Strong Electric Field Experiments on Positronium in Gases*†

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The increase in positronium formation on applying a dc electric field is measured using a two-photon angular correlation method as well as pulse-height analysis. In argon and in nitrogen the field dependence at pressures ranging up to 28 atmospheres is consistent with results of other investigators. In oxygen an increase by a factor of 2 is demonstrated, the field and pressure dependence having the same characteristics as for argon and nitrogen. In sulfur hexafluoride this normal increase in positronium formation is produced, requiring for given density an order of magnitude higher fields than in these other gases; also, the main features of an abnormal decrease in formation with increasing field which occurs at comparatively low fields are determined. A certain range-energy systematic effect crucial to field reversal experiments is explored empirically, and it is concluded that the earlier reported $\boldsymbol{\sigma}\cdot\mathbf{E}$ effect for positronium can be traced in large part to this effect.

I. INTRODUCTION

THE application of strong electric fields to positrons and/or positronium became of interest in late 1957 for the purpose of examining into whether a hypothetical energy term $\xi\boldsymbol{\sigma}\cdot\mathbf{E}$—involving the product of positron or electron spin with an electric field—was effectively zero or not.

Formation and decay of positronium in fluid media is perhaps better understood than in crystalline surroundings. The work to be presented here concerns annihilation in gases only. This research was undertaken primarily to obtain a better understanding of a certain asymmetry previously reported[1] which seemed to imply a nonzero value of ξ.

It has been known for some time that when positrons have been stopped in certain gas samples the fraction which will form positronium can be increased due to an applied static electric field.[2–5] Therefore in the present work considerable contact is necessarily made with such an increase in formation. It is here studied by means of pulse-height analysis of single annihilation photons and by means of two-photon angular correlation, the latter method of measurement being applied for the first time to the detailed aspects of this phenomenon. Measurements made on argon and on nitrogen are compared with the work of the Columbia group.[4] Results are given for the increased formation in oxygen, which for practical purposes is accessible only to the angular correlation technique. The increased formation is identified in sulfur hexafluoride, where it has been separated from an anomalous decrease in formation for this gas, which was discussed by Marder *et al.*[4]

A range-energy systematic effect associated with reversal of $\mathbf{E}$ with respect to the direction of incidence of the positive betas, which is studied empirically, is discussed in relation to the asymmetry reported (reference 1).

For ease of presentation of results, Sec. III presents data with the direction of $\mathbf{E}$ averaged, while Sec. IV is concerned primarily with any differences on reversing $\mathbf{E}$. There are instances when the same raw data are processed for inclusion in both sections.

II. APPARATUS

Measurements for this experiment were made using two-photon angular correlation and single-photon pulse-height analysis equipment. The essential features of the angular correlation apparatus have been described previously.[5,6] Basically this equipment consists of two scintillation counters [NaI(Tl)] crystals mounted on DuMont 6292 photomultipliers connected in coincidence and a chamber located at the center of the apparatus, which contained the gas sample and Na^{22} positron source. One counter was fixed in position while the other was mounted on a movable arm pivoted about an axis normal to the z axis at the center of the apparatus. The defined angle θ is the projected angle.

Three different chambers[7] were used during this experiment; two of them are shown in Fig. 1. Either

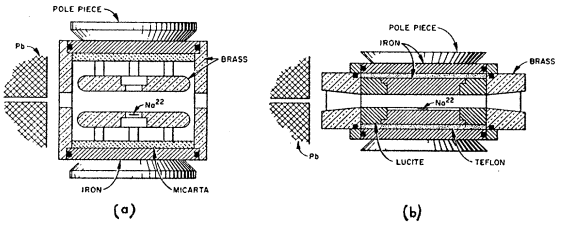

FIG. 1. Gas sample chambers. (a) Interelectrode spacing continuously variable to one centimeter, $|H|\cong 4$ kgauss; (b) interelectrode spacing fixed at 0.81 cm, $|H|\cong 9.4$ kgauss. The annihilation gammas leave the chamber through thin brass windows. The z axis is vertical.

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† Based on Ph.D thesis, Felix E. Obenshain, University of Pittsburgh, Pittsburgh, Pennsylvania, 1960 (unpublished).

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[1] F. Obenshain and L. Page, Phys. Rev. **112**, 179 (1958).
[2] M. Deutsch and S. Brown, Phys. Rev. **85**, 1047 (1952).
[3] M. Deutsch, Progress in Nuclear Physics (Butterworths-Springer, London, 1953), Vol. 3, p. 131.
[4] S. Marder, V. Hughes, C. S. Wu, and W. Bennett, Phys. Rev. **103**, 1258 (1956).
[5] M. Heinberg and L. Page, Phys. Rev. **107**, 1589 (1957).
[6] L. Page and M. Heinberg, Phys. Rev. **102**, 1545 (1956).
[7] The first chamber used is shown in reference 1.

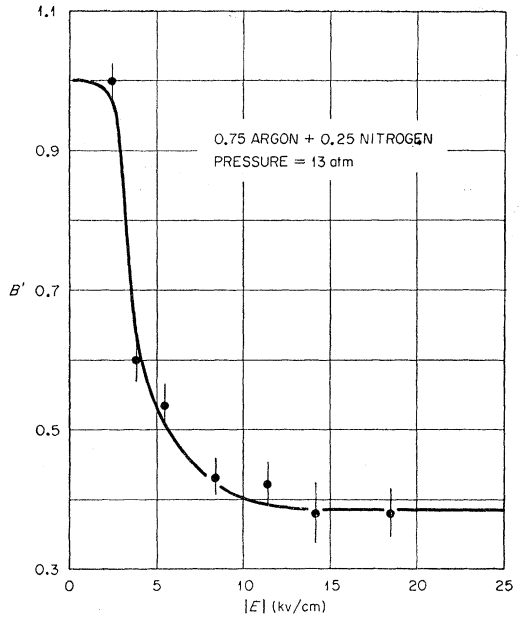

FIG. 2. A typical B' versus $|E|$ curve in the wing of the angular distribution, $\theta=10$ mrad, $\Delta\theta=5$ mrad. Total pressure 13 atm of 0.75 Ar+0.25 N_2. The value of $B_{sat}'=0.38$ indicates that the fraction of positrons that can be made to form positronium at this pressure is $F_E=0.76$, whereas the fraction at 3 atm is 0.84. The dependence of F_E upon pressure was linear over the region 3 to 25 atm.

chamber could be located between the pole pieces of an electromagnet having cylindrical symmetry about the z axis. The chamber shown in Fig. 1(a) has two adjustable electrodes, and the interelectrode spacing could be varied according to the requirement of the particular measurement to be made. The variability of the electrode spacing was achieved at some sacrifice in magnetic field strength. The chamber shown in Fig. 1(b) was used for measurements requiring large magnetic fields. The increase in magnetic field intensity was made possible by using mild steel for the electrodes instead of brass as in the other chamber. The electrodes of both chambers were insulated from the body of the chambers and a positive high-voltage supply was connected to one electrode and a negative supply to the other. The high voltage was balanced about ground (the chamber walls).

One of the two electrodes of each chamber contained the 15-mC Na^{22} source (the source was deposited on a quartz backing) and positrons emitted from the source were stopped in the gas sample between the two electrodes. It was arranged so that the detector viewed a differential element of the annihilating region above the source. The differential in the z direction (average direction of the positron emission) was determined by the slit arrangement. The slit system for this apparatus consisted of three slits: A pair of lead bricks adjacent to the chamber, called the main defining slit, and two additional pairs of lead bricks adjacent to each of the coincidence counters. The scintillation spectrometer crystal when needed could be set up about half way between the chamber and the stationary counter of the coincidence apparatus. The main defining slit at the chamber was then used with the spectrometer, and the observed differential element of the annihilation region could be defined with nearly the same precision as with the coincidence equipment. The scintillator was a NaI(Tl) crystal mounted on a DuMont 6292 photomultiplier. The signal from the photomultiplier was fed to a linear amplifier and then into two single-channel pulse-height analyzers.

III. RESULTS ASSOCIATED WITH THE MAGNITUDE OF E

Throughout this section a given measurement "at electric field strength $|E|$" shall mean that the data were taken alternately at $\mathbf{E}=+|E|\mathbf{k}$ and $-|E|\mathbf{k}$ with equal weights, the two results being averaged before presenting them. The asymmetry, if any, between respective results $\pm|E|$ is discussed separately under Sec. IV. Here $\mathbf{k}$ stands for the unit vector parallel to the axis of symmetry of the gas chamber—specifically upward in Fig. 1.

A. Angular Correlation Method

The angular correlation between the two quanta from two-photon annihilation of positrons in gases is thought of as consisting of three main components. These may or may not be reasonably well resolved depending on experimental circumstances, yet it is useful to speak in this fashion for purposes of outlining the method of these experiments.[8] Departures from collinearity greater than about 25 mrad are excluded from further discussion. The three components[9] of interest here are: Firstly, the narrow component with typical width at half-maximum ≤ 2 mrad,[10] which can only come from free positronium atoms having kinetic energy ≤ 0.5 ev. Examples are the magnetically quenched triplet states in argon, the exchange quenched triplet states in oxygen, and the singlet annihilation of "new" positronium produced by applying an electric field to positrons in argon. Secondly, there is the broad component, typically about 10 mrad wide, exemplified by the direct annihilations against atomic electrons of free positrons (usually almost thermalized). Thirdly, a component of intermediate width, which has never been directly observed,[11] is ascribed to singlet annihilation of

[8] This is no different from speaking of a total rate as though it consisted of a background rate plus a true rate, simply because there are circumstances where one or the other of the rates may be isolated.

[9] These have been discussed in a number of papers. See references 5 and L. Page and M. Heinberg, Phys. Rev. **106**, 1220 (1957).

[10] The actual residual true width undoubtedly varies with the gas sample and conditions. It is very expensive to measure accordingly since the coincidence rate has to fall off at least as fast as the *cube* of the resolution width.

[11] Its presence is, however, inferred from experimental data. See references 5, L. Page and M. Heinberg, Phys. Rev. **106**, 1220 (1957), and L. A. Page and F. E. Obenshain, Office of Ordnance Research Report, University of Pittsburgh, 1958 (unpublished).

positronium in motion with kinetic energy of perhaps one or two electron volts.

For the present measurements of the increase in positronium formation, one chooses an angular interval $\Delta\theta$ centered at some θ sufficiently far from zero that the narrow component is completely excluded from being counted. Then, the removal of a positron from the region below the Ore gap where it can only annihilate directly (broad component) to just above the threshold for positronium formation must remove it entirely from being recorded. Although we speak of projected angle throughout the paper, the argument, of course, would be the same for true angle. Suppose that through the agency of the applied electric field no positron failed to form positronium. Under such circumstances the only residual counting rate in the wing of angular distribution (aside from accidental background) would be excluded by choosing a sufficiently large θ.

To illustrate with typical data the curve of Fig. 2 is shown. B' denotes the ratio of coincidence rate at applied field $|E|$ to coincidence rate at $E=0$ with background taken into account. The angular resolution was $\Delta\theta=5$ mrad centered about $\theta=10$ mrad. (This curve happens to have seven points overall; in taking the data, zero field was run seven times as long on the whole as was a given nonzero field value.) For orientation it should be stated that for this same sample of gas similar data taken at $\theta=13$ mrad and $\Delta\theta=5$ mrad at some sacrifice in coincidence rate were consistent with the same-shaped curve. But most importantly both sets of data had the same saturation value of B', implying that the intermediate component was already excluded for $\theta=10$ mrad. The other possibility, namely that this component effectively coincides in width with the broad component, is considered to be less likely.

A curve such as that in Fig. 2 is found to be unchanged when data are taken with magnetic field **H** constantly applied of sufficient strength to quench out most of the triplet ($m=0$) states of positronium. This is simple proof that the narrow component has been excluded.[5] Again at the same resolution, but at the center of the angular distribution ($\theta=0$), the curve of B' vs $|E|$ has the same shape as in Fig. 2 but saturates much closer to unity (for this particular gas sample the saturation value was 0.82). If at $\theta=0$ the resolution is narrowed considerably, one can make B' to be flat against E. That is to say, the geometric efficiency for counting a two-photon annihilation event from newly-formed positronium relative to that for counting a direct annihilation can be made 4:1, the ratio of the pertinent statistical weights (for this particular gas sample one requires $\Delta\theta=0.82$ milliradian). Finally, in the presence of applied **H** (5–10 kgauss) the curve of B' vs $|E|$ can be made to be monotonically increasing, saturating at 2.0 for very narrow slits or at about 1.15 for very broad resolution.

It seems quite sure from numerous tests on samples of different gases at a variety of pressures that $E_{\frac{1}{2}}$ [defined to be the value of $|E|$ at which $B'=\frac{1}{2}(1+B_{\text{sat}}')$] has the same value independent of θ, $\Delta\theta$, and **H**. It was decided to adopt the procedure of running B' as a function of applied **E** at $H=0$ and in the wing of the angular distribution (as for the data of Fig. 2) for the sake of being in the position to interpret rather directly the physical meaning of the saturation value[12] of B'. To interpret B_{sat}' in terms of residual fraction of *total* positrons requires only multiplication by $1-F_0$, where F_0 is the fraction of all positrons forming positronium in the absence of applied **E**. (This latter number is rather quickly determined from measurement of Q_∞.[13]) Thus $(1-F_E)=B_{\text{sat}}'(1-F_0)$, where F_E is the fraction of all positrons forming positronium with saturation E applied.

The fraction $(1-F_E)$ is always an upper limit on the residual positrons due to the following: (1) The angular distribution of the direct annihilations has to be broadened, if anything, because of the higher average energy of the positrons in the presence of applied **E**; (2) the intermediate component may contribute to the rate even at large θ; (3) a fraction of the positronium atoms may annihilate during relatively high-energy collisions with the gas.

B. Argon and Nitrogen Results

The results are now summarized for measurements on argon and nitrogen made according to the techniques exemplified by the data of Fig. 2. Argon was run at four different pressures, 4, 9, 18, and 26 atm, nitrogen at 14 and 28 atm, and the mixed gases (0.75 argon plus 0.25 nitrogen by pressure) at 3, 7, 13, 18, 24, and 27 atm. All B' vs $|E|$ curves showed the characteristic delay, i.e., B' remains practically unity until $|E|$ exceeds a certain value which is typically 0.5 to 0.7 of $E_{\frac{1}{2}}$. All curves showed a well-defined "edge" so that $E_{\frac{1}{2}}$ could be easily estimated to within ten percent. Beyond the knee a flat region was always reached, termed saturation. For a given gas or mixture, $E_{\frac{1}{2}}/P$ (P is the gas pressure) was constant as it should be. Numerically, the results for $E_{\frac{1}{2}}/P$ are 180 v/cm-atmosphere for argon, 440 v/cm-atmosphere for nitrogen, 300 v/cm-atm for 0.75 argon +0.25 nitrogen. These values are not in disagreement with the work of the Columbia group,[4] which were 150 v/cm-atm for argon and 400 for nitrogen. In the present experiment, no provision was made for purifying the gases or baking-out the annihilation chamber.

The interpretation of B_{sat}' in terms of the fraction of positrons forming positronium in the presence of applied E is relatively straightforward and the fractions at saturation are: argon, 0.87; nitrogen (N_2), 0.86; 0.75 argon+0.25 nitrogen, 0.84.

[12] B_{sat}' under these circumstances is just the residual fraction of those positrons which in the absence of applied E would annihilate directly against atomic electrons.

[13] The quantity $(Q_\infty-1)$ is defined as the fractional increase in two-photon yield with a magnetic field applied ($E=0$). [See L. A. Page and M. Heinberg, Phys. Rev. **106**, 1220 (1957).]

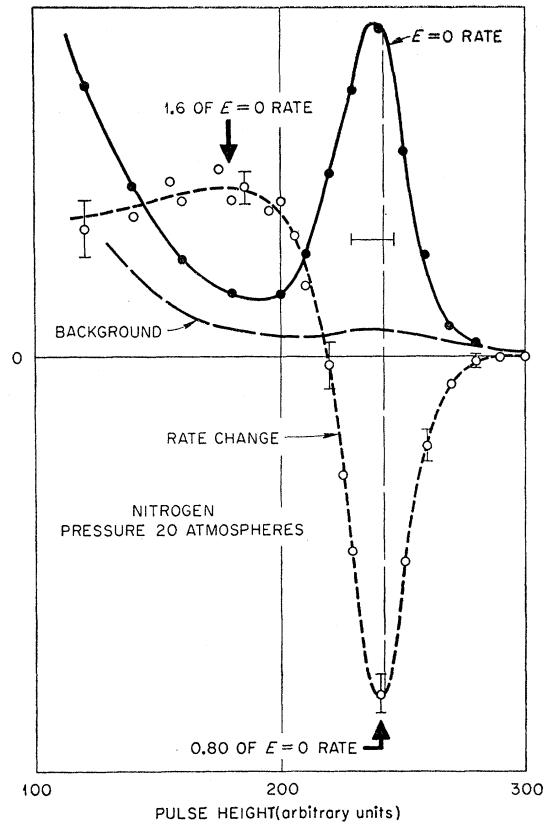

FIG. 3. Differential pulse-height spectrum for $E=0$ and the rate change on applying E (23 kv/cm). Total pressure is 20 atm of nitrogen. The spectral distribution of three quanta annihilations is evidenced by the shift of the minimum of the rate change curve away from the maximum of the rate curve. Arrows indicate key running positions.

The results given here are representative[14] of many such determinations of $E_{\frac{1}{2}}$ values and fractional increase in positronium formation. The general features of the curves are consistent for all pressures and mixtures of gases used. The value of $E_{\frac{1}{2}}/P$ for nitrogen is larger by a factor 2.4 than for argon. The consistency of these values with those of Marder *et al.* is good and indicates the purity of the gas sample is not of prime importance. The fraction of positrons which form positronium in various gases when saturation $|E|$ is applied is larger than the results of other investigators (reference 4), and the results of this experiment are not inconsistent with the assumption that all the positrons can be made to form positronium.

The direct approach of this method aids considerably the interpretation of the data in terms of the quantities of interest, and the quite rapid accumulation of the data insures good comparison between gas samples and different conditions for the same gas sample.

[14] F. Obenshain, thesis, University of Pittsburgh, 1960 (unpublished).

C. Pulse-Height Analysis on Single Photons

The principle of this method is by no means novel. One measures the differential pulse-height spectrum as a function of applied **E**, using the apparatus described under Sec. II. The method we use here to treat the data (which is essentially not to treat it at all) will be clear from Fig. 3. For orientation the figure shows the differential spectrum (total counts) at zero applied field, the solid curve. The background spectrum is also sketched; it is obtained by closing the photon slit adjacent to the chamber, which throughout the paper is called the "defining slit." The change in the differential rate on applying E is the quantity of primary interest. This is plotted directly in Fig. 3, as counts per unit time and *not* as a fractional change. By viewing the rate change in this way one does not get involved with having to ascertain the differential spectrum when all positronium is quenched. A difference curve such as that in Fig. 3 is obtained by sweeping across the pulse-height region of interest five or six times using the two single-channel analyzers (they are never both set at the same part of the spectrum). From such a plot one then chooses two key pulse-height settings indicated by the two arrows in the figure. It is interesting that both the key settings are shifted, respectively, from the peak point and the valley point on the solid curve. This is not a matter of backgrounds but is entirely reasonable inasmuch as the continuum representing the triplet annihilations is large immediately at and below the 0.51-Mev line[15] and again a certain fraction of the 0.511-Mev photons must "leak" down into the valley region of the actual pulse-height spectrum. However, both these effects should certainly be independent of applied E, and therefore the difference rate at either of the key points (or at any point for that matter) ought to be strictly proportional[16] to the transfer of positrons from singlet to triplet annihilation.

Figure 4 illustrates as a function of $|E|$ the difference rate for the two key points whose choice was just discussed. The 1.8:1 ratio of difference-rate-peak to difference-rate-valley, of course, depends primarily on discriminator channel width and scintillator efficiency as a function of photon energy. No attempt was made to predict this ratio accurately; it was merely noted that it was qualitatively plausible. What is important about this ratio is that it was found to be preserved when different gases were used (even for the abnormal features of SF_6 (see Fig. 8), once saturation was reached).

Specific results for $E_{\frac{1}{2}}$ were obtained by the pulse-height-analysis method for nitrogen at several pressures and for argon-nitrogen mixtures at several pressures. All features of the angular correlation curves B' vs $|E|$ were reproduced in the ΔR vs $|E|$ curves,

[15] R. Drisko, Phys. Rev. **102**, 1542 (1956).

[16] Checks were made using additional gamma sources to show that the circuitry was not overloaded.

namely the delayed onset of the transfer process, the well-defined edge, and the presence of the flat saturation region. The numbers obtained for $E_{\frac{1}{2}}$ divided by pressure for nitrogen and argon-nitrogen were so close to those already obtained by the angular correlation approach that we need not quote a separate set of numbers.

To interpret pulse-height data such as those of Figs. 3 or 4 in terms of actual fractional increase in positronium formation or in terms of residual fraction of positrons at saturation would require far more detailed knowledge of counter efficiency than was felt worthwhile pursuing. An alternative would be to quench out (presumably with a strong quenching agent, such as O_2 or NO) all positronium so as to have a reference pulse-height spectrum. In which case, one would be repeating in rather complete detail the work of Marder *et al.*[4]

To summarize the pulse-height approach as used, it can be said to give results entirely consistent insofar as it is able with the angular correlation method already discussed. This is of some concern when the complicated case of SF_6 is taken up later in this section.

D. Electric Field Experiments in Oxygen

There are at least two gases, oxygen and nitric oxide, which have the property of converting singlet ↔ triplet positronium. Deutsch[17] was the first to use these gases for their so-called "self-quenching" properties to demonstrate the existence of positronium in gases. The self-quenching (i.e., singlet-triplet) in oxygen has been attributed to the paramagnetic property and to the spin-flip involving unpaired electrons. The spin-flip mechanism can occur in oxygen, but the energy involved is the order of an electron volt.

Ferrell[18] has shown that spin-flip is not necessary for self-quenching, and the quenching is possible provided the molecule has at least one unpaired electron with which exchange (with the electron of the positronium atom) may take place. The angular correlation experiments of Heinberg and Page[5] showed that about 50% of the positrons in O_2 formed positronium and that the triplet positronium was largely quenched giving a very narrow component near 180°.

There does remain, however, the possibility of forcing more of the incident positrons to form positronium by means of an electric field, just as with the many other gases where this is possible.

When this problem was approached via the pulse-height analysis technique, it could be shown by progressively adding partial pressure of oxygen to argon with total pressure fixed at 27 atm, that the transfer of counts from 0.511 Mev to the continuum below was not being shifted particularly in terms of $|E|$ divided by pressure required, thus indicating that the positron energy loss in collisions with the O_2 molecule is not extremely large. But, of course, the additive oxygen was very active in attenuating such transfer of counts. The partial pressure of O_2 for 50% attenuation was only about 0.6 atm, the attenuation is extremely rapid, and at 27 atm (100% oxygen) there remained only about 3% of the original transfer for (pure) argon at the key peak point and at the key valley point.

Therefore, it was interesting to examine (pure) oxygen via the angular correlation approach. The data of Fig. 5 serve to illustrate what may be obtained via this method. It is found that saturation E field can essentially double the number of counts in the narrow component, and

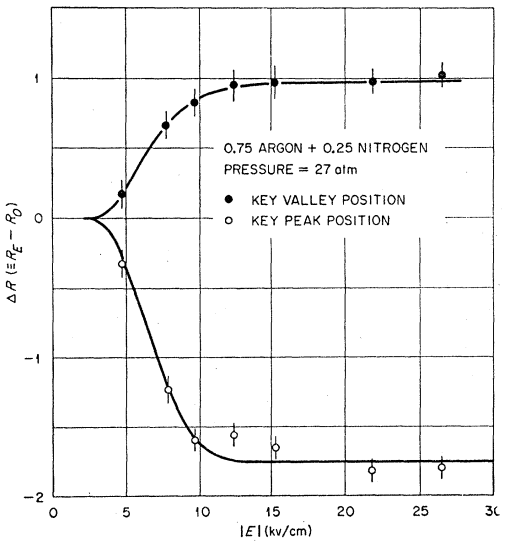

FIG. 4. A typical ΔR vs $|E|$ curve taken at the key peak and key valley position. Total pressure 27 atm 0.75 Ar+0.25 N_2.

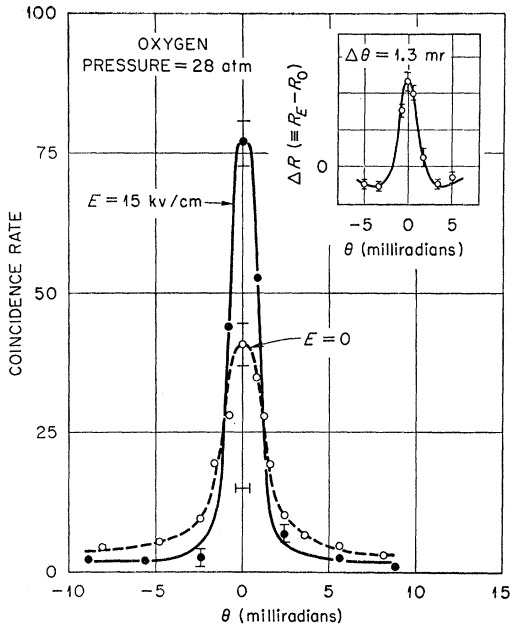

FIG. 5. An angular distribution curve ($\Delta\theta$=0.83 mrad) for 28 atm of oxygen, dashed curve E=0 and solid curve $|E|$=15 kv/cm. The insert is the rate change on applying E, ΔR vs θ.

[17] M. Deutsch, Phys. Rev. **82**, 455 (1951).
[18] R. Ferrell, Phys. Rev. **110**, 1355 (1958).

these counts must be drawn (at least some 0.94 of them) from the broad component according to the rather good conservation of 0.511-Mev gammas for high-pressure oxygen. It is concluded from this that the broad base under the narrow component for the $E=0$ curve of Fig. 5 (see also Fig. 2 of reference 5) has to be of the order of 20 milliradians wide at half maximum.

The E dependence of this transfer from broad to narrow is indicated by the typical curves of Fig. 6. Note that the resolution width is changed for counting rate reasons; B' has the same meaning as when used earlier. Complete curves similar to Fig. 6 were run for three different pressures of (pure) oxygen, 28, 22, and 15 atm. $E_{\frac{1}{2}}/P$ was the same for all six curves as far as the eye could judge. The numerical result is $(E_{\frac{1}{2}}/P)$ $=300$ v/cm-atm. This result may be compared with 180 for argon and 440 for nitrogen as given above. Since the value of $E_{\frac{1}{2}}/P$ for O_2 is less than for N_2, one is led to the conclusion that the positron energy loss in collisions with these two molecules is different. Further experimentation using purified gases could presumably lead to information concerning the energy loss mechanism and consequently information on the molecular energy levels.

The magnitude of B_{sat}' for O_2, both at $\theta=0$ and $\theta=5$ milliradians, approaches closer to unity as the pressure is increased and could be interpreted as a decrease in the fraction of positrons which can be made to form positronium with **E** applied. Assuming fraction of positrons which at zero E form positronium is 0.50 as stated above; then the fraction formed with applied **E** is between 0.80 and 0.90 at 28 atm.

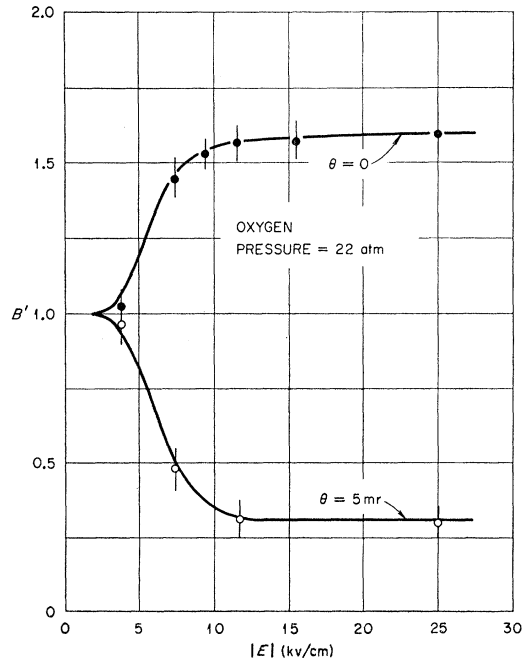

FIG. 6. A B' vs $|E|$ curve at $\theta=0$, $\Delta\theta=1.8$ mrad (upper curve) and $\theta=5$ mrad, $\Delta\theta=5$ mrad (lower curve) for a total pressure of 22 atm of oxygen.

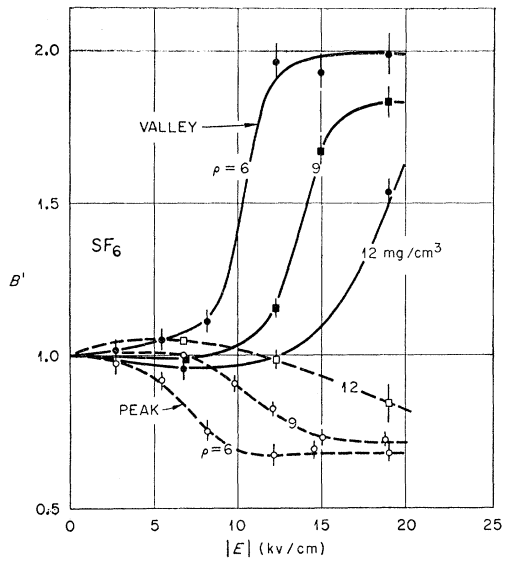

FIG. 7. Curves of B' vs $|E|$ for 6, 9, and 12 mg/cm³ of SF_6, demonstrating the *normal* positronium enhancement.

E. Sulfur Hexafluoride

Sulfur hexafluoride has been frequently used in the study of positronium.[3,19] In the present work it is used primarily for the sake of its large and controllable density (see particularly the discussion of the range-energy systematic effect under Sec. IV). Its characteristics with respect to electric-field enhancement of its fractional positronium yield were first examined by the Columbia group,[4] and an anomalous decrease in apparent positronium formation was found for pressures of one to two atmospheres and field strengths up to 2.2 kv/cm.

In the course of the present experiments the normal increase of positronium formation by virtue of an applied electric field has been identified for the first time. Figure 7 shows curves obtained using the pulse-height analysis method which has been described above. (The data happen to be plotted here in terms of the ratio B' of field-on-rate to field-off-rate which does not change the features we wish to discuss.) For the first time one finds that the valley curve generally lags[20] the peak curve as though there were a nonconservation of annihilation events.[21] In any case the ratio of the saturation values of rate changes, for example, at 6 and 9 mg/cm³

[19] S. DeBenedetti and H. Corben, Ann. Rev. Nuclear Sci. 4, 191 (1954).

[20] Some months later in a different chamber and using a different bottle of SF_6, this feature was found to still persist.

[21] At 6 mg/cm³ and $|E|=8$ kv/cm a search was made for the "missing" gamma rays (the curves of Fig. 7 are based only on the two key points in the pulse-height spectrum, cf. Fig. 3), scanning from 200 kev up to 1.2 Mev with special attention paid to $\frac{4}{3}mc^2$ and $2mc^2$, but within statistics none were found.

does come out to be 1.8:1 (with the discriminator window set the same as for the data of Fig. 4) as with the other gases discussed above.

From Fig. 7 we see that $E_{\frac{1}{2}}$ is of the order of 10 000 v/cm-atm (compare 180 v/cm-atm for argon) but, of course, we are dealing here with a comparatively large and complicated molecule. A fairer comparison might be made in terms of v/cm-(mg/cm^3). In this case the SF_6 requires about a factor 10 more field than argon for example.

Returning to Fig. 7 one notes for the 9- and 12-mg/cm^3 curves that in the region (below about 9 kv/cm) where one should normally expect B' to remain essentially unity, there is a definite "overshoot" which applies both to peak and valley curves. This is the anomalous effect noted by Marder *et al.* where seemingly there is a *decrease* in positronium formation on applying E. From the fact that the overshoot seemed to be absent at 6 mg/cm^3 (one atmosphere), it seemed reasonable to see whether the anomalous decrease would become more prominent at still higher densities than shown in this figure.

Figure 8 shows typical results obtained at a higher density where the normal increase presumably would not set in until about 30 kv/cm. The saturated portion of the peak-curve corresponds to about 5% of the peak rate, while the valley-curve appears to saturate at about 10% of the estimated valley rate. The remarkable feature of these results is that the half-point should be reached so early as $|E|$ is increased. At this rather high density equivalent (considering mg/cm^3) to about 15 atm of argon, nitrogen, or oxygen, the half-points for the peak curve and the valley curve are, respectively, 400 and 800 v/cm. Figure 8 is typical of curves obtained for 71, 50, and 11 mg/cm^3 of SF_6, except that the abscissa must be scaled approximately as the density of the gas. The earliness of the onset of the anomalous decrease in positronium formation is accentuated also by the fact that SF_6 (quite reasonably) is a very lossy medium in which to attempt to accelerate positrons—apparently an order or magnitude more lossy than argon for example.

Analysis of this early component in SF_6 via the angular correlation approach showed that it was readily identified in the wing of the curve (8 to 10 mrad) and could hardly be found at the center of the angular correlation. At 32 mg/cm^3, for example, B' as a function of $|E|$ run at $\theta=8.3$ mrad, $\Delta\theta=5.5$ mrad had its half-point at one kilovolt/cm and was flat from 8 kv/cm to 20 kv/cm with $B_{\text{sat}}'=1.14\pm0.01$.

Adding oxygen (equal pressures) to SF_6 did not appreciably affect the early component when observed in the wing of the angular distribution, but when observed by the pulse-height analysis method, both the early component and the normal component were made consistent with zero. The absolute pressure of O_2 added was never more than the pressure of the SF_6 sample being observed.

The application of a magnetic field ($|H|=9.4$ kgauss) did not have any effect outside the statistical limits on the early component in either the wing of the angular distribution or in the peak of the pulse-height analysis, but the valley showed a decrease, H applied, in the size of $\Delta R=(R_E-R_0)$, $|\Delta R(|H|=9.4 \text{ kgauss})|\cong\frac{2}{3}\times|\Delta R(H=0)|$, as did the normal component at both the peak and valley key points.

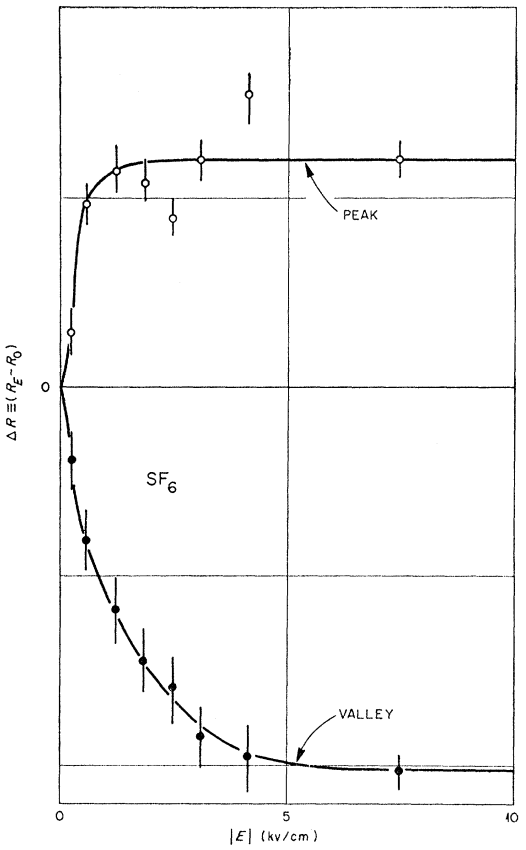

FIG. 8. Data taken at the key peak and valley positions of the differential pulse-height spectrum ΔR vs $|E|$, which show the anomalous decrease in positronium formation for SF_6, $\rho=18$ mg/cm^3. Note the small $|E|$ required to saturate the effect.

Detail investigation of the early component showed that in both the wing of the angular distribution and in the peak of the pulse-height analysis, structure in the curve was at times apparent and small $|H|$ (of the order 4 kgauss) would tend to wash out the structure. Upon close examination positron annihilation in SF_6 is anomalous in many ways, but the gross features, which are of interest here, are always reproducible.

To summarize the findings with SF_6: At high densities and at remarkably low fields, the anomalous apparent decrease in formation is investigated in considerable detail. It is not clear to what mechanism this decrease should be ascribed. At low densities and very high E fields the normal increase in positronium formation has been identified. $|E|/\rho$ (ρ the gas density) is the parameter of importance. It appears that about 0.75 of the incident positrons can be made to form positronium.

IV. RESULTS ASSOCIATED WITH THE SIGN OF E

A previous experiment carried out in this laboratory used strong electric fields applied parallel and antiparallel to the direction of emission of the positrons from a Na^{22} source. The asymmetry observed in the positron annihilation rate upon the reversal of **E** was thought to imply a nonzero value for the hypothetical energy term $\xi\boldsymbol{\sigma}\cdot\mathbf{E}$. During the course of the experiment, a number of systematic effects was considered. Of the systematic effects considered, one was the range-energy systematic effect and it was argued (footnote 9 of reference 1) that such an effect had the sign opposite to that observed.

The later more detailed experimental investigation of the range-energy systematic effect, which is now being reported, shows that the sign of the effect given by the argument could obtain but could only obtain at a considerably larger distance (mg/cm²) from the source than the distance used for the original experiments of reference 1.

Positrons slowing down in a gas sample which has a strong electric field applied may either gain or lose energy depending on the direction of the electric field with respect to the incident positron. The gain or loss of energy does, of course, have an effect on the range of the particles, and the spatial distribution of positrons of kinetic energy less than 20 ev is perturbed accordingly. In first approximation the average of the spatial distributions for the two directions of **E** should be just the $E=0$ distribution. Therefore, the spatial distributions for $+|E|$ and $-|E|$ must cross each other at a certain distance from the source if positrons are to be conserved (i.e., a retarding field will give a greater number of slow positions near the source than an accelerating field).

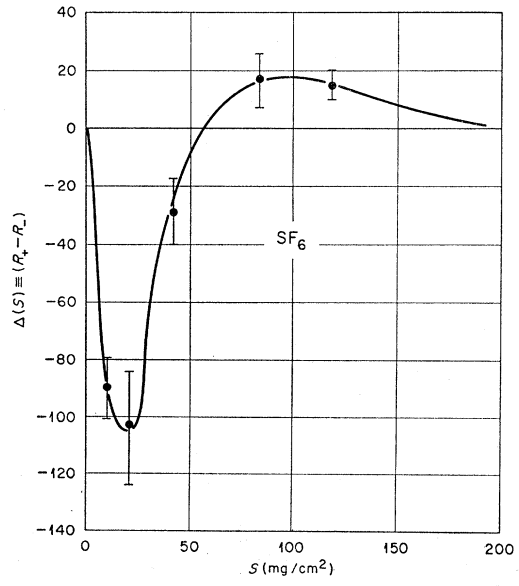

FIG. 9. The rate change $\Delta(S)=(R_+-R_-)$ as a function of distance from the source S (mg/cm²) on applying $E=+|E|$ and $E=-|E|$ to a sample of SF_6.

When the positron annihilation rate is measured for $+E$ and $-E$ as a function of distance (mg/cm²) from the source, it is possible to find a position where $\Delta R(=R_+-R_-)$[22] changes sign, R_+ and R_- are the coincidence rates for $+E$ and $-E$, respectively. Figure 9 shows the results of such a measurement. The sign of ΔR is negative for $S\leq 60$ mg/cm² (where S is the distance from the source in mg/cm²) and positive otherwise. In order to obtain the data of Fig. 9, it was necessary to use a dense gas and SF_6 was selected for this purpose so that a large fraction of positrons could be stopped in the visible region of the chamber.

From these and other data it was determined that the sign of ΔR, the range-energy effect, could only be negative for the less dense gases argon and N_2 within the observable annihilation region of the apparatus. The sign of this systematic asymmetry is the same as that observed for the asymmetry of reference 1. At the time the experiments (reference 1) were completed, the true $\Delta(S)$ was unknown (i.e., had not been measured). It was then erroneously concluded that the range-energy systematic effect could only give a positive Δ in the region $S\cong 30$ mg/cm² while the observed Δ was negative.

The size of the range-energy Δ (expressed as a percentage change in coincidence rate) is ample to account for the magnitude of the original asymmetry, but it fails to account for certain detail features, for example the observed E and H dependence.

The Δ was measured in SF_6 mainly to establish the features of the range-energy systematic effect. Small variations in the percentage Δ in SF_6 were observed at times, but these could not be traced to any malfunction of the electronic equipment.

The observed Δ (in particular for 0.75 Ar+0.25 N_2) is sensitive to the application of a magnetic field. This sensitivity alone indicates that at least one other effect in addition to the range-energy effect is involved. This conclusion may be drawn from consideration of the parameters of the experiment which do not affect the magnitude of the range-energy asymmetry (expressed as a percentage). They are as follows: (1) the quenching of the $1\,^3S$ ($m=0$) triplet state when H is applied, (2) the magnetic "focussing" of the positrons, which was found to be negligible for pressures greater than 15 atmospheres, and (3) the increased positronium formation due to applied **E**. Each of these affects the coincidence rate, but the change may be viewed as a simple change in the detector efficiency and thus the magnitude of the range-energy effect is not affected.

Figures 10 and 11 serve to illustrate the characteristic features of the Δ for certain conditions of the experiment. Figure 10(a) shows the percentage change in coincidence rate as a function of applied $|E|$ for 0.75 argon +0.25

[22] The sign of ΔR as defined here is *opposite* to that defined in reference 1. In present work, the *positive* sign denotes *upward* **E** in Fig. 1.

nitrogen at a total pressure of 25 atm. The Δ is consistent with zero out to about 9 or 10 kv/cm and for higher voltages, Δ increases with nearly a linear dependence on $|E|$. Similar curves were obtained for pressures down to about 15 atm, but below this pressure, the curves of Δ vs $|E|$ were characteristic of the one shown in Fig. 10(b). That is, no definite delay in Δ could be established for these pressures.

The data of Fig. 11(a) and (b) are measurements of Δ for two pressures, 27 and 18 atm, and at the center of the angular distribution curve $\Delta\theta=5.5$ mrad. The asymmetry again exhibits the characteristic delay and then begins to increase at higher $|E|$. Figure 11(a) is a comparison of the form of Δ with the increased formation of positronium with E applied, measured in the wing of the angular distribution as described in Sec. III. Here it is observed that the departure of Δ from zero follows rather closely the onset of the positronium enhancement curve. The Δ's at 10.5 and 15 kv/cm are both about 4.5% and indicate possibly a saturation of Δ in that region again following the curve of B' versus $|E|$ which saturates at about $|E|=12$ kv/cm at this pressure. The data of Fig. 11(a) also show the delay in $\Delta(H=0)$ and a tendency to saturate following the curve of B' versus $|E|$. The filled circles of Fig. 11(b) are Δ's with a magnetic field applied ($H=9.4$ kgauss)[23] and the Δ's ($|E|\geqslant 5$ kv/cm) generally lie below those for $H=0$. The Δ vs $|E|$ with H applied follow more closely a linear dependence with E as should be the case when Δ is entirely due to the range-energy effect.

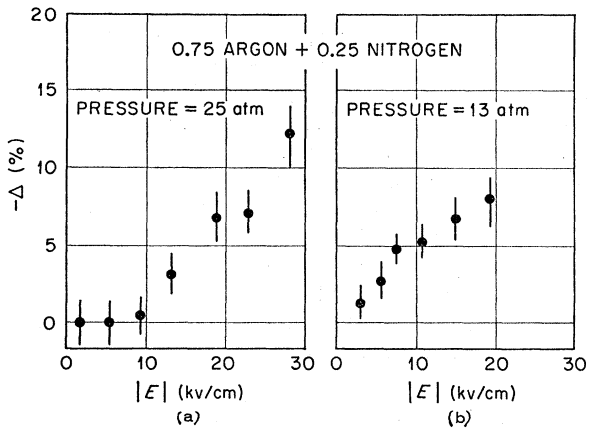

FIG. 10. The percentage change in coincidence rate Δ as a function of applied $|E|$. (a) Total pressure 25 atm of 0.75 Ar +0.25 N_2, $\theta=0$, $\Delta\theta=5$ mrad; (b) total pressure 13 atm of 0.75 Ar +0.25 N_2, same resolution. Note that Δ is consistent with zero up to about 9 kv/cm at 25 atm, but there is no evidence of this behavior at 13 atm.

[23] These data were taken during the same run for which the Δ's with $H=0$ were obtained.

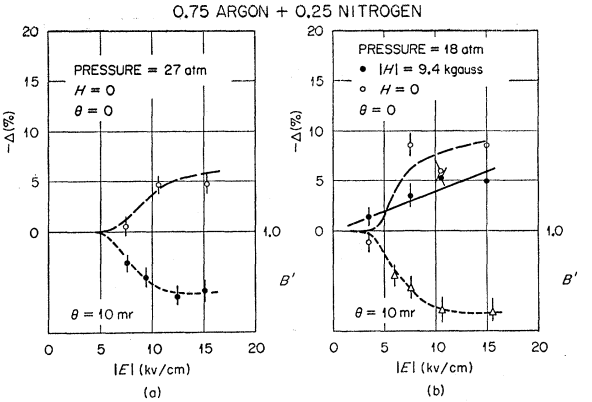

FIG. 11. Comparison of Δ vs $|E|$ and B' vs $|E|$ for two pressures of 0.75 Ar+0.25 N_2. (a) Δ measured at $\theta=0$, $\Delta\theta=5.5$ mrad (upper curve) and B' measured at $\theta=10$ mrad, $\Delta\theta=5.5$ mrad (lower curve), $H=0$ 27 atm; (b) Δ measured at $\theta=0$, $\Delta\theta=5.5$ mrad, $H=0$ (open circles) and $H=9.4$ kgauss (filled circles), B' measured at $\theta=10$ mrad, $\Delta\theta=5.5$ mrad, $H=0$ (lower curve), at pressure 18 atm. The data points with $|H|=9.4$ kgauss are characteristic of what one expects for the range-energy effect; whereas the points for $H=0$ follow a different functional form.

The data obtained during the course of this investigation show that Δ is sensitive to applied H in almost all cases. The magnitude of Δ with H applied is generally less than the $\Delta(H=0)$, the difference of the two being the order of 2 or 3%. As mentioned earlier, the magnitude of the energy-range effect cannot be decreased on application of the magnetic field and the Δ does not follow exactly $|E|/P$ at high pressure; therefore one is led to the conclusion that the observed asymmetry must be due to the range-energy effect, plus some other effect which depends on the reversal of **E**.

The pertinent features of the range-energy effect are measured. The sign of this effect is established as well as its general size for the electric fields of interest. For field reversal experiments, knowledge of this effect is crucial. On the other hand, it is felt that it cannot interact appreciably with the experiments which depend on the magnitude of **E**, Sec. III.

The size of the range-energy effect is ample to account for the observed asymmetry (reference 1), but the details, the **E** and **H** dependence, cannot be explained on the basis of this effect.

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