-absorption is ex-
pected to be low and the line is not easily reversed. This has the advantage that optical integrating cavities are more effective; there is no absorption with radiation of other wavelengths by the unexcited tin in the discharge tube. It has the disadvantage that the line is not as easily excited as a resonance line, as the radiating level is less easily excited from the ground state. Although excitation from the ground state by electron impact is less dependent on angular momentum change than electromagnetic excitation, the large angular momentum change between this level and the ground state inhibits the transition.

There are several types of discharge in which tin may be excited. The spark discharge is of low intensity and tends to yield spectra of the ionized atoms. The d.c. and a.c. arcs are unsatisfactory because the use of tin involves the use of non-oxidizing atmospheres and the arcs operate with low efficiency. Medium or high pressure quartz enclosed arcs (16) with thoriated tungsten electrodes work with indifferent success, mainly due to the tin which must be used as stannous chloride, attacking the electrodes. The low pressure arc, similar to the glow discharge, is incapable of the required output; any attempt to increase the current results in a shift in the operating characteristic toward high pressure arc operation.

The electrodeless discharge, excited by microwaves, has proved so far to be the most efficient trouble-free light source. A small quantity of stannous chloried is introduced into a
quartz tube with one or two centimeters Hg pressure of a noble gas. When placed in a coaxial line or wave guide carrying microwave power, an intense atomic tin spectrum is observed, nearly free from radiation of the chlorine or the noble gas. The noble gas serves as a starter and as a source of electrons for the plasma. The tin chloride volatilizes with difficulty (B.P. 650°C) and, unlike mercury, requires the gas even after the discharge has started.

Stannous chloride may be introduced into the plasma even when the tube operates at a low temperature (∼120°C) by the following artifice. It is known that this salt forms a semiconducting layer on the discharge tube wall (23). If the salt is forced to condense on the tube wall in a region of high electric stress, the induced surface currents will carry a sufficient quantity of the chloride into the discharge to maintain the tin spectra, probably due to the high local temperatures near thin areas of the film. Such selective condensation is effected by local air blast cooling.

The use of argon was expected to be the most efficient filling gas as it has an excited state slightly higher in energy than the group of tin levels of interest. Collisions of the second kind would excite this group of levels in tin and inhibit radiation from this argon level. Neon has a less favorably situated level but experiments (20) have shown that this gas selectively excited the U.V. spectra of some materials more than argon, or indeed more than any of the other noble
gases. We have confirmed this result with tin chloride and find also that it makes the tubes easier to start under pulsed conditions.

A theoretical limitation to the free electron density in a microwave excited discharge exists which sets an upper limit to the power absorption and thus the emission of light. This limitation arises at the onset of plasma oscillations, a collective oscillation of the free electrons in the plasma. As these oscillations become more intense, the impedance of the plasma becomes more reactive and the microwave power is reflected more strongly. In a sense, the plasma becomes "diamagnetic"; the discharge region moves to the walls of the container as the exciting field fails to penetrate. Estimates by Professor S. C. Brown indicate that this effect might become important at the power levels at which the lamps were operated.

Experimental considerations set a limit to the mean microwave exciting power. The statistical nature of the quenching detection allows an advantage to be gained by operating the lamp under pulsed conditions, keeping the mean power constant. If the lamp is on a fraction $f$ of the time, and the lamp output remains proportional to exciting power, then during this period the probability of excitation of positronium atoms is a factor $1/f$ larger than under c.w. (continuous) excitation. The positronium excitation probability is linear in the exciting power, and the peak power is $1/f$ the constant mean power.
If the annihilation spectrum of the positronium is observed only during the duty cycle the observed effect of excitation is $1/f$ larger than under c.w. conditions. As the standard deviation of the total counts varies, at constant counting rate, as $1/\sqrt{t}$ where $t$ is the duration of the counting period, it is seen that to preserve the same ratio of observed effect to fractional standard deviation under pulsed conditions (compared to c.w.) it is only necessary to observe the positronium a fraction $f^2$ of the time necessary with c.w. excitation. The pulsed lamp is only on a fraction $f$ of the time so the period needed to accumulate the required observation time is a factor $1/f$ larger than the observation time.

If $f < \frac{1}{2}$ the lamp-off period may be utilized to find the base counting rate with equal or better accuracy than the "excited" rate if the effect of excitation is small. Therefore, for equal time of collecting data pulsed operation yields an increase in the "statistics" of a factor of $1/f$ and allows the determination of the base-line data, as compared to c.w. operation.

If plasma oscillations begin to destroy the linearity between intensity of the tin line and exciting power at some particular peak excitation power, there exists little benefit by further decreasing the duty cycle. In the present experiment the optimum value of $f$ was about 0.3 and for other reasons (cf. III, 4) a value of 0.500 was chosen.
Such pulsed operation has the additional advantage that slow drift of the electronic equipment, gas pressure changes in the cavity, and other random variations of the parameters of the experiment are averaged over in the data taking. The accuracy of the present experiment could not have been attained by other than pulsed operation.

4. Detection of Excitation

The detection of the positronium formed and the recognition of any induced excitation formed an important part of the experimental problem. As in the ground state splitting determination, the region of positronium formation is observed with a scintillation counter, using a lead collimator to prevent γ-rays from the source itself (or its image on the opposite cavity wall) from scattering into the counter. The pulse height spectrum of the monochromatic γ-rays from pure 2-quantum decay would consist of the photoelectric peak at $m_0c^2$, with the Compton edge at lower energy. The spectrum from 3-quantum triplet decay does not exhibit a sharp peak at $m_0c^2$ as the incident γ-rays are not monochromatic. Changes in the counting rate of that part of the pulse spectrum between the 2-quantum Compton edge and the photo-peak (the "valley") are associated directly with changes in the rate of triplet positronium formation.

If the positronium is formed and excited in a sufficiently high magnetic field, the Zeeman singlet-triplet mixing will ensure that many of the atoms, upon optical de-excitation, will be
in the singlet ground state and will annihilate into two photons. This "quenching" of the 3-quantum decay will then lower the "valley" counting rate. In fields for which \((L S J M)\) is not a satisfactory representation (i.e., region of Back-Goudsmit spin decoupling), each of ten \(P\) states will be mixed 3:7 singlet-triplet or 0.3 of atoms excited into these states will de-excite to singlet states. The essentially complete spin decoupling for \(n = 2\) occurs at much lower fields (below 5000 gauss) than for \(n = 1\), so there is little quenching of the ground state. The lifetime of \(1S_0\) is so short that all atoms are excited from the \(1^3S_1\) state. There are twelve \(P\) states so \(10/12 \times 0.3 = 0.25\) of all excited atoms will decay by 2-quantum decay from the ground state. This calculation neglects possible Stark effect which would mix in the approximately metastable \(\tau = 10^{-6}\) sec \(3S_1\), and the \(1S_0\) from which 2-quantum annihilation occurs directly. At this writing the estimates of the Stark effect make quantitative calculations of the quenching untrustworthy.

If positronium formation occurs in a gas whose pressure is large enough to give a collision rate large compared to the de-excitation rate, then collision mixing among \(n = 2\) states (with essentially no selection rule on \(\Delta L\)) will lead to equal populations of all the triplet levels. There is little induced de-excitation \(\text{}_{\text{I,7}}\). (The probability of collisions with \(\Delta S \neq 0\) is very small for most gases, as discussed in \(\text{}_{\text{II,1}}\).) At least one collision in ten will produce a change of state \(\text{}_{\text{17}}\).
so that in one atmosphere of gas, a positronium atom will undergo at least $10^{11}$ collision-induced transitions sec$^{-1}$ which is large compared to the optical decay rate: $10^9$ sec$^{-1}$. The three $^3S_1$ states are long-lived and the $^1S_0$ annihilates about three times faster than the optical decay rate of the P states, so to a good approximation about 25 percent of the excited atoms will eventually annihilate from the singlet state in the presence of complete collision mixing between the Zeeman split $n = 2$ states.

5. The Lamb Shift

The object of the work reported here is to lay the foundation for actual determinations of the fine structure of the $n = 2$ group of levels. Consideration of these subsequent experiments has influence much of the present design and suggested the program of computation. These considerations form the basis of this section.

In order to measure most accurately the energy difference between two levels of a fine structure pattern, the techniques of microwave spectroscopy are employed to induce the transition directly. Similar to the ground state splitting experiment (15), the expectation in the present case is to induce additional 3-quantum quenching which is directly measurable. As is customary, it would be convenient to use a fixed microwave frequency with a constant power oscillating field and sweep the resonance by changing an external static magnetic field. The transition would be between Zeeman split states. The unperturbed level
differences would be found by extrapolating the energy difference at non-zero field to the field-free case.

If the Stark effect is neglected for the moment, the $^3S_1 - ^1S_0$ energy difference could be measured by the methods used in the ground state, or since the splitting is $1/n^3 = 1/8$ smaller, the transition could be induced directly by a K-band (25,000 Mc/sec) magnetron. Two considerations make the latter course extremely difficult. The transition involves a spin flip and thus the transition probability in a given field is $\sim 157$ times lower than for an El transition. As only a few watts power are available at this frequency, even from experimental c.w. magnetrons (18), the use of an extremely high Q cavity is indicated. In addition, the desired field can only be obtained if the cavity is very small (largest dimension $\sim 1''$) and this appears incompatible both with the satisfactory detection of positronium S-quantum decay quenching and with the rather rigorous demands of the optical excitation.

The experiment analogous to the ground state splitting determination will prove impossible if the expected Stark effect is realized. (See (I, 3).) An accurate energy difference determination by this method would require the Stark effect to be small compared with the Zeeman splitting, but this is incompatible with the Doppler broadening of the 1st Lyman absorption line which allows excitation by the off-frequency tin line.

Any one of the $(^1S - ^1P)(^3S - ^3P)$ energy differences is not only more easily measured (El transitions) but provides more in-
teresting information as it is the angular momentum dependent \( \alpha^5 \) corrections which are the largest: the Lamb shift, vacuum polarization, etc. In addition to the energy requirements to induce those being smaller, there are at least two orders of magnitude more microwave power available for the lower energy transitions involved. The use of low Q cavities (or even the free radiation field) becomes possible.

In view of the expected Stark broadening of many of the lines, it is advantageous to induce the largest energy transition possible which means, with present equipment, a 10,000 Mc/sec or X-band transition. A transition from some sublevel of \( ^3P_2 \) to one of \( ^3S_1 \) is immediately suggested.

In order to detect quenching of 3-quantum annihilation, it is necessary to make the El transition to a state having an admixed singlet state. This restricts the final state to the \( ^3S_{1/2} (m = 0) \), for with a favorably small Stark effect (corresponding to atomic energies \(< 0.2\) e.v.) there would be only an inconsequential singlet component in the \( ^3S_{1/2} (m = \pm 1) \) states.

In the absence of collision induced transitions, the \( ^3S_{1/2} \) states are effectively metastable, but the \( M = 0 \) does annihilate by 2-quantum decay through the field dependent admixture of \( ^1S_0 \). A microwave induced transition to this state may thus give detectable quenching, depending on the admixing.

A second transition would be from the singlet component in the \( ^3P_0 \) state to the \( ^1S_0 \), which (for fields below 4000 gauss) is also an X-band transition. This latter state is more severely
broadened by the Stark effect than the $^3S_0$; it is possible that this broadening would rule this transition out.

All of the above discussion has been under the tacit assumption that the final states of these induced transitions were unoccupied. The effect of positronium collisions with the gas atoms in which it was formed (cf. II, 1) is to induce the above-mentioned (EL) transitions. Unless this induced collision rate is very small compared with the mean life of the excited states, there will be an approximate equilibrium population among all the triplet levels which further microwave transitions will not alter. The principal method of reducing the collision-induced transition rate is to reduce the gas pressure (i.e., the collision rate). As the stopping power of the gas (per cm$^2$) is directly related to the pressure, it is seen that any drastic reduction in pressure involves an equally drastic reduction in the amount of positronium formed in the useful region of the cavity. It will be a difficult problem to reconcile this lowered positronium production at low gas pressures with the statistical problem of recognizing the minute changes in quenching involved in excitation and subsequent microwave induced transitions.
PART III. THE EXPERIMENT

1. Introduction

In the experimental excitation the positronium is produced in a cylindrical cavity. The positron source, Na$^{22}$Cl, is mounted at the center of one end plate. A magnetic field parallel to the axis of the cavity collimates the positrons from the source in a beam along the axis. Positrons slowing and stopping in the filling gas can form positronium along the region of the positron beam. A 270° toroidal electrodeless discharge tube is incorporated as part of the cavity wall (see Fig. 1). A pair of Al reflectors helps form an optical integrating cavity. Some of the positronium is produced in this sub-cavity and is irradiated by the lamp which in turn is excited through coaxial line coupling to three 100-watt S-band magnetrons. The region of irradiation is "observed" with a NaI(Th) scintillation counter, using a 10" lead collimator to reduce spurious scattered γ-rays. The spectrum of pulses from the counter is passed to a single channel differential discriminator and to the registers. The following sections describe these components in detail.

2. The Positron Source

Positrons are furnished from the radioactive decay of Na$^{22}$ prepared in the MIT cyclotron by the Mg$^{24}$($d,α$)Na$^{22}$ reaction. With a 2.6 year half-life and a maximum positron energy of 0.54 mev,
this isotope emits a 1.28 mev gamma ray in coincidence with the positron. A 200-hour deuteron bombardment of Mg$^{24}$ (15 mev, 60 microamperes) yielded about 25 mc. activity of which $\sim 15$ mc. was vacuum evaporated, as the chloride (with $\sim 0.3$ mg. Na$^{23}$Cl as carrier), on a $1\frac{1}{2} \times 1/8"$ silver-plated brass disc. Using an oven design which produced a narrow beam, about 80% of the final source activity was deposited on a region 1/8" in diameter, the remainder was confined to a 3/8" diameter spot with a glass collimator. A 100-microgram/cm$^2$ layer of gold was evaporated over the face of the disc to protect the chloride. Adhesion of the active material to the silver plate was ensured by the use of a layer of Vinylseal cement approximately 80 micrograms/cm$^2$ which was spread on the source plate as a solution in toluene and allowed to dry. The thickness was gauged by the interference colors. Vinylseal, when properly degassed, has an extremely low vapor pressure ($< 10^{-7}$ mm. Hg) and its softening point of 150°C coupled with its superior adhesive properties make it an admirable material on which to deposit Na$^{22}$Cl. The pre-baking to eliminate residual toluene usually routine in the use of Vinylseal was omitted in view of the very thin films involved and the duration under vacuum before evaporation.

The utility of a small source lies in the reduced size of the resulting collimated positron beam which, in turn, prevents positrons from annihilating on the cavity wall where the annihilation radiation may be "seen" by the $\gamma$ counter. Efforts were made to design the oven so that the atomic NaCl beam was
narrow, rather than depending on defining apertures to limit the source size. In this way a higher percentage of the active material could be deposited on the source plate.

The stopping power of the gas in the cavity is not very great, particularly at low gas pressures. It might be expected that an absorber of properly chosen thickness would reduce the mean energy of the positron spectrum in such a manner as to furnish more low energy positrons (e.g., below 50 kev) although attenuating the whole beam, so that more positrons would stop in the gas. That this expectation is in error follows from the fact (21) that a beta ray spectrum is attenuated approximately exponentially in an absorber so that the energy distribution remains very nearly the same. Thus any absorber will reduce the numbers of low energy particles; there can be no increase in the present situation of "poor" geometry. The amount of positronium formed in a given region of a gas is greatest when the stopping power of that region is greatest and there is a minimum of absorber between the region and the source.

3. Cavity Design

The cavity design was similar to that employed by Dulit (19); it consisted of an open-ended Vycor cylinder 2\(\frac{1}{2}\)" O.D. by 3\(\frac{1}{2}\)" long, with a closed "side-arm" at the center of 1" Vycor 2" long, which formed part of the \(\gamma\) collimator. This projected downward into the oval hole in the lead collimator. The cav-
ity ends were ground flat so as to form reliable vacuum seals onto the flat silicone rubber gaskets mounted in grooves in the two 1/4" brass end plates. A recess on the inner surface of one disc retained the Na\textsuperscript{22} source plate by means of two beryllium-copper clips. See Fig.1.2.

Advantage was taken of the backscattering properties of platinum (21) to utilize the "image" source on the opposite wall of the cavity from the source. As many as 80% of the positrons arriving at the image could be returned with only a slight lowering of their mean energy. Only the central 1/2" diameter of the opposite wall was utilized, however, by constructing a platinum foil spot of this diameter. Lucite was used to form the remainder of the wall as it backscatters less than 10% of the positrons and lowers their mean energy strongly (21).

This construction reduced counting-background from positrons backscattered from the outer regions of the image, for the reduced backscattering insured annihilation in the lucite. The use of the platinum becomes increasingly effective at lower gas pressures when the mean free path of the positrons is longer than the cavity length, and fewer positrons are stopped by the gas.

A sidearm of 5 mm Vycor projected horizontally from the cavity near one end and was connected to the gas handling system.

The toroidal discharge lamp was constructed of 10 mm (I.D.) quartz tube and formed 270° of a circle the diameter of the cav-
Figure 2
END VIEW: LAMP CAVITY, MICROWAVE EXCITATION
ity. See Fig. 2 The lamp was sealed into and formed part of the wall of the Vycor cavity. The lamp was cemented with Sauereisen Insalute cement, which forms a good mechanical bond and is nearly as heat resistant as Vycor. The lamp fit was very carefully made so as to avoid the use of an appreciable thickness of cement with its high thermal expansion. The vacuum properties of the joint were established by the use of DC 801 Silicone baking resin, a clear thermosetting varnish of high thermal resistance. This material was mixed with titanium dioxide to form a thick paint, applied to the exterior of the cemented joint and cured at 250°C for 18 hours.

The optical cavity was formed by the lamp and two circular discs of aluminum foil, the internal diameter of the cavity whose axes coincide with the cavity axis. These discs were placed inside the cavity on each side of the lamp. The outer 1/8" of the discs is made from 10 mil Dural sheet; thin aluminum foil (160 micrograms/cm²) formed the center. The thin foil was made to adhere with an extremely thin layer of Silicone high vacuum grease. The discs were retained in their positions by 16-gauge copper-wire snap rings. The foils increased the vacuum valley-counting rate by 10%, a negligible contribution to the background rate.

The exterior of the optical cavity (i.e., the lamp exterior) was smoked with MgO. MgO is one of the best reflectors for ultraviolet (22) and, contrary to some sources, preserves this property for some time following exposure to the atmosphere.
Because of the delicate nature of the foils no attempt was made to construct an electrode system to apply a re-accelerating electric field to the region of positronium formation. The field would have to be very uniform so as not to exert a force on these thin foils. The excitation experiments therefore used a gas chosen to inhibit r.f. re-acceleration, although with some modification the present design could be adapted to the use of d.c. fields.

The θ collimator was designed to allow observation of a region \( \frac{1}{2}'' \) (the discharge tube thickness) by 1'' using criteria established by Weinstein (13). The collimator was cast of lead and was 10'' high, 2'' more than necessary but chosen so as to remove the scintillation counter from the interior of the magnet.

4. The Lamp

Of the several methods described in (III, 2) of exciting the atomic tin spectrum the most useful has proved to be the electrodeless discharge employing microwave exciting power. Early experiments immediately eliminated the tin electrode a.c. or d.c. arcs primarily due to the low melting point of tin (232°C), but the necessity of operating the arc in a non-oxidizing atmosphere would have introduced gas handling, cooling, and optical problems in addition to the objection of having to operate the arc in the magnetic field required for positronium formation.
1. 7/8" Coaxial Line
2. Tin Discharge Tube
3, 4. Al Foils
5. Vycor Cavity
6. End Plates and Water Cooled Clamp
7. Magnet Pole Pieces
8. Gas Filling Inlet
9. Source Plate
10. Lead Collimator
11. Scintillation Counter

(Not drawn to scale)

Figure 3
SIDE VIEW: MAGNET, CAVITY, COLLIMATOR
An enclosed arc operated in a quartz bulb with sealed-in electrodes may not be entirely unfeasible but the principal difficulty appears to be the elimination of chemical attack by the tin salts on the hot electrode structure. Metallic tin cannot be used in the tube for two reasons. Its boiling point is so high (2270°C) that to introduce an appreciable amount into the discharge plasma requires that the entire tube be operated at a temperature far in excess of the devitrification temperature of quartz (≈1000°C). In addition, metallic tin sputters badly and soon coats the walls of the tube. This latter defect can, in general, be reduced by proper choice of filling gas pressure but not to a satisfactorily low level. The use of a readily volatilized salt such as stannous chloride does indeed introduce sufficient tin into the discharge to dominate the emitted spectra, but decomposition of the salt at the hot electrodes still gives an undesirable level of sputtering of metallic material. The use of low work function emitters (e.g., thoriated tungsten) would lower the electrode temperature greatly and might eliminate the decomposition.

Cold cathode discharges cannot be operated at high power inputs without having the discharge change to the (high current) arc type. Cold cathode low pressure discharges are inherently unsuited to high power inputs and have not been investigated during the present work.

Microwave excitation of quartz electrodeless discharge tubes containing stannous chloride and a rare gas operates
quite satisfactorily. The halides of many metallic elements have been used to allow excitation of the metal spectrum (24, 25, 26). Such discharges operate without sputtering, with little halide or filling gas spectra and no trace of the ionized metal spectra. The quartz tubes are placed, for example, in 7/8" S-band coaxial line carrying ten or more watts r.f. power. The discharge is initiated by application of the spark from a Tesla coil.

Preliminary experiments established that a pressure of \( \sim 1.8 \text{ cm (Hg)} \) of neon gas in a 10-mm (i.d.) quartz tube in which a trace of stannous chloride had been placed radiated the most energy at 2430 A.U. The tubes were excited by using them as a continuation of the center conductor of the 7/8" coaxial line from a QX61 S-band magnetron. Part of the exterior conductor was cut away to allow the light to escape. The magnetron was operated at an output of \( \sim 100 \text{ watts} \) and a double-slug coaxial tuner prevented the high standing wave ratios (15 db.) existing at the lamp from interfering with the magnetron operation. The tuner was adjusted to maximize the lamp output at 2530 A.U.

Lower than optimum gas pressures (e.g., \(< 1.0 \text{ cm Hg}\)) appear not to load the discharge sufficiently; it is considered that the filling gas inhibits the diffusion and condensation of the tin chloride vapor and that at low gas pressures an insufficient amount of the salt is in the plasma. At higher pressures (\( > 2.5 \text{ cm Hg} \)) the noble gas spectrum becomes increasingly pre-
dominant and there is no further increase in the intensity of the tin spectrum.

The total power radiated at 2430 A.U. was measured in the following manner. A mercury discharge tube was prepared from 9-mm Vycor tube, 60 mm in length. The tube contained about 100 mg of mercury and no filling gas. At low excitation (≈ 20 watts microwave power) and with carefully adjusted air cooling it finished a copious source of resonance radiation at 2537 A.U. The tube was difficult to start; it had to be heated to ~400°C and initiated with a Tesla coil, although once in operation its temperature was near 50°C. Using a $2\frac{1}{2}$" diameter fluorite lens of 10" focal length an image of the source was formed with unit magnification. The image was formed after the beam had passed through a filter to isolate the 2537 A.U. line, and a portion was intercepted by a type 25A W.E. thermistor which formed one leg of a bolometer bridge circuit. See Fig. 4. The bridge unbalance caused by the change of resistance of the thermistor heated by absorbed radiation was corrected by a calibrated potentiometer in another bridge leg. This gave a direct measure of the radiant energy at the bridge thermistor. Knowing the solid angle subtended by the lens at the lamp, one finds the source brightness (watts/cm²/steradian) and using the lamp dimensions and projected area in all directions the calculation of total radiated power is immediate. The assumption of uniform source brightness in any direction (Lambert's Law) was checked experimentally, as was the uniformity of all parts of
the source radiating in a single direction. The mercury tube was uniform in both these respects, unlike the tin-containing tubes.

A standard carbon filament lamp with a National Bureau of Standards calibration was used to calibrate a ribbon filament projection lamp (GE 18A/T10/2P-6V). The two lamp filaments were adjusted to the same color temperature with a Leeds and Northrup optical pyrometer and the relative radiated energies measured with a GE DW58 light meter. The 1/8" x 3/8" radiating area of the projection bulb made the absolute calibration of the relatively insensitive thermistor bolometer bridge far easier than the direct use of the standard bulb with its 1 1/2" loop filament.

About 90% of the energy radiated by the mercury tube was in the 2536 A.U. resonance line if the tube was operated at low excitation power. Filters of the usual type designed to isolate a narrow spectral region do not exist for this part of the ultraviolet spectrum so recourse was had to a liquid filter modified from a design by Kasha (27). A 2.0 cm optical path of 2 parts Kasha's Ni So₄ - CoSO₄ solution diluted with 1 part H₂O provided a peak transmission of 0.33 at 2700 A.U. and 0.30 at 2537 A.U. The solution was retained in a 20-mm diameter glass cell with 1-mm Vycor ends. The transmission was approximately zero for wavelengths below 2200 A.U. and above 3600 A.U. The transmission was measured from 2000-4000 A.U. with the Carey recording spectrophotometer.
THERMISTOR BOLOMETER CIRCUIT

Figure 4

Thermistors mounted in vacuum enclosure with a 0.01" thick quartz window allowing radiation to fall on one thermistor

Calibration constant

17 Microwatts/cm²/scale division (Vacuum)
170 Microwatts/cm²/scale division (Atmos. air)

Time Constant

∼ 30 sec. (Vacuum)
∼ 3 sec. (1 Atmos. air)
That no other lines within the transmission region of the filter radiated an appreciable energy was verified by the quartz monochromator described below. It was found with this latter instrument that $\sim 80\%$ of the radiated energy was radiated in the 2537 A.U. line and $\sim 10\%$ in the persistent line at 3650 A.U. It is interesting to note that this energy distribution can be altered radically by increasing temperature and higher exciting power.

The mercury tube radiated a maximum of 10 watts in the line at 2537 A.U.

Light from the lamp was intercepted by a MgO smoked plate 2 cm square at a distance of about 10" from the tube. This plate was mounted at the entrance slit of a Bausch and Lomb quartz crystal monochromator at $45^\circ$ to the entrance axis. The diffusely reflecting plate formed the "source" for the monochromator. At the exit window was mounted a photomultiplier socket which would allow the use of either a LP21 or LP28 photomultiplier. The phototube was supplied with 1500 volts and the current to the last dynode measured with a 0-100 microammeter.

The MgO smoked plate had two advantages as a monochromator source. It made the instrument insensitive to small changes in orientation about the position of maximum light collection and, as it viewed the entire discharge tube, the meter reading was proportioned to the total power radiated in some spectral line, independent of the tube shape or size.
This instrument was in operation during the absolute radiated energy measurements at 2537 A.U. on the mercury tube. By substituting a tin discharge lamp and observing the reading at 2430 A.U. a direct comparison was obtained of the total power radiated into these two lines. It was assumed that the monochromator detector had a uniform wavelength response. The detector was designed so as to have this property in good approximation. A thin layer of sodium salicylate was deposited by evaporation of a saturated solution of the material in high purity acetone on the glass jacket of a 1P21, over the photocathode. The 1P21 jacket is opaque to wavelengths below 3000 A.U. but the sodium salicylate phosphor has a constant quantum yield (28) of fluorescent radiation in the blue visible. Thus the detector efficiency is approximately constant in the region of 2500 A.U.

By the use of the monochromator the best tin-containing straight discharge tube was found to radiate 1.0 watt at 2430 A.U. with a microwave exciting power of 100 watts. The estimated uncertainty is 20%.

Pulsed operation of the lamp, with 50% duty cycle and the same average excitation, gave about 2.0 watts peak power radiated at 2430 A.U.

The circular lamps were not well adapted to such quantitative measurements due to the poor geometry. The best comparison measurements indicate that single magnetron excitation yields the same radiated power as from the straight lamps.
Through the courtesy of the Spectroscopy Laboratory the spectra of the best straight tin-containing tubes was investigated with the high resolution echelle spectroscope. The line at 2430 A.U. shows no reversal (as expected) and is somewhat narrower than 0.01 A.U. The observed breadth is attributed to pressure broadening.

The preliminary program of "tin" lamp development included an investigation of the effects of variation of the parameters (1) gas filling (2) tube size (3) microwave matching (4) microwave frequency (5) duty cycle and repetition rate of pulsing. The monochromator previously described was used throughout this series of experiments as a reproducible instrument giving quantitative relative data on the effect of varying these parameters.

The gases, neon, argon, and xenon, were tried. Xenon would not support a stable discharge. Argon was satisfactory over a range of pressures from 0.05-2.5 cm (Hg) with optimum pressure in the range below 2.0 cm. These tubes were frequently difficult to start and gave a non-uniform filamentary discharge of satisfactory stability. Neon, tested over the same pressure range (10 cm microwave excitation), yielded about 30% more radiation than argon at 2430 A.U. at the optimum pressure of 1.8 cm (Hg). At low excitation (~ 50 watts) the tubes could be kept reasonably cool and the discharge with neon was uniform throughout the tube. At maximum excitation the increase in pressure due to heating caused the discharge to be-
come non-uniform and filamentary. This did not appear to inhibit the radiation of the tin spectrum.

The total energy radiated at 2430 A.U. depended approximately linearly on the tube volume; the final choice of 10 mm i.d. tube was dictated by the maximum size useful with the cavity-collimator design. The circular tubes were made as long as possible; longer ones could not have been sealed in the cavity.

The large tubes had the advantage that excitation was possible with almost any arrangement of coaxial line and tube. As long as the lamp was partially surrounded by the coaxial line exterior, and the inner conductor made contact with the tube, excitation was satisfactory. Adjustment of the slug tuners adequately corrected variations in the mismatch.

A 3-cm magnetron with an output of 200 watts was tried briefly. It excited the filling gas strongly but not the tin.

Using 10-cm single-magnetron excitation on a 2" straight tube, with $\sim 100$ watts mean exciting power, the peak pulsed light output at 2430 A.U. was approximately linear for the peak power below 200 watts. There was no increase in the line intensity for peak powers $\geq 300$ watts. As a source of precision square waves was not difficult to construct the above results justified the final choice of 50% duty cycle pulsing.

Lamp output was essentially independent of pulse repetition rate. This rate was kept as high as consistent with
the 50 μsec rise time of the light pulses, to avoid long periods of high emission from the overloaded magnetron cathodes. The repetition rate as finally used was 400 c.p.s.

5. Microwave Components

Three QK61 10-cm magnetrons furnished the microwave exciting power for the excitation experiment. They were mounted external to the magnet and individually blower-cooled. A standard 7/8" coaxial double-slug tuner was connected directly to each magnetron and 7/8" coaxial line coupled these directly to the lamp. Two of the coaxial lines were fitted against the sides of the Vycor cavity by cutting away cylindrical portions of the lines. The center conductors were in direct contact with the discharge tube. The third line ended at the top of the cavity; its center conductor was also in contact with the discharge tube. See Figure 2.

Power for each magnetron was provided by separate 3000-volt 400-ma. d.c. supplies. Current regulation and pulsing control was furnished by a series 813 tube in each supply. The three 813's were pulsed from a common source, and a single 500-volt d.c. variable screen-grid supply allowed one-control operation of the three magnetrons.

Although nominally rated at 120 ma. each, the QK61's oper-
ated satisfactorily at currents up to 200 ma. average, 400 ma. peak. The use of current rather than voltage stabilized power supplies probably contributed to this overload tolerance.

6. Magnet

A high intensity precision magnet of a design of Professor F. Bitter of MIT furnished the required magnetic fields. This magnet was used in the positronium ground state splitting experiment and is described by Weinstein (13).

The proton resonance field stabilization was not used—sufficient stability was obtained from the current control unit.

The magnetic field was measured with a laboratory gauss meter for the range of currents used in the experiment.

7. \( \gamma \)-Ray Detection

\( \gamma \)-ray detection was effected with the same scintillation counter, BNL design non-overloading amplifier and single channel differential discriminator, as used by Weinstein (13). The face of the DuMont photomultiplier was ground optically flat and a 1" lucite light pipe used to move the tube away from the NaI (Th) crystal. The tube was fitted with a double cylindrical shield, the inner of Mu-metal, the outer of Nicoloi, and the entire counter was assembled within a 1/8"-thick demagnetized iron tube 2 1/4" in diameter. This prevented the magnetic field from effecting the photomultiplier gain.

The differential discriminator showed indications of intermodulation distortion, for the pulses from the discriminator had
a 60-cps variation in counting rate amounting to about 2\% of the gross rate.

This eliminated the possibility of using a 60-cps pulsing of the lamp, for the counting rate during one-half cycle was about 5\% higher than during the other. Use was made of the 400-cps power line to furnish a repetition rate which would average over these 60-cps variations. This proved satisfactory.

The amplifier and discriminator power supplies, the scintillation counter supply, and the master pulse and gating circuits were all supplied from a constant voltage 110 v transformers.

3. Pulsing and Gating Circuits

The master pulse unit was designed to furnish a square wave with a rise time of the order of 0.5 \mu sec and stability and equality of the on-off intervals to better than 0.1\%. A Schmidt trigger circuit with a 0.5 megohm voltage divider circuit using precision resistors, including a 10,000-ohm wire-wound potentiometer to set the d.c. level accurately, allowed the exact zero of the 110-volt 400-cps signal to initiate the circuit. Using the potentiometer to make small readjustments in the d.c. level, the square wave intervals could be adjusted accurately to within 0.04\%. Although the circuit frequently remained stable within this figure for several hours at a time, a test oscillator was built on the same chassis to facilitate fast and accurate checking.
A negative pulse from the Schmidt circuit cut off a 6SN7, which in turn removed cut-off bias from the three 813's controlling the individual magnetron currents. The large inter-electrode and filament transformer capacitances associated with the magnetrons and their current supplies resulted in the rise time of the light pulses being of the order 50 μsec, but this was of little importance as long as the light was on most of the interval, as was indeed the case.

Two gated beam tubes with their #1 control grids connected to the output of the differential discriminator were gated out of phase, using the precision 400-cps square wave, each output going to an Atomic Instrument scale-of-64, and also to a scale-of-256 register.

An unstabilized 884 relaxation oscillator operating as a pulse generator with a repetition rate near 2.5 kc was used as the oscillator for making rapid determinations of the balance or equality of the square wave intervals.

By switching the oscillator output into the gated tubes, and using the scale-of-256 registers in each channel, 1000x256 pulses could be accumulated in each register in a few minutes. The difference between the registered counts was a direct measure of the unbalance of the intervals.

The test oscillator was used to check the balance directly before and after each data-taking run. Using this as a standard the balance could be checked and reset to a reproducible value.
BLOCK DIAGRAM: EXCITATION EXPERIMENT
Figure 5
Such a test could not, of course, be relied on as the absolute determination of balance to the highest accuracy; it was used primarily to enable accurate check and reset.

The scale-of-64 registers were used during data taking.

9. Ultraviolet Line Intensity Monitoring

In order to monitor the ultraviolet intensity in the cavity during excitation runs, the monochromator previously described was mounted outside the magnet where it could view directly a \( \frac{1}{2} \)"-hole in the coaxial line which, in turn, exposed a 1/4"-hole in the MgO reflection coating on the discharge tube.

A 0-100 \( \mu \)ampere meter measured the average current to the last dynode of a LP21 detector; in addition, the current pulses were observed on a DuMont oscilloscope. The meter was used in the very critical tuning of the microwave equipment before and during excitation runs. In this way the intensity of the desired tin line could be maximized with precision.

The pattern of the light pulses was constantly observed and incipient instability in the lamp output corrected by readjustment of the tuners. The monochromator could not easily be used for any but relative intensity measurements. No attempt was made to measure the absolute light intensity in the cavity.

10. Experimental Procedure

Previous to any data-taking runs, the electronic equipment was allowed at least a six-hour warm-up and several balance
checks were taken to insure that drift was negligible. With the cavity in place in the collimator, the optical reflectors installed, and the coaxial lines in contact with the lamp, the source would be placed in position and a vacuum established. After about 20 minutes of pumping the cavity would be flushed and then filled to exactly 1 atmosphere pressure with the desired filling gas.

The differential discriminator was used to check the position of the annihilation peak of the pulse height spectrum and small readjustments of the photomultiplier high voltage made to reset this to a standard value. The acceptance window of the discriminator was set at a width 1/7 the pulse height of the peak and positioned in the valley of the pulse height spectrum. Cf. Weinstein (13).

The lamp was then initiated at low excitation and the magnetic field established. Air blast cooling of the lamp was started. At about 0.8 of maximum excitation the coaxial tuners were adjusted to maximize the lamp output at the desired wavelength. Final adjustments were made at full power, usually 185 ma per magnetron.

A balance check in progress during the tuning process would be concluded and, if satisfactory, an actual run would be started. Between 7-10,000 x 64 counts would be accumulated in light-on and the light-off registers, requiring from 40 minutes to one hour. If during the run the light intensity could not be pre-
vented from falling below 85% of its initial value by retuning, the run was terminated. A balance check followed immediately in all cases. If the balance had not changed more than 0.15%, the average of the initial and final balance values was used to correct the data. If the final unbalance exceeded 0.15% the data was rejected. The average change was about 0.03% and was frequently < 0.02%.

Most of the runs were taken at night to avoid the power-line fluctuations and transients so frequent during the day.

The heat from the lamp would raise the gas pressure in the cavity to 1 or 2 psi (gauge). This was tolerated as it rendered leakage into the cavity impossible.
Figure 7

Cavity with end plates and water-cooled clamp, in position in collimator. One magnet pole piece is shown. The coaxial line discharge tube excitation is in place but discharge tube is not MgO coated.
Figure 8

Magnet open, showing cavity, collimator and microwave components in place. The magnetrons with the slug tuners are mounted above the magnet; the monochromator (left foreground) views the discharge tube through a hole in the coaxial line.
PART IV. RESULTS

1. Excitation and Auxiliary Experiments

Propylene was used in all the experimental runs. Experiments by Dulit (15) show that more than three times as much positronium is formed in this gas than in argon of an equal pressure. Three-quantum positronium annihilation accounts for two-thirds of the observed valley counting rate. Propylene is expected to inhibit almost entirely re-acceleration of positrons in any r.f. fields penetrating the cavity. There is no interaction of the ultraviolet radiation with the double bond structure of propylene; no wavelength transmitted by quartz excites or ionizes propylene (33).

All runs were taken at a magnetic field of 5200 gauss. This is more than sufficient to entirely decouple the electron and positron spins in the excited state and ensure maximum three-quantum quenching. For example, the branching ratio for two-to three-quantum annihilation from positronium initially in the \( 2 \, ^3P \_1 \) state is 24:25 at 3000 gauss, 24.5 : 25 at 5000 gauss; for the \( 2 \, ^3P \_o \), 16 : 33 and 20.5 : 28.9 (from calculations with thermal Stark effect.)

To test the equality or balance of the precision square wave intervals, the balance was adjusted to equality using the test oscillator and the true balance found using the valley counting rate from a small Na\(^{22}\) source placed near the scintillation counter. The same discriminator settings were used as
in actual runs. In this manner the balance error was found to be

\[-0.18 \pm 0.07\%\]  \hspace{1cm} (A)

where the negative sign indicates that the light-on interval was shorter than the light-off by the amount indicated.

The actual excitation runs, with complete optical cavity including an opaque MgO exterior reflector gave the fractional difference in total counts

\[-0.34 \pm 0.08\%\]  \hspace{1cm} (B)

The error shown is the fractional standard deviation of the difference between the total counts in the light-on and light-off channels after individual runs had been corrected for balance error. The sign indicates a decrease in the light-on rate and thus a decrease in the three-quantum decay rate during excitation.

To test that this was indeed an optical effect, the magnetic field was established before the magnetrons were energized. Several runs were taken under these conditions in which the lamp did not strike on. With the magnetrons operating at 0.6 of normal current it was expected that the r.f. field in the cavity would be of the same order of magnitude as in normal light-on operation for the high r.f. reflection of the ionized gas plasma would be absent. In this way unexpected r.f. effects would be detected. The unbalance was measured as

\[-0.15 \pm 0.07\%\]  \hspace{1cm} (C)

in satisfactory agreement with result (A).

* The experimental data is tabulated in Appendix VII.
An additional confirmation consisted of removing entirely the MgO reflection coating, but otherwise preserving the conditions of the excitation runs. With the magnetrons operating at full input, and the ultraviolet intensity maximized, the r.f. and thermal conditions of an excitation run were duplicated. With the 98% reflectance of the entire exterior of the optical cavity removed, the optical "Q" would be lowered to such an extent that excitation would be undetectable. This is true assuming that the exciting radiation is effectively reflected completely only twice before extinction, a conservative assumption. The removal of this thin dielectric coating, only a few mg/cm² thick, would not alter the electrical properties of the system, particularly after adjusting the slug tuners. The resulting unbalance was

$$(-0.10 \pm 0.09)\%$$

which is in agreement with results (A) and (C). Progressive deterioration of the source prevented any further increase in the accuracy of the above results.

Results (C) and (D), taken with r.f. fields present in the cavity, are both lower than (A) with no r.f. present. Although in no sense could a real effect be inferred from the poor statistics, an effect in the observed direction might be expected by residual r.f. re-acceleration of positrons whose energies were only slightly below the positronium formation threshold. This would increase the rate of positronium formation during r.f. -on periods, leading to a higher light-on counting rate,
an effect opposite to that of excitation.

2. Conclusions

The change in the gross valley counting rate attributed to excitation is the difference between results (A) and (B). This is

\[ (-0.16 \pm 0.10)\% \] (E)

Any re-acceleration of positrons during excitation would cause a reduction in the observed effect, and this is an argument to use the r.f. -on, light-off data (C) as the base. This yields a counting rate change attributed to excitation of

\[ (-0.20 \pm 0.09)\% \] (F)

Results (E) or (F) are of the order of magnitude expected.

Lamp intensity measurements yielded the result that about 1 watt was radiated at 2430 A.U. per 100 watts microwave exciting power. An average power output of 100 watts was obtained from each of three magnetrons operating on a 50% duty cycle so that about 6 watts peak ultraviolet of the proper wavelength was available. Assuming a mean reflectivity of the optical cavity of \( \sim 0.7 \) the optical \( Q \) was about 3. (\( Q \): the ratio of stored to dissipated energy.) Most of the light would be expected to travel approximately perpendicular to the cavity axis as the best reflector and the light source were located at the edge of the pill-box shaped region. Thus most of the light would pass through the 6 cm² cavity cross section parallel to the axis. Of the order of 3 watts/cm² would be avail-
able for excitation. Combining the results of Part I, Section 4 and Part II, Section 3, about 0.2% three-quantum quenching would be expected. As about 0.6 of the observed valley counting rate is from triplet positronium annihilation, one finds from (E) or (F) that the observed quenching of three-quantum decay is between 0.25% and 0.3%.

These figures are in good agreement with the estimate and it thus appears reasonable to assume that excitation has been induced.

3. Suggestions for Further Work

The present equipment could be modified in several respects to enhance the observed effect. By either smoking the Al foils with MgO or replacing them with a higher reflectance metal the optical Q could be raised. More careful design of the microwave fittings would avoid small dislocations of the external MgO film and possibly improve the microwave coupling to the discharge tube. More efficient cooling (using air at dry ice temperatures) would allow the use of more powerful magnetrons.

It is possible that the observed effect could be increased by a factor of 5. By making the Vycor cavity longer, thus moving the source away from the excitation region would allow the γ-counter to be moved much closer to the cavity. A fourfold increase in the counting rate would help greatly in reducing the statistical errors by decreasing the counting time.

The possible extension of the present methods to enable actual fine structure measurements to be made is hopeful. The experiment reported here is felt to be the first successful step in this direction.
APPENDIX I

Zeeman Calculation

We wish to evaluate

$$\langle S L J M \mid G_2, -G_2, \mid S' L J' M \rangle$$

There is no loss of generality in setting $S = 0$, $S' = 1$. Decompose the initial state in eigenfunctions of $m_s m'_s$

$$= \sum_{m_s m'_s} \langle 0 L J M \mid G_2, -G_2, \mid 1 m_s L, m'_s \rangle (J' M \mid 1 L M_s, m'_s)$$

where $M_s + M'_s = M$ and $(J' M \mid 1 L m'_s m'_s)$ is a Clebsch-Gordon coefficient.

$$= \sum_{m'_s} \langle 0 L J M \mid G_2, -G_2, \mid 1 m_s L, M-M'_s \rangle (J' M \mid 1 L M_s, M-M'_s)$$

$$= (J' M \mid 1 L 0 M) (0 L L M \mid G_2, -G_2, \mid 1 0 L M)$$

since $M_s$ can have value zero only.

$$= (J' M \mid 1 L 0 M) (\chi, G_2, -G_2, \mid \chi^0)$$

using the spin function notation of (9)

$$= 2 (J' M \mid 1 L 0 M) (\chi^0, \chi^0) = 2 (J' M \mid 1 L 0 M)$$

$H'$ for S states is

$$H' = \frac{\hbar}{\mu} \sum_{\ell S \ell' S'}$$

We disagree on the signs of several entries (with Ferrell).
**APPENDIX II**

**Stark Calculation**

Let $H = \frac{H_2}{2}$ and $\nu = \nu_y$ so the transformed electric field is in the $x$-direction.

$$E_t = \frac{\nu}{c} H$$

$$H_s = e E_t \overline{X}_o = e E_t n \cdot \sin \theta \cdot \cos \theta$$

$$= e E_t n \cdot \frac{1}{2} \sqrt{\frac{\theta \pi}{3}} (Y^{-1}_1 - Y^1_1)$$

using the normalized spherical harmonics of Blatt and Weisskopf (9).

We want

$$(nLsJM | H_s | n'L's'J'M')$$

specializing for $n = 2$ set $L' = 0$, $S' = J'$, $n = n' = 2$.

$$(LSJM | H_s | OS'S'M') = \sum (JM | 1S_m_s m_s) \left\{ S_m_s | S'm'_s \right\} \cdot (L,M | H_s | 00)$$

Now $m_s = m - M$

$$= \sum (JM | 1S_w M') \delta_{ss'} R \cdot (L,M | H_s | 00)$$

$$= RG \frac{1}{\sqrt{4\pi}} \int Y_L^{M-M'} (-Y^{-1}_1 + Y^1_1) dS \cdot (JM | 1S_w M')$$

where $Y_o^0 = \frac{1}{\sqrt{4\pi}}$ and $L$ must = 1
and \( g = eE_t \sqrt{\frac{8\pi}{3}} \)

\[
R = \int R_2^2(n) \cdot n \cdot R_2'(n) \cdot n^2 \, dn = -3\sqrt{3} \, a.
\]

So

\[
(L_{SM}|H_s|0S'\,S'M') = (J_M|M',M-M')(-1)^{J'J-1} \delta_{ss'} \frac{3\sqrt{2}}{2} (a_0E_t) \left( \delta_{M-M'_1} - \delta_{M-M'_1} \right)
\]
APPENDIX III

Excitation of Positronium

The square of the induced transition matrix element from a state $L S J M$ to the states $L' S' J' M'$ is desired, summed over $J', M'$:

$$\sum_{J'M'} \left| \langle LSJM | \vec{r} \cdot \vec{E} | L'SJM' \rangle \right|^2$$

(1)

If $\vec{E}$ represents isotropic radiation we can average over $\theta$ in

$$\left| \vec{r} \cdot \vec{E} \right|^2 = \left| \vec{r} \right|^2 E^2 \cos^2 \theta = \frac{E^2}{3} \left| \vec{r} \right|^2$$

$$= \frac{E^2}{3} \frac{4\pi}{3} \sum_{J'M'K} \left\{ \left| \langle LSJM | Y^{(i)}_K | L'SJM' \rangle \right|^2 \right\}$$

(2)

where $Y^{(i)}_K$ is a normalized spherical harmonic as in Blatt and Weisskopf (9). Using the Wigner-Eckart theorem (2) becomes

$$= \frac{4\pi E^2}{9} \sum_{J} \frac{1}{(2J+1)} \left| \langle L \parallel Y \parallel L' \rangle \right|^2 \left( 2J+1 \right) \left( 2J'+1 \right) \left| W(LJLJ' ; S1) \right|^2$$

where $W (\ldots)$ is a Racah coefficient (31)

$$= \frac{4\pi E^2}{9} \frac{1}{2L+1} \left| \langle L \parallel Y^{(i)} \parallel L' \rangle \right|^2$$

(3)

and thus the probabilities of excitation from all initial states are equal.
To show that the transition yields equal populations in each final $M$ state, write Eq. (2) in terms of the Racah function:

$$
\frac{\xi^2}{9} \sum_{J'} \frac{1}{(2J'+1)} \left| Z(LJ'J'; S1) \right|^2
$$

(4)

The $Z$ function is tabulated (Biedenharn (32)) so (4) becomes, for the final states $J'$ of the transition $3_{S_1} \rightarrow 3_{P_{J'}}$:

$$
\frac{\xi^2}{27} \left\{ \begin{array}{cl}
1 & J' = 0 \\
3 & J' = 1 \\
5 & J' = 2 \\
\end{array} \right.
$$

but these are the statistical weights of the states so that for all $M'$ levels the square of the matrix element is $\frac{\xi^2}{27}$.

To evaluate (1), write the reduced matrix element in (3) in terms of $V$ coefficients, for $3_{S_1} \rightarrow \sum_{J'} 3_{P_{J'}}$:

$$
= \left( \frac{\xi^2}{3} \right) \cdot 3 \left| V \left( 011; 000 \right) \right|^2
$$

$$
= \left( \frac{\xi^2}{3} \right) \times 1
$$

which is the square of the angular matrix element. The square of the radial element is tabulated in Condon and Shortley (8) and is $1.66 \, a_o^2$, where $a_o$ is the positronium "Bohr radius."

The matrix element $\left| \langle \hat{n} \rangle_{1k} \right|^2$ in Eq. (35.23) of Schiff (12) is thus $\left( \frac{\xi^2}{3} \right) \times 1$. 

= $1.66 \, a_o^2$. 
APPENDIX IV

Incoherence of Spontaneous Radiation from Perturbed States

Assume spontaneous El radiation occurs from a perturbed state \( \Sigma_s' \) to final states \( \Sigma_{s'j'm'} \), the radiation operator (treated semi-classically) is \( \chi^{(i)} \) a normalized spherical harmonic [Cf. (9)], where we neglect the radial operator. We wish the transition matrix element (using the notation of Dirac (7))

\[
\sum_{j'm's'k} \left| (a | \chi^{(i)}_k | s'j'm') \right|^2
\]

where

\[
(a | = \sum_{lsm} (a | lsjlm) (lsjlm |)
\]

we can write

\[
(a | \chi^{(i)}_k | s'j'm') = \sum_{lsm} (a | lsmjlm) (lsmjlm | \chi^{(i)}_k | s'j'm')
\]

\[
= \sum_{lsmjlm} (a | lsmjlm) \frac{s_{s'}!}{(2j'+1)} (-)^{s_{s'}-j'-j} (4j!j')^{2j'+1} W(xy; s'l'; s) (s'j'm')
\]

by using the Wigner-Eckart theorem to write the sum in terms of a sum over the reduced matrix elements \( (d | \chi^{(i)}_0 | l') \)

\( W (abcd; \alpha \beta) \) is a Racah coefficient. Now
\[
\sum_{m_1 s_1 k} \left( \sum_{l_1 j_1 m} \delta_{ss_1} (-)^l (j_1 m_1 k_1 | j_1 m) \sum_{f_2 g_2 m_2} (m_2 s_2 f_2 | a) (-)^l (j_2 m_2 k_2 | j_2 m) \right) \\
= \sum_{s_1 j_1 m_1} (a | s_1 l_1 j) (s_1 l_1 j | a) (-)^l \\
= \sum_{s_1 j_1 m_1} (a | s_1 l_1 j) (s_1 l_1 j | a) (-)^l (2j_1 + 1) W(l_1 l_1', s 1) \\
\cdot \langle l l' y y' | W(l_1 l_1', s 1) (a | s_1 l_1 j) \rangle \\
= \sum_{s_1 j_1 m_1} \frac{1}{2j_1 + 1} |(a | s_1 l_1 j)|^2 |(a | s_1 l_1 j)|^2 = \sum_{a l s_1 m_1} |(a | l_1 s_1 m_1)|^2 \frac{1}{4\pi^2} (2j_1 + 1) W(l_1 l_1', 1, 00) \right|^2 \\
= \sum_{s_1 j_1 m_1} \frac{1}{8\pi} \frac{3}{(2j_1 + 3)(2j_1 + 1)} \\
\text{where the sum is over the unperturbed components of state } a, \\
\text{and there is no interference. If we wish to consider the} \\
\text{annihilation process from } S \text{ states } (l = 0, j, m) \text{ we add the sum} \\
\sum_{(l_0 j_0 m_0)} |(a | l_0 j_0 m_0)|^2 |H'|^2 
\]
where \((H')\) is the annihilation matrix element from the state \(J = S\).

Notice that in Eq. (5) the non-zero interference terms do not allow us to sum the probabilities of transitions from the components of \((a)\) to a particular final state \(\ell'j'm'\). It is only in the sum over \(\ell'j'm'\) that the cross terms vanish; a particular example of a general sum rule.
APPENDIX V

DETAILS OF COMPUTATION PROGRAM

The positronium atom is assumed to have an energy
\[ E_{\text{eff}} = \frac{1}{2m} (\frac{\mathbf{p}^2}{m} + \frac{\mathbf{p}^2}{m}) \]
with velocity vector perpendicular to the magnetic field. This motion gives rise to the electric field in the atomic rest frame responsible for the Stark effect. The true energy \[ E = \frac{3}{2} E_{\text{eff}} \] includes motion parallel to the magnetic field lines.

The Hamiltonian matrix was constructed using as the diagonal elements the energy eigenvalues for the unperturbed \( n = 2 \) states as calculated to order \( m \omega^5 \) by Fulton and Martin (14). The off-diagonal elements were found from the expressions derived in Appendices I and II. for the range of the parameters of interest: \( H \) and \( v \). This data, plus the radiative widths of the unperturbed states \( \gamma_i \), all expressed in \( \text{Mc}/\text{sec} \) were read into Whirlwind computer on punched tape. The results were displayed by oscilloscope and photographed on microfilm. For each matrix the display consisted of the fourteen perturbed eigenvalues, each with its associated 14-component eigenvector, total radiative width, and two partial widths corresponding to singlet and triplet components for each energy eigenvalue. The results were in \( \text{Mc}/\text{sec} \).

The criteria for the diagonalization procedure was chosen so that no off-diagonal element was larger than 0.1 \( \text{Mc}/\text{sec} \). Thus the perturbed energy eigenvalues are correct to 0.01 \( \text{Mc}/\text{sec} \).
which is more accurate than the initial eigenvalues.

For the values of $E_{\text{eff}} = 0, 0.025, 0.25, 0.50$ e.v. the Zeeman-Stark pattern was computed for values of the magnetic field of 500, 1000, 2000, 3000, 4000, 5000 gauss. These results are displayed graphically in Part I, Sections 2-3. The $M$ values for the pure Zeeman pattern are shown, as $M$ remains a good quantum number at all values of $H$. For $E_{\text{eff}} = 0.025$ e.v. the $M$ values are also shown but are only approximately good quantum numbers for the larger values of $H$. For $E_{\text{eff}} = 0.25$ and 0.50 e.v. these are omitted.

A sample of the radiation width calculations is included at the end of this Appendix. The singlet and triplet radiation widths for $E_{\text{eff}} = 0$ are shown for all levels, for values of $H$ of 500, 2000, and 5000 gauss.

The level crossings in the pure Zeeman pattern are allowed either because the Zeeman operator does not mix the crossed levels or through the vanishing of a matrix element. (See Part I, Section 2.)

For non-vanishing Stark entries none of the level crossings are allowed, as a consequence of the Wigner-von Neumann theorem. The $^{3}S_{1}(m = \pm 1)$ and $^{3}P_{2}(m = 0)$ states are connected by a non-zero matrix element.

The $^{1}P_{1}(m = 0)$ $^{3}P_{1}(m = 0)$ states are not mixed directly, but are mixed in "higher order," e.g., matrix elements connect
successively the states $^1P_1(m = 0) \rightarrow ^3P_0 \rightarrow ^3S_1(m = \pm 1) \rightarrow ^3P_1(m = 0)$.

The effect is to lift the level degeneracies at the crossings. This is clearly evident for the larger values of $E_{\text{eff}}$, but is not shown explicitly for $E_{\text{eff}} = 0.025$ e.v.
Radiative Widths Zeeman Split
n=2 States of Positronium (Mc/sec)

<table>
<thead>
<tr>
<th>State</th>
<th>Unperturbed Width</th>
<th>500 gauss</th>
<th></th>
<th>2000 gauss</th>
<th></th>
<th>5000 gauss</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Singlet</td>
<td>Triplet</td>
<td>Singlet</td>
<td>Triplet</td>
<td>Singlet</td>
</tr>
<tr>
<td>$^3S_1$, m=0</td>
<td>0.29</td>
<td>0.48</td>
<td>0.29</td>
<td>6.79</td>
<td>0.27</td>
<td>26.2</td>
</tr>
<tr>
<td>$^3S_1$, m=±1</td>
<td>0.29</td>
<td>—</td>
<td>0.29</td>
<td>—</td>
<td>0.29</td>
<td>—</td>
</tr>
<tr>
<td>$^3P_2$, m=0</td>
<td>49.5</td>
<td>6.68</td>
<td>42.8</td>
<td>20.9</td>
<td>28.6</td>
<td>24.6</td>
</tr>
<tr>
<td>$^3P_2$, m=±1</td>
<td>49.5</td>
<td>5.78</td>
<td>43.7</td>
<td>20.5</td>
<td>28.9</td>
<td>23.8</td>
</tr>
<tr>
<td>$^3P_2$, m=±2</td>
<td>49.5</td>
<td>—</td>
<td>49.5</td>
<td>—</td>
<td>49.5</td>
<td>—</td>
</tr>
<tr>
<td>$^1P_1$, m=±1</td>
<td>49.5</td>
<td>34.7</td>
<td>14.7</td>
<td>6.57</td>
<td>42.9</td>
<td>1.18</td>
</tr>
<tr>
<td>$^1P_1$, m=0</td>
<td>49.5</td>
<td>41.6</td>
<td>7.8</td>
<td>17.2</td>
<td>32.2</td>
<td>4.2</td>
</tr>
<tr>
<td>$^3P_1$, m=0</td>
<td>49.5</td>
<td>—</td>
<td>49.5</td>
<td>—</td>
<td>49.5</td>
<td>—</td>
</tr>
<tr>
<td>$^3P_1$, m=±1</td>
<td>49.5</td>
<td>9.0</td>
<td>40.5</td>
<td>22.4</td>
<td>27.1</td>
<td>24.5</td>
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<tr>
<td>$^3P_0$</td>
<td>49.5</td>
<td>1.16</td>
<td>48.3</td>
<td>11.4</td>
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<td>20.6</td>
</tr>
<tr>
<td>$^1S_0$</td>
<td>160.0</td>
<td>159.5</td>
<td>0.009</td>
<td>153.</td>
<td>0.01</td>
<td>133.8</td>
</tr>
</tbody>
</table>
APPENDIX VI

SOURCE PREPARATION

The preparation of the radioactive source differed only in minor respects from the procedure of Weinstein (13) and reference is made to that paper for the details.

The active Na$^{22}$Cl with approximately 0.3 mg of carrier was transferred to a Weinstein oven, and dried by passing 1.25 amperes through the 14 turns of #16 Nichrome wire used as the oven heater.

After drying six hours the oven was plugged with a 1/4" long stainless steel plug, 3/16" in diameter, with a 1/16" axial hole, which fitted into the oven flush with the oven top. The material to be evaporated had to emerge through this 1/16" hole.

The silver-plated brass source disc was cleaned with acetone, and one drop of Vinylseal solution allowed to evaporate in the center, leaving a deposit about 1/2" in diameter, $\sim 80$ micrograms/cm$^2$. The source disc was set in place one inch from the oven exit channel. A glass plate with a 3/8" hole, flush against the disc, limited the region of the disc on which active material could be deposited.

With a vacuum $\sim 10^{-5}$ mm (Hg) established, the evaporation was conducted by operating the oven for 15 minutes at 3.5 amps, and 30 minutes at 3.75 amps.
After cooling the vacuum chamber was demounted and the source plate mounted 1¾" from a 2" length of 0.05" tungsten wire whose center had been wound with two inches of .01" gold wire.

After renewing the vacuum, a few seconds of heating the tungsten by the passage of 35 amps sufficed to deposit a barely opaque layer of gold over the active material, a layer estimated of ~100 micrograms/cm².

The source plate received about two-thirds of the initial activity of 25 mc, most of which was in a region ~1/8" in diameter. It remained mechanically stable until it was accidentally exposed to the pulsed r.f. output of the magnetrons. The gold layer then split and curled with some powdering of the active material and contamination of adjacent apparatus. The source was then saturated with Vinylseal solution, which dried to a coating ~1 mg/cm², effectively retaining active material. This allowed the completion of most of the data-taking, but slow deterioration prevented the completion of the program to reduce the experimental errors to negligible amounts.

The present techniques of source preparation are not entirely satisfactory for the sources are not stable against exposure to microwaves or to temperatures >100°C. It is not known at present just what steps should be taken to improve these techniques.
APPENDIX VII

DATA

1. Excitation Runs: register readings (scale 64) of light-on light-off registers corrected for balance error.

<table>
<thead>
<tr>
<th>Light-on</th>
<th>Light-off</th>
<th>% Difference</th>
<th>% Std. Dev</th>
</tr>
</thead>
<tbody>
<tr>
<td>3,631</td>
<td>3,649</td>
<td>-0.05</td>
<td>0.28</td>
</tr>
<tr>
<td>1,198</td>
<td>1,204</td>
<td>-0.05</td>
<td>0.49</td>
</tr>
<tr>
<td>4,913</td>
<td>4,933</td>
<td>-0.01</td>
<td>0.24</td>
</tr>
<tr>
<td>7,846</td>
<td>7,886</td>
<td>-0.04</td>
<td>0.19</td>
</tr>
<tr>
<td>2,732</td>
<td>2,743</td>
<td>-0.02</td>
<td>0.33</td>
</tr>
<tr>
<td>7,993</td>
<td>8,001</td>
<td>-0.01</td>
<td>0.19</td>
</tr>
<tr>
<td>6,870</td>
<td>6,884</td>
<td>-0.02</td>
<td>0.21</td>
</tr>
<tr>
<td>10,000</td>
<td>10,043</td>
<td>-0.04</td>
<td>0.17</td>
</tr>
<tr>
<td>45,185</td>
<td>45,343</td>
<td>-0.04</td>
<td>0.28</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(0.34+0.08) %</td>
<td></td>
</tr>
</tbody>
</table>

2. Check Run: (R.F. on) corrected for balance error.

<table>
<thead>
<tr>
<th>Light-on</th>
<th>Light-off</th>
<th>% Difference</th>
<th>% Std. Dev</th>
</tr>
</thead>
<tbody>
<tr>
<td>17,360</td>
<td>17,377</td>
<td>-0.01</td>
<td>0.12</td>
</tr>
<tr>
<td>27,197</td>
<td>27,261</td>
<td>-0.03</td>
<td>0.09</td>
</tr>
<tr>
<td>23,400</td>
<td>23,406</td>
<td>-0.02</td>
<td>0.10</td>
</tr>
<tr>
<td>24,948</td>
<td>24,994</td>
<td>-0.02</td>
<td>0.10</td>
</tr>
<tr>
<td>92,905</td>
<td>93,037</td>
<td>-0.08</td>
<td>0.10</td>
</tr>
</tbody>
</table>

3. Check Runs: (External source)

<table>
<thead>
<tr>
<th>Light-on</th>
<th>Light-off</th>
<th>% Difference</th>
<th>% Std. Dev</th>
</tr>
</thead>
<tbody>
<tr>
<td>29,354</td>
<td>29,410</td>
<td>-0.02</td>
<td>0.10</td>
</tr>
<tr>
<td>2,201</td>
<td>2,202</td>
<td>-0.03</td>
<td>0.37</td>
</tr>
<tr>
<td>15,001</td>
<td>15,015</td>
<td>-0.04</td>
<td>0.14</td>
</tr>
<tr>
<td>15,001</td>
<td>15,043</td>
<td>-0.04</td>
<td>0.14</td>
</tr>
<tr>
<td>61,557</td>
<td>61,669</td>
<td>-0.02</td>
<td>0.10</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(0.18+0.07) %</td>
<td></td>
</tr>
</tbody>
</table>

- (0.18 ± 0.07)% difference
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(33) R.C.Lord: Private communication
BIOGRAPHICAL NOTE

Henry W. Kendall, son of Mr. and Mrs. H. P. Kendall of Sharon, Massachusetts, entered Amherst College in 1946 following sixteen months with the U. S. Merchant Marine Academy, at Kings Point, New York and overseas. Awarded a B.A. degree (in mathematics) from Amherst in 1950, he entered the Graduate School of Physics at the Massachusetts Institute of Technology that fall. He is a member of Phi Beta Kappa, Sigma Xi, and Delta Kappa Epsilon. He is unmarried.