Effect of an Electric Field on Positronium Formation in Gases: Experimental*†

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The increase in positronium formation due to a static electric field has been measured in various gases including He, Ne, A, H_2, D_2, N_2, and several more complex molecular gases by a study of the energy spectrum of the annihilation γ radiation. For the rare gases the increase has a sigmoid dependence on the ratio of electric field to pressure. The fraction of the positrons that form positronium increases by a maximum factor of 1.5, 1.4, and 2.1 for He, Ne, and A, respectively. A similar increase was observed for H_2, D_2, and N_2. No effect of the electric field was found in the polyatomic gases CO_2, CH_4, C_2H_6, and CCl_2F_2, but a small anomalous decrease in positronium formation was seen in SF_6. Some data are given using a microwave field at 2460 Mc/sec. The detailed theory of the increase given in the following paper provides a basis for obtaining from the observed data values of the elastic scattering cross section of positrons by the rare gas atoms, which are $0.023\pi a_0^2$, $0.12\pi a_0^2$, and $1.2\pi a_0^2 \pm 25\%$ for He, Ne, and A, respectively. Quantitative interpretation for the polyatomic gases is more difficult. At the high electric fields for which positronium formation has its maximum value, the fraction of positrons forming positronium is between 50% and 80% for the rare gases and for H_2 and N_2; the mode of decay of the remaining positrons is not understood.

1. INTRODUCTION

PREVIOUS work on the formation of positronium by positrons stopping in gases has been discussed in several review articles.[1,2] Deutsch[3] first reported the abundant formation of positronium in gases from his studies on the time delay and energy spectrum of the annihilation γ radiation. A basic tool in his studies was the marked effect on the annihilation radiation of a small amount of NO gas, which was interpreted as due to the conversion of orthopositronium to parapositronium in collisions with the paramagnetic NO molecules. Pond[4] measured the fraction of the positrons that form positronium in a gas by observing the effect of NO gas on the two-quantum coincidence rate. From these data, together with the assumption that the ratio of the number of orthopositronium atoms formed to the number of parapositronium atoms formed equals the statistical weight factor of 3, it can be deduced that the fraction of positrons forming positronium in the gases He, A, H_2, and N_2 is between 0.2 and 0.4. De-Benedetti and Siegel[5] compared the amount of positronium formation in various gases by observation of the relative three-quantum coincidence counting rates, and concluded that the fraction of positrons forming positronium in the gases He, H_2, N_2, SF_6, and CCl_2F_2 is the same within $\pm 25\%$.

Ore[6] has given a qualitative discussion of the formation of positronium in gases. Positrons emitted from a radioactive source are slowed down in the gas by ionization and excitation processes until they reach kinetic energies of the order of the ionization energy of the gas, and the last inelastic collision leaves the positrons uniformly distributed in energy. That only several percent of the positrons will annihilate in flight in the slowing down process has been indicated both experimentally[7] and theoretically.[8] From Ore's arguments, it then follows that the fraction ϕ of the positrons that form positronium in a gas will lie in the range

$$(E_{\text{exc}}-E_{\text{thr}})/E_{\text{exc}}<\phi<(E_{\text{ion}}-E_{\text{thr}})/E_{\text{ion}},$$

in which E_{ion} is the ionization energy of the gas atom, E_{exc} is the excitation energy of the lowest excited state of the atom, and E_{thr} is the threshold energy for positronium formation ($E_{\text{thr}}=E_{\text{ion}}-6.8$; 6.8 ev is the binding energy of the ground state of positronium). The experimental data on all gases studied fall within the theoretical limits calculated from this expression. Any positron which falls to an energy below E_{thr} will be annihilated in a collision with an electron in a gas atom without the formation of positronium. Since the cross section for annihilation of a free positron is small compared to the cross sections for ionization, excitation, or positronium formation when these latter processes are energetically possible, all except several percent of the annihilation of free positrons is expected to occur for positrons with energies below E_{thr}.

The increase in positronium formation due to an electric field was first observed by Deutsch and Brown[9] during their radio-frequency measurement of the Zeeman effect of positronium. They suggested that the enhanced formation is due to the gain of kinetic energy from the

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[1] M. Deutsch, Progr. Nuclear Phys. **3**, 131 (1953).

[2] S. DeBenedetti and H. C. Corben, in *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1954), Vol. 4, p. 191.

[3] M. Deutsch, Phys. Rev. **82**, 455 (1951).

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[6] A. Ore, Universitetet i Bergen Arbok 1949, Naturvitenskapelig rekke No. 9.

[7] Gerhart, Carlson, and Sherr, Phys. Rev. **94**, 917 (1954); H. W. Kendall and M. Deutsch, Phys. Rev. **101**, 20 (1956).

[8] H. A. Bethe, Proc. Roy. Soc. (London) **A150**, 129 (1935).

[9] M. Deutsch and S. C. Brown, Phys. Rev. **85**, 1047 (1952).

rf electric field by positrons which fall to a kinetic energy below the threshold energy for positronium formation. In the absence of an electric field, all such positrons would annihilate with electrons in the gas atoms without any formation of positronium; in the presence of an electric field these positrons can gain energy from the field to cross the threshold energy for positronium formation, and thus an increase in positronium formation occurs. Deutsch[1] has also reported briefly that an increase in positronium formation was observed upon application of a static electric field.

The detailed investigations in this and the following paper[10] on the effect of an electric field on positronium formation were undertaken to obtain a better understanding of the behavior of positrons in a gas. Such understanding is helpful to experiments on the fine structure of positronium and to searches for the excited states of positronium.

This paper presents the experimental observations on the effect of an electric field on positronium formation in various gases including He, A, Ne, H_2, D_2, N_2, and several more complex molecular gases. The majority of the data were taken using a static electric field, but some data were obtained using a microwave field at about 2460 Mc/sec. The experiment consists in observing the shape of the energy distribution of the annihilation γ radiation as a function of the applied electric field. From these data the change in the fraction of positrons that form positronium as a function of the electric field is obtained. The accompanying paper by Teutsch and Hughes gives a theory of the effect of an electric field on positronium formation in gases. The experimental results can be interpreted with the aid of this theory to yield a value for the elastic scattering cross section of a positron by a gas atom.

2. APPARATUS AND PROCEDURE

The apparatus used in this experiment was basically the same as that used by Hughes, Marder, and Wu,[11] and only the few differences will be noted here. A cavity of the same external dimensions was used, but the interior was modified to allow for the application of a uniform electric field. A static voltage was applied to two parallel copper plates which are 10 cm in diameter and are separated by a distance of 4 cm. These field plates were insulated from the cavity by Teflon sheets which had circular depressions to hold the field plates, and the voltage leads were brought into the cavity through two Kovar to glass seals. The Cu^{64} source was the same as in the experiment of HMW, but was set into a depression $\frac{1}{2}$ in. in diameter and $\frac{1}{32}$ in. deep at the center of one of the field plates and then held in place by a copper spring. No internal lead shielding could be used inside the cavity, because of the distortion this would introduce in the electric field.

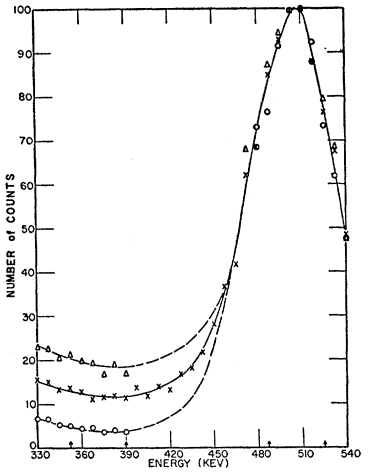

FIG. 1. Positron annihilation spectra. Number of counts, normalized to 100 at the peak, *vs* γ-ray energy for Cu^{64} source in argon at 1.2 atmos with a magnetic field of 7200 gauss. For the experimental curves with crosses and open triangles, the electric field had the values 0 and 706 volts/cm, respectively; for the curve with open circles an NO gas concentration of 6% was used. The peak region from 485 to 530 kev and the valley region from 345 to 390 kev are indicated by arrows. A point in the valley represents approximately 480 counts in the case of argon alone, and approximately 170 counts in the case of argon plus 6% NO.

In order to obtain reproducible results, it was found necessary to observe certain precautions to insure cleanliness of the cavity and purity of the gas. The size of the pumping line to the cavity was increased, and a liquid air trap was inserted. After the source was placed in the cavity, the system was pumped for about three hours to a pressure of about 5×10^{-5} mm of Hg, and on some occasions further removal of residual gas was attempted by applying 3000 volts to the field plates. The cavity was then filled with gas and the system was shut off from the gas supply. The helium and argon gases used were of commercial grade with a minimum purity of 99.9%. In order to obtain reproducible results for argon it was found helpful to pass the gas into the system through a calcium purifier. Research grade neon was used with an impurity of less than 1 part in 10^4. All of the other gases employed were of commercial grade.

A magnetic field of between 5000 and 7500 gauss, whose direction was parallel to that of the electric field, was employed for almost all the measurements, because it focused the positrons in the center of the cavity. This focusing effect increased the counting rate due to positron annihilations in the gas, and decreased the background counting rate due to positron annihilations in the cavity walls. Furthermore it maintained the positrons in the region of homogeneous electric field.

The energy spectrum of the annihilation γ radiation was observed for a given gas as a function of the applied electric field, and, in addition, the annihilation spectrum

[10] W. B. Teutsch and V. W. Hughes, Phys. Rev. **103**, 1266 (1956), following paper.

[11] Hughes, Marder, and Wu, Phys. Rev. **98**, 1840 (1955). This paper will be referred to hereafter as HMW.

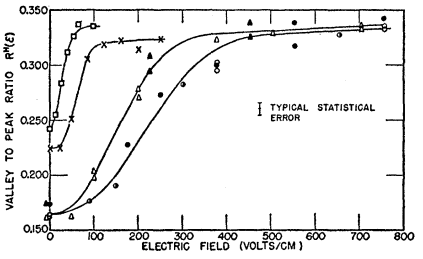

FIG. 2. Valley to peak ratio $R^H(\mathcal{E})$ of γ-ray spectrum *vs* electric field for the rare gases, experimental relationships. Open circles, full circles, and half-full circles are for argon at a pressure of 2.0 atmos; open and full triangles are for argon at 1.2 atmos; crosses are for neon at 2.0 atmos; squares are for helium at 2.7 atmos.

was observed when a small amount of NO was added (partial pressure of the NO from 1% to 5%). Typical spectra are shown in Fig. 1 for the case of argon at a pressure of 1.2 atmospheres. The curves have been normalized to the value of 100 at the peak, and, except for the curve involving the electric field, are of the type discussed with reference to Fig. 2 of HMW.

3. ANALYSIS OF DATA AND RESULTS

The application of the electric field is seen (Fig. 1) to cause an increase in the number of counts in the valley energy region (345 to 390 kev) relative to the number of counts in the peak energy region (485 to 530 kev). This increase is interpreted as an increase in the number of three quantum decays from orthopositronium (whose γ-ray energies are lower than 510 kev) relative to the number of two-quantum decays, and hence as an increase in the fraction of positrons that form positronium.

Figure 2 shows the observed ratio $R^H(\mathcal{E})$ of the number of counts in the valley energy region to the number of counts in the peak energy region as a function of the applied electric field $\mathcal{E}$ for the gases helium, neon, and argon. In all cases the value of $R^H(\mathcal{E})$, which is a measure of the amount of positronium formation, increases with electric field in a sigmoid fashion to a maximum value. The amount of the increase in $R^H(\mathcal{E})$ and the electric field region in which the increase occurs differ for the different gases. The effect of pressure is indicated by the argon data at 1.2 and 2.0 atmos.

To obtain an expression for the fraction of positrons forming positronium, $\phi(\mathcal{E})$, in terms of the experimentally observed quantities, the following equations are written for the number of counts appearing in the peak energy regions under the various experimental conditions. For the number of counts in the peak energy region:

$$P^H=N_p{}^H[g_p+Kg_p+(FG_0+2G_1)g_0+(1-F)g_p], \quad (1a)$$

$$P^H(\mathcal{E})=N_p{}^H(\mathcal{E})[g_p+K(\mathcal{E})g_p+(FG_0+2G_1)g_0+(1-F)g_p], \quad (1b)$$

$$P^0=N_p{}^H[4+K], \quad (1c)$$

$$P^0(\mathcal{E})=N_p{}^H(\mathcal{E})[4+K(\mathcal{E})]. \quad (1d)$$

There will be corresponding expressions for V^H, $V^H(\mathcal{E})$, V^0 and $V^0(\mathcal{E})$ in which there will appear H_0 and H_1 instead of G_0 and G_3 and h_p and h_0 instead of g_p and g_0. These equations are almost identical to Eqs. (8) of HMW, and the discussion of notation and justification of the equations given in HWM will not be repeated in detail here. All of these equations apply to measurements made with a strong magnetic field applied. Equation (1a) holds for the condition of pure gas and no applied electric field, and Eq. (1b) holds for the condition of pure gas and an applied electric field. Equations (1c) and (1d) are the corresponding expressions when a small amount of NO is added. The quantity g_p and the quantities g_0G_0 and g_0G_1 are the probabilities that an annihilation γ ray from parapositronium and orthopositronium decays, respectively, will be counted in the peak region of the energy spectrum, in which G_0 and G_1 account for the difference in angular distribution of the annihilation quanta from the different magnetic substates of orthopositronium. The quantity F is the fraction of $M=0$ orthopositronium atoms which decay by three-quantum annihilation in the presence of the magnetic field H.

The quantities $N_p{}^H$ and $N_p{}^H(\mathcal{E})$ are the number of parapositronium atom annihilations with and without an electric field present respectively. It is assumed that orthopositronium and parapositronium atoms are formed in the statistical ratio of 3 to 1 under all experimental conditions. The quantities K and $K(\mathcal{E})$ are the ratios of the number of positrons which annihilate directly to the number of positrons which form parapositronium with and without an electric field present respectively. Hence the fraction of positrons which form positronium when an electric field is present, $\phi(\mathcal{E})$, is given by: $\phi(\mathcal{E})=4/[4+K(\mathcal{E})]$. The quantity $\Phi=\phi(\mathcal{E})/\phi(0)$, which is the ratio of the fraction of positrons that form positronium in an electric field to the fraction that form positronium in zero field, is the quantity obtained from the measurements. Under the assumption that the number of positrons annihilating in the volume viewed by the scintillation spectrometer is independent of the electric field, Eqs. (1) can be solved without approximation for Φ to yield:

$$\Phi=\frac{P^H(\mathcal{E})}{P^H}\frac{R^H(\mathcal{E})-R_0}{(R^H-R_0)}. \quad (2)$$

It is to be noted that Φ is determined entirely by quantities obtained from the annihilation spectra, and hence a knowledge of probability factors, angular distribution corrections, and the fraction of orthopositronium quenched by the magnetic field is not

required. If it is assumed that K and $K(\mathcal{E})$ do not depend on the magnetic field, then Φ will be independent of H. This assumption only requires that the mechanisms of the formation of positronium and of the energy gain by the positrons from the electric field be independent of the magnetic field. Furthermore, Φ will be independent of any collision quenching of orthopositronium, provided such quenching is independent of $\mathcal{E}$.

Many factors must be considered in deciding upon the accuracy with which $\Phi(\mathcal{E})$ is determined by the measurements. The statistical probable error in the R and P values associated with the number of counts was between 1.5 and 2.5% for all gases and for all values of the electric field. Because no internal lead shielding could be used in the cavity owing to the need for a uniform electric field, background wall counts were larger than for the experiment of HMW. If the background counting rate is defined as one-half of the counting rate observed when the cavity is evacuated, it amounts to between 2.5 and 14% of the total counting rate for the various peak and valley readings taken. Still Φ depends only on quantities which are ratios of observed counts, and the background effects in the numerator and denominator of each ratio tend to cancel, so that the error introduced in Φ is approximately that of the statistical counting error.

The positrons were collimated by the strong magnetic field of 5000 to 7500 gauss within about 2 cm of the field axis, so that edge-effect inhomogeneities in the electric field were less than 2%. The electric field was known to about 2% in view of the meters used and the deviations of the actual geometry from plane parallel plates. It will be recalled that the derivation of the expression (2) for Φ assumed that the electric field did not affect the number of positron annihilations occurring in the region of the cavity viewed by the scintillation spectrometer. It might be expected that this assumption is valid because the applied potentials (<8 kv) were small compared to the initial energies of the positrons (for Cu^{64} the maximum kinetic energy is

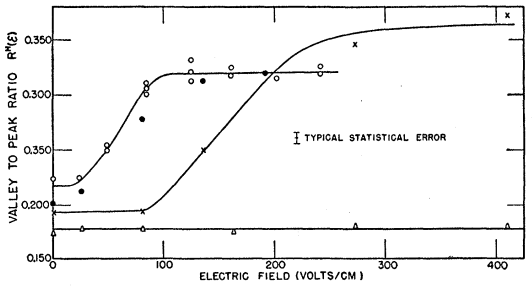

FIG. 3. Figure illustrating the effect of impurities in neon. Valley to peak ratio $R^H(\mathcal{E})$ of γ-ray spectrum *vs* electric field, experimental relationships. Open and full circles show data obtained with neon at a pressure of 2.0 atmos; crosses show data obtained in neon which has a contamination of CCl_2F_2 of the order of one part in 10^4 at a total pressure of 2.0 atmos; triangles show data obtained in a mixture of neon plus 3% by volume of CCl_2F_2 at a total pressure of 2.0 atmos.

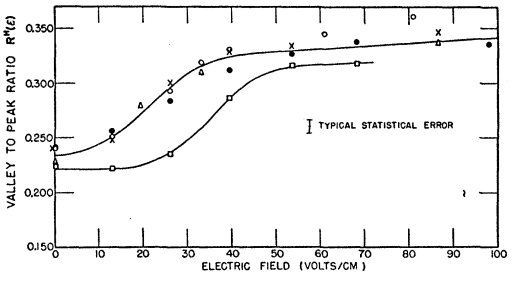

FIG. 4. Figure illustrating the reproducibility of data on helium. Valley to peak ratio $R^H(\mathcal{E})$ of γ-ray spectrum *vs* electric field, empirical relationships. Each symbol indicates data obtained from a different filling of the cavity with helium at a pressure of 2.7 atmos.

650 kev), and, furthermore, when the positrons have been slowed down their drift velocities due to the electric field are too small for them to move an appreciable distance during their lifetime. The validity of this assumption was supported by the experimental observation that no variation in the various counting rates was seen when the polarity of the electric field was changed. The pressure was measured with an Ashcroft pressure gauge, calibrated against a mercury manometer, and hence is known to about 1%.

Slight impurities in the gas were found to be a principal cause of lack of reproducibility in the experimental data. Deutsch[1] has pointed out that polyatomic gas impurities can suppress the enhancing effect of the electric field on positronium formation, and the explanation of this effect will be discussed further in the next section and in the following paper. The suppression of the electric field effect in neon by small amounts of freon is shown in Fig. 3. The gases SF_6 and NO were found to produce a similar effect. On the other hand, admixture of a few percent of argon with neon or the reverse did not noticeably affect the electric field effect characteristic of neon or argon, respectively. Because of this pronounced effect of polyatomic impurities, the precautions on cleanliness of the system and purity of the gas mentioned in Sec. 2 were taken. Typical reproducibility of data on $R^H(\mathcal{E})$ *vs* $\mathcal{E}$ is illustrated for five different runs on helium (Fig. 4). Except for the one run, the reproducibility from run to run is within the statistical error of the points. The anomalous run is shifted in the direction to be expected for polyatomic impurities. Data were taken for neon both with and without the use of the calcium purifier and no difference was observed. Particular difficulty was experienced with argon, for which an increase of R^H with time was often observed. This drift was reduced considerably, and in some cases eliminated entirely, by passing the argon gas into the system through a calcium purifier. This observed drift corresponds to an increase in positronium formation with time and is presumably due to some evolution of polyatomic impurity. Data with argon were used only if no drift in R^H was present.

The final results on the increase in Φ with increase

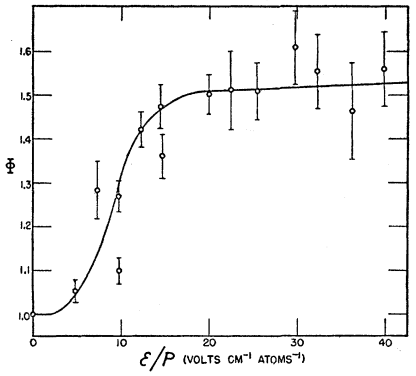

FIG. 5. The ratio Φ of the fraction of positrons that form positronium in an electric field to the fraction that form positronium in zero field as a function of the ratio of electric field to pressure, empirical relationship for helium. Values are obtained using the data of Fig. 4.

in $\mathcal{E}/P$ are shown in Figs. 5, 6, and 7. The values of Φ were calculated from the observed quantities for the various runs taken (2 to 4 runs for each gas) by use of Eq. (2), and the errors indicated represent our estimate of the experimental error, which arises primarily from statistical counting errors and from background effects. It is seen that the probable error in Φ is about 6%. Any systematic error due to impurities could not, of course, be included in this estimate. Argon and neon data were taken at pressures of 1.2 and 2.0 atmospheres, and for both gases the data for the two pressures lie on the same curve when the abscissa is taken to be $\mathcal{E}/P$. The maximum values obtained for Φ were 1.53±0.06, 1.41±0.03, and 2.08±0.12 for He, Ne, and A, respectively. For the polyatomic gases only data on $R^H(\mathcal{E})$ *vs* $\mathcal{E}$ were obtained (Figs. 8 and 9). The increase in positronium formation with $\mathcal{E}$ for H_2, D_2, and N_2 is similar in form to that for the rare gases. The more complex gases exhibit no effect at all except in the anomalous case of SF_6 in which a decrease in positronium formation is observed with increase in the electric field.

Some few data were also obtained on the effect of a radio-frequency electric field on positronium formation

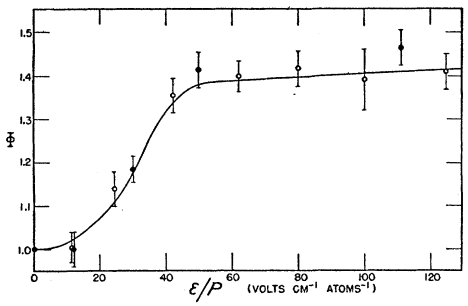

FIG. 6. The ratio Φ *vs* the ratio of electric field to pressure for data obtained in neon, empirical relationship. Open circles are obtained at a pressure of 2.0 atmos and closed circles at a pressure of 1.2 atmos.

in gases during the course of a measurement of the Zeeman effect of positronium in an experiment similar to that of Deutsch and Brown.[12] Radio-frequency power from a 1-kilowatt CW magnetron operating at about 2460 Mc/sec was fed into a high Q cavity filled with the gas under study, and the power in the cavity was measured with a bolometer fed from an output loop of known dimensions inserted in the cavity. From this power measurement, it was deduced that the rms electric field in the region of the cavity viewed by the spectrometer was known to within about a factor of 2. A magnetic field of about 8000 gauss was used. For argon at a pressure of 1.95 atmospheres, the ratio $R^H(\mathcal{E})/R^H(0)$ (which will be approximately equal to Φ) was found to be 1.46±5%. For SF_6 at a pressure of 1.95 atmospheres, $R^H(\mathcal{E})/R^H(0)$ was 0.98±5%.

In addition to their use in the measurement of the relative value $\Phi(\mathcal{E})$, the experimental data also determine the absolute value, $\phi(\mathcal{E})$, of the fraction of positrons which form positronium in a field $\mathcal{E}$ although the

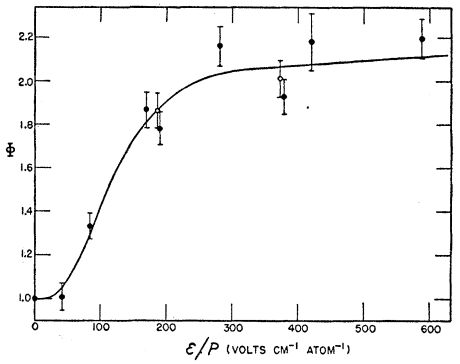

FIG. 7. The ratio Φ *vs* the ratio of electric field to pressure for data obtained in argon, empirical relationship. Open circles are obtained at a pressure of 2.0 atmos and closed circles at a pressure of 1.2 atmos.

latter quantity is not determined as accurately as is the quantity Φ. Equations (1b) and (1d) can be solved to yield

$$\phi(\mathcal{E})=\frac{4[P^0(\mathcal{E})-P^H(\mathcal{E})]}{P^0(\mathcal{E})[(2+F)-\frac{1}{2}(FG_0+2G_1)]}.$$

The quantities F, G_0, and G_1 can be computed theoretically from Eqs. (3), (4), (5), and (9) of HMW, and the quantities $P^0(\mathcal{E})$ and $P^H(\mathcal{E})$ are experimental observables. Values for $\phi(0)$ of 0.55±0.06 and 0.36±0.06 were obtained for neon and argon, respectively, at a pressure of 1.2 atmospheres. The value for argon is in agreement with the result of Pond[4]; no previously reported value for neon has been found in the literature. Because of the small positron stopping power of helium, no reliable value was was obtained for this gas.

[12] M. Deutsch and S. C. Brown, Phys. Rev. **85**, 1047 (1952). Our Zeeman effect measurement will be described in detail in a later publication.

4. DISCUSSION AND CONCLUSIONS

A simple discussion of the theory will now be presented in order that certain general points can be made and in order that conclusions based on the comparison of the experimental results with the more complete theory of the accompanying paper can be drawn. It was pointed out in Sec. 1 that the increase in positronium formation due to an electric field was believed to be due to the gain of kinetic energy from the electric field by positrons which had fallen to a kinetic energy below the threshold energy for positronium formation and the consequent possibility that these positrons will form positronium.

In the absence of an electric field a positron which has a kinetic energy less than the threshold energy for positronium formation will eventually annihilate with an electron in a gas atom. The cross section for this process is so small compared to the cross section for elastic scattering that a positron will make on the

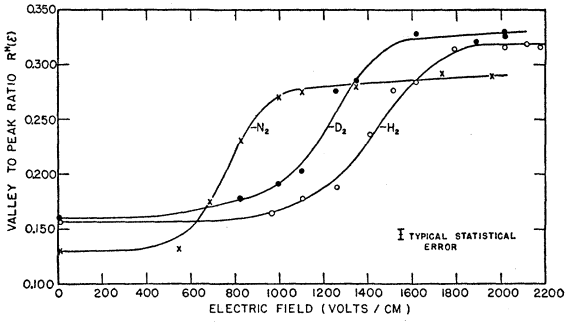

FIG. 8. Valley to peak ratio $R^H(\mathcal{E})$ of γ-ray spectrum *vs* electric field for the gases H_2, D_2, and N_2. Data for H_2 and D_2 were obtained at a pressure of 2.7 atmos, and data for N_2 at a pressure of 2.0 atmos.

average some 10^4 to 10^6 elastic collisions and thus attain thermal energy before it annihilates.

In the presence of an electric field, the positron can gain or lose energy from the field between elastic collisions. It experiences an acceleration $\mathbf{a}=e\mathcal{E}/m$ in which e is the positron charge, m is the positron mass, and $\mathcal{E}$ is the electric field, and hence the change in energy between collisions is

$$\tfrac{1}{2}m(\mathbf{v}+\mathbf{a}t)^2-\tfrac{1}{2}mv^2=e\mathcal{E}\cdot\mathbf{l}+(e\mathcal{E}l)^2/2mv^2,$$

in which t is the time between collisions, v is the velocity at the beginning of the free path, and l is the length of the free path. The expression on the right-hand side follows from the expression on the left-hand side provided the change in velocity due to the field is small compared to the initial velocity. The term $e\mathbf{E}\cdot\mathbf{l}$ can be either a gain or loss of energy depending on the relative direction of $\mathbf{E}$ and $\mathbf{v}$, and, indeed, its average value over many free paths will be zero. On the average, however, a positron gains a small amount of energy from the field association with the term $(e\mathcal{E}l)^2/2mv^2$. If this energy gain is equated to the average energy loss

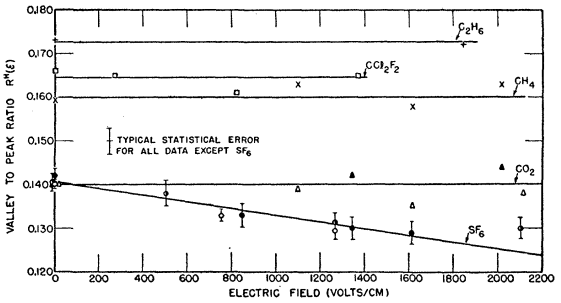

FIG. 9. Valley to peak ratio $R^H(\mathcal{E})$ of γ-ray spectrum *vs* electric field for the gases C_2H_6, CCl_2F_2, CH_4, CO_2, and SF_6. Data for C_2H_6 and CH_4 were obtained at a pressure of 2.7 atmos; for CCl_2F_2 and SF_6 at 2.0 atmos; for CO_2 at 1.2 and 2.3 atmos.

$(2m/M)(\frac{1}{2}mv^2)$ in an elastic collision, then it can be seen that the positrons tend towards an equilibrium energy of the order of

$$(M/m)^{\frac{1}{2}}e\mathcal{E}l.$$

And, indeed, the positrons will attain an energy distribution with a mean energy of this order, and with a spread in energy due to the random term $e\mathcal{E}\cdot\mathbf{l}$. Increase in positronium formation occurs with increase in the electric field because the positron energy distribution is shifted towards higher energies, and there is then an increased probability that a positron will cross the threshold energy for positronium formation before it annihilates with an electron in a gas atom. The characteristic term is $e\mathcal{E}l$ and since $l=1/N\sigma_e$, in which N is the number of atoms/cm^3 and σ_e is the elastic scattering cross section, it is clear that the appropriate experimental variable is $\mathcal{E}/P$ (P being the pressure), in agreement with the experimental observations. Such a variable is characteristic for gas kinetic processes. Further, it is seen that the elastic scattering cross section is a very important parameter.

The following paper treats the changes in energy of the positrons by a diffusion equation derived from the Boltzmann equation. With this equation the increase in positronium formation as a function of the applied electric field and gas density is calculated. The result describes correctly the experimental observations and yields information on the pertinent atomic cross sections. In particular, for the rare gases helium, neon, and argon it can be concluded that the annihilation cross section, σ_a, for a positron incident on a gas atom is $\zeta\pi r_0^2c/v$ in which the parameter ζ is equal to the atomic number of the atom to within about a factor of two. (The quantity $\pi r_0^2c/v$ is the nonrelativistic Dirac annihilation cross section per electron, in which r_0 is the classical electron radius and v is the relative velocity of the positron and electron.) The cross section for the elastic scattering of a positron by a gas atom, which is the most sensitive quantity in the theory, is determined to have the following values for the rare gases: $0.023\pi a_0^2$, $0.12\pi a_0^2$, and $1.5\pi a_0^2\pm25\%$ for

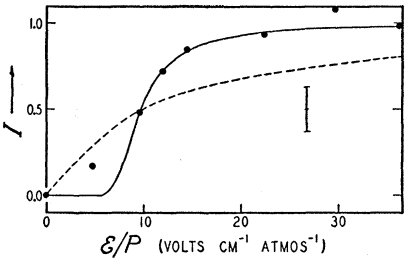

FIG. 10. Increase in positronium formation I *vs* the applied electric field divided by the pressure, for helium. [$I=(\Phi-1)/(\Phi_{max}-1)$, in which Φ is the fraction of positrons that form positronium in an electric field divided by the fraction that form positronium in zero field.] The points are computed from the experimental data of Fig. 5. The dashed curve is computed from the simple theory associated with Eq. (4), in which $\sigma_a=2\pi r_0^2c/v$ and $\sigma_e=0.03\pi a_0^2$. The solid curve is computed from the theory of the following paper, in which $\sigma_a=2\pi r_0^2c/v$ and $\sigma_e=0.023\pi a_0^2$.

helium, neon, and argon, respectively. Figure 10 shows the agreement of theory and experiment for helium. The comparison of theory and experiment for the other rare gases is given in the following paper. It is to be noted that these cross sections are about an order of magnitude smaller than the corresponding elastic scattering cross sections for electrons.[13] Theoretical estimates in the following paper indicate that this is to be expected because of the change in the sign of the interaction potential for the positron.

It is interesting to make a simple calculation of the effect of an electric field on positronium formation which neglects the terms $(e\mathcal{E}l)^2/2mv^2$ and $(2m/M)\times(mv^2/2)$. Then the positron is pictured as executing a simple random walk in energy space, gaining and losing energy in units of $e\mathcal{E}l$. The question of interest is what is the probability that a positron which has fallen to some kinetic energy below the threshold energy for positronium formation shall cross the threshold energy before it annihilates with an electron in a gas atom. It will be assumed that if the positron crosses the threshold energy it will form positronium. This assumption will be a good one provided the cross section for positronium formation is large compared to the elastic scattering cross section. Hence the problem is that of a random walk with an absorber barrier at the threshold energy.

The probability that a positron which lands at an energy E at time $t=0$ shall cross the formation threshold energy E_{thr} between times t and $t+dt$ is given by[14]:

$$dP=(e^{-\lambda_a t})\left[\frac{E_{thr}-E}{2t(\pi Dt)^{\frac{1}{2}}}\exp\left(\frac{-(E_{thr}-E)^2}{4Dt}\right)\right]dt, \quad (3)$$

in which $e^{-\lambda_a t}$ is the probability that the positron has survived direct annihilation with λ_a being the direct annihilation rate in the gas. The bracketed expression is the probability that a positron in the absence of free annihilation shall cross the threshold energy for the first time in the time interval t to $t+dt$, in which $D=(\lambda_e/2)(e\mathcal{E}l)^2$ and $\lambda_e=Nv\sigma_e$, where $\lambda_e=$frequency of elastic collisions. The validity of this expression requires that σ_e be independent of energy and also that λ_e be independent of energy. These two requirements are inconsistent because of the relationship $\lambda_e=Nv\sigma_e$. However, a result which is correct in order of magnitude will be obtained by use of some average value of λ_e in formula (3). When the expression (3) is integrated over time and averaged over the energy region from 0 to E_{thr}, an expression is obtained for the fraction I of the positrons reaching this energy region that will form positronium in the presence of an electric field:

$$I=(1-e^{-y})/y,$$
$$y=(2\sigma_a/\sigma_e)^{\frac{1}{2}}(E_{thr}/e\mathcal{E}l). \quad (4)$$

In taking the average over energy it is assumed that positrons have equal probability of landing in any energy interval between 0 and E_{thr}.

The expression (4) for the increase in positronium formation has the general form of the experimental curves. The theoretical curve is 0 at $\mathcal{E}=0$ and rises to a saturation value of 1 at $\mathcal{E}=\infty$. Figure 10 shows this curve fitted to the experimental data for helium by taking $\sigma_a=2\pi r_0^2c/v$ and $\sigma_e=0.03\pi a_0^2$. A value of v corresponding to a kinetic energy of 10 ev was used. The value of σ_e was chosen to make the theoretical curve agree with the experimental curve at the halfway point in the rise. These values of the cross sections are in reasonable agreement with the values obtained by comparison with the theory of the accompanying paper.

The interpretation of the experimental data on the polyatomic gases is considerably less certain because of their low-lying energy levels, including rotational, vibrational, and electronic levels, which can be excited by inelastic collisions with a positron whose energy is below the threshold energy for positronium formation. This problem is discussed in the accompanying paper, and on the basis of the observed difference in the effect of an electric field on positronium formation in H_2 and D_2, some evidence is presented that for these gases excitation of vibrational states is an important process. For the gases CO_2, CH_4, C_2H_6, and CCl_2F_2, no increase in positronium formation was observed up to the maximum electric fields used (approximately 2000 volts/cm at pressures of about 2 atmospheres). Thus in these gases energy losses in collisions, presumably involving excitation of low-lying electronic levels, still compete favorably with energy gain from the electric field even at the highest fields used, and prevent the positrons from reaching the threshold for positronium formation before they annihilate as free positrons. The fact that the energy loss in a collision of a positron with a molecule of a polyatomic gas is large compared to the recoil energy loss in an elastic collision with an atom of a

[13] H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Oxford University Press, New York, 1952).
[14] S. Chandrasekhar, Revs. Modern Phys. **15**, 1 (1943).

monatomic gas suggests why minute traces of such polyatomic gases strongly suppress the enhancing effect of the electric field on positronium formation in those gases which do exhibit an increase.

The present experiment measures primarily the ratio Φ of the fraction of positrons which form positronium in an electric field to the fraction which form positronium in zero field, although somewhat less accurate values for $\phi(0)$, the fraction of the positrons which form positronium in zero electric field are also obtained. If the values of 1.53, 1.41, and 2.08 for helium, neon, and argon are combined with measurements of the fraction of positrons which form positronium in the absence of any electric field, which is 0.34 for helium,[4] 0.55 for neon (Sec. 3 of this paper), and 0.33 for argon[4] (Sec. 3 of this paper), it is found that at high electric fields the fractions 0.52 ± 0.05, 0.78 ± 0.09, and 0.70 ± 0.08 of the positrons form positronium in helium, neon, and argon, respectively. For H_2 and N_2, the complete determination of Φ was not made, but data on the rare gases indicate that the quantity $R^H(\mathcal{E})/R^H$ is a good approximation to Φ. If the maximum values of $R^H(\mathcal{E})/R^H$, equal to 2.0 and 2.2 for H_2 and N_2, are used for Φ_{max} and combined with Pond's values of 0.38 and 0.23, it is concluded that at high electric fields the fraction of positrons forming positronium is 0.76 and 0.51 for H_2 and N_2, respectively. Hence it is indicated that rather generally only about $\frac{1}{2}$ to $\frac{3}{4}$ of all the positrons form positronium at the high fields.[15]

General theoretical considerations given in Sec. 1 indicate, however, that except for several percent of the positrons which annihilate in the slowing-down process, all the positrons which do not form positronium will annihilate as slow positrons with electrons in the gas atoms. At sufficiently high electric fields all of the positrons will be accelerated to velocities above the threshold for positronium formation, and hence the observed saturation of the increase in positronium formation at large values of the electric field should correspond to positronium formation by all the positrons in the gas except the few that decayed in flight. Possible sources of the discrepancy are: (1) the formation of positron compounds and hence another mode of decay of positrons,[16] (2) annihilation of parapositronium atoms formed from positrons with energies above the ionization energy of the atom,[17] (3) failure of the formation of orthopositronium and parapositronium in the statistical ratio of 3 to 1, perhaps by positrons which are accelerated by the field and form positronium just above the threshold energy. Four recent experiments by Dulit, Gittelman, and Deutsch[18] have indicated that the addition of even small amounts of certain polyatomic impurities changes the fraction of positrons that form positronium at zero electric field. Thus in the cases of He, H_2, and N_2 the combination of results from different experimenters should be viewed with caution. Further experimental evidence is needed to clarify these questions.

A decrease in positronium formation was observed with increase in electric field for the gas SF_6 alone, as shown in Fig. 9. A decrease was also observed with no magnetic field present and at a pressure of about $\frac{3}{4}$ atmospheres with a strong magnetic field. The experimental data were not accurate enough to judge the dependence of this decrease on magnetic field or on pressure. Part of the motivation for the data taken on other polyatomic molecules shown in Fig. 9 was to seek some understanding of this effect. None of the other gases exhibited the effect. These observations on SF_6 remain unexplained.

The interpretation of the experimental data taken with the radio-frequency electric field can be made with only slight modifications in the theory discussed for the effect of a static electric field. Since the period of the radio-frequency field is short compared to the lifetime of the positrons in the gas but long compared to the elastic collision time, it is only necessary to replace the field $\mathcal{E}$ appearing in Eq. (4) by the rms value of the radio-frequency field. And, indeed, if the value of Φ obtained for argon and SF_6 is plotted *vs* $\mathcal{E}$, the points fall on the static field curve of Figs. 6 and 9 to well within the experimental error.

[15] S. DeBenedetti and H. C. Corben, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1954), Vol. 4, p. 191. Our result disagrees with a statement in this review article that at high fields 100% of the positrons form positronium.

[16] J. A. Wheeler, Ann. N. Y. Acad. Sci. **48**, 219 (1946); A. Ore, Phys. Rev. **73**, 1913 (1948); A. Ore, Universitetet i. Bergen Arbok 1952, Naturvitenskapelig rekke No. 5.

[17] C. B. O. Mohr, Proc. Phys. Soc. (London) **A68**, 342 (1955).

[18] Dulit, Gittelman, and Deutsch, Bull. Am. Phys. Soc. Ser. II, **1**, 69 (1956).