The Three-Photon Annihilation of Positrons and Electrons*

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(Received January 22, 1954)

The physical characteristics of the radiations emitted in the annihilation of positrons and electrons in the triplet state have been studied with a triple coincidence method. The energies of the three gamma rays, the angular distribution, and their polarization were found to agree with theory. Triplet annihilation was studied both from positrons stopping in gases where positronium was formed, and from positrons stopping in condensed materials. In the case of metallic absorbers, we verified within the accuracy of the experiment that the number of triplet annihilations is 1/370 of the number of singlet events. Experiments on the number of three-quantum annihilations in different materials are described.

I. INTRODUCTION

It is well known that a pair of positive and negative electrons can annihilate with the transformation of their rest masses into electromagnetic radiation. In order to satisfy the law of conservation of momentum at least two gamma rays must be emitted, and the main features of the process can be computed as a second-order effect of the interaction of the electrons with the electromagnetic field.

One can expect, however, that higher-order annihilation processes will occur, and in particular that positron-electron pairs should annihilate with the emission of three gamma rays in about 1/137 of the cases. Among the first to carry out a calculation of this effect were Lifshitz,[1] Ivanenko,[2] and Ore and Powell.[3] The result of these last authors (later confirmed by Radcliffe[4] and by Drisko[5]) is that the relative frequency of the three-quantum and two-quantum processes is 1/370 for a random distribution of spin orientations. The effect of spin orientation is simply that singlet pairs annihilate only with the emission of two quanta,[6,7] triplet pairs only with the emission of three.[8]

The purpose of the present work was an experimental investigation of the three-quantum annihilation process, including the probability of its occurrence as well as the physical properties of the emitted gamma rays.[9]

Before our first results were obtained, Rich[10] presented some evidence of the existence of this effect, obtained with a triple coincidence method, and indicating a probability of occurrence of the predicted order of magnitude. Shortly afterwards Deutsch[11] discovered that positronium, the bound positron-electron system, is formed with large probability when positrons are stopped in certain gases. This result was of great help in our work because the formation of positronium, destroying the random distribution in the spin orientations, considerably increases the probability of three-quantum annihilation and thus provides sources from which the characteristics of the radiations emitted can be conveniently studied.

II. INSTRUMENTATION

The three gamma rays emitted in the triplet annihilation process were detected by means of three NaI(Tl) scintillation crystals, each 2.5 cm thick and 4 cm in diameter. The light pulses from the crystals were viewed by RCA 5819 photomultipliers, whose anodes were connected in coincidence, with a resolving time measured to be 1×10^{-7} sec. In addition, the pulses from the last dynode of each tube were amplified by standard linear amplifiers and passed through single channel differential pulse height selectors. The pulse height selector outputs were placed in slow (2×10^{-6} sec) coincidence with the (delayed) output of the fast coincidence circuit. Thus a set of three coincident

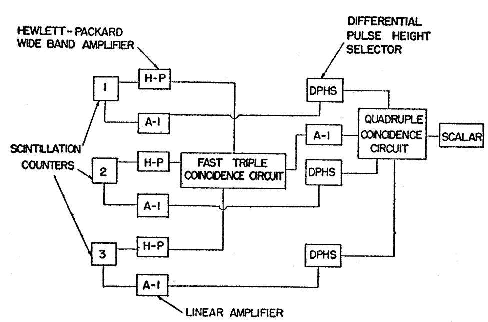

Fig. 1. Block diagram of electronic apparatus.

* Work supported in part by the U. S. Atomic Energy Commission (1951–1952), and in part by the National Science Foundation (1952–1953).

† U. S. Atomic Energy Commission Predoctoral Fellow (1950-1952). Portions of this work were submitted by R. T. Siegel in partial fulfillment of the requirements for the degree of D. Sc. at Carnegie Institute of Technology.

[1] E. M. Lifshitz, Doklady Akad. Nauk. S.S.S.R. **60**, 211 (1948).

[2] D. Ivanenko and A. Sokolov, Doklady Akad. Nauk. S.S.S.R. **61**, 51 (1948).

[3] A. Ore and J. L. Powell, Phys. Rev. **75**, 1696 (1949).

[4] J. M. Radcliffe, Phil. Mag. **42**, 1334 (1951).

[5] R. M. Drisko (private communication).

[6] C. N. Yang, Phys. Rev. **77**, 242 (1950).

[7] L. D. Landau, Doklady Akad. Nauk. S.S.S.R. **60**, 207–209 (1948).

[8] L. Wolfenstein and G. D. Ravenhall, Phys. Rev. **87**, 217 (1952).

[9] For a prelimary communication, see S. DeBenedetti and R. Siegel, Phys. Rev. **85**, 371 (1952).

[10] J. A. Rich, Phys. Rev. **81**, 140 (1951).

[11] M. Deutsch, Phys. Rev. **82**, 455 (1951); M. Deutsch and E. Dulit, Phys. Rev. **84**, 601 (1951); M. Deutsch, Phys. Rev. **83**, 866 (1951).

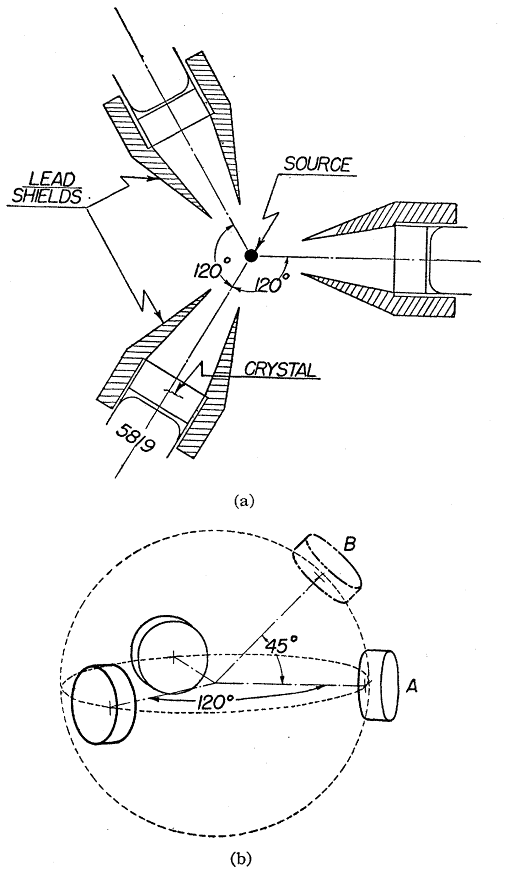

FIG. 2. Arrangement of the counters for detection of three-quantum annihilation (a) Top view, (b) Perspective of the crystals.

gamma rays was accepted only if each yielded a pulse whose amplitude lay in the band accepted by the corresponding differential pulse-height selector. Figure 1 shows a block diagram of the electronic equipment used.

As a source of positrons we used Na^{22}, which is convenient because of its long life. In addition to the positrons and their annihilation radiations, this isotope emits a nuclear gamma ray of 1.3 Mev, which is undesirable since it increases the number of random coincidences. However, when the pulse-height selectors were properly set, this effect was reduced and the random coincidences did not interfere with our measurements.

The three counters (Fig. 2) were placed at a distance of 12.0 cm from the source, and in this position each covered a solid angle of 0.087 sterad. For the measurement of the effect under investigation the counters and the source were coplanar [A, Fig. 2(b)] since most pairs annihilate with negligible momentum. For the measurement of the background one of the counters was removed from the plane defined by the source and by the other two, without altering its distance from the source [B, Fig. 2(b)].

In order to reduce spurious coincidences due to scattering, each counter was protected by a lead shield as can be seen in Fig. 2.

III. THE PHYSICAL PROPERTIES OF THE THREE QUANTA

(a) The Energy of the Three Quanta

For any given orientation of the counters, the energy of the three quanta of triplet annihilation can be computed in an elementary way from the laws of conservation of energy and momentum. Though the result of such computation needs no experimental confirmation, we have performed the energy measurement to prove beyond any doubt that the coincidences recorded were due to the phenomenon of three-quantum annihilation.

For this purpose a positronium source was used. This consisted of a bell shaped container (1-in. diameter, $\frac{1}{16}$-in. wall thickness) holding SF_6 vapor at 10 atmospheres; the Na^{22} activity (1.4×10^6 dis/sec) was deposited between two thin Zapon foils supported in the center of the container in such a manner that most positrons would stop in the vapor, where the formation of positronium is favored. The pulse-height distributions of the coincident pulses were studied for each counter in succession, with a band width of 90 kev; the background was obtained as previously described by rotating one of the counters away from the coplanar position. The angular arrangements for which data were taken and the computed energies for each configuration are those indicated with an asterisk in Table I. The data are plotted in Fig. 3 after subtraction of background, and after multiplication by a convenient factor in order to equalize the height of the photopeaks. The uppermost curves show the pulse-height distribution of the two-quantum annihilation line, and provide the energy scale for each counter. The other curves, which are the pulse height distributions of the three-quantum annihi-

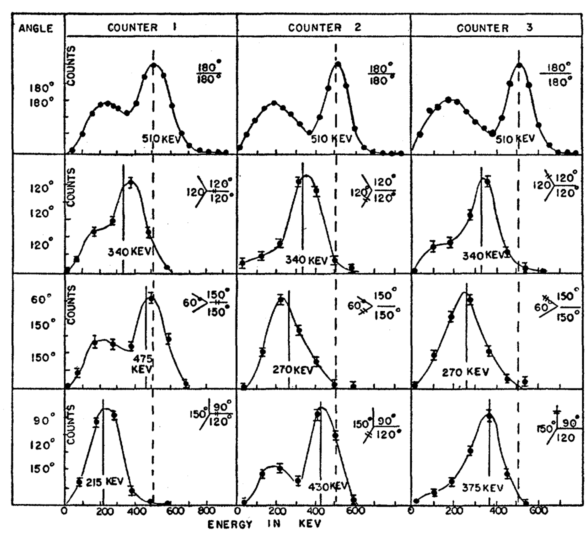

FIG. 3. Pulse-height spectra of three-photon annihilation for different angular arrangements.

TABLE I. Angular distribution data.

A Angles between counters 2–3	3–1	1–2	*B* Energy in each counter (kev) E_1	E_2	E_3	*C* Relative intensity (exp.)	*D* Rel. int. (exp. corrected)	*E* Rel. int. (theoretical)	*F* Exp./theory (D/E)	*G* Exp./stat. theory
*120°	120°	120°	341	341	341	1.00	1.00	1.00	1.00	1.00
100°	120°	140°	263	355	404	1.01±0.04	0.925	0.97	0.95	1.00
100°	100°	160°	435	435	150	1.11±0.05	0.95	0.82	1.16	4.45
*90°	120°	150°	430	215	375	0.86±0.04	0.74	0.89	0.83	0.95
80°	140°	140°	444	289	289	1.09±0.05	0.89	0.97	0.92	1.01
80°	120°	160°	160	404	456	1.05±0.04	0.84	0.75	1.12	1.50
*60°	150°	150°	475	272	272	1.25±0.04	0.82	0.97	0.85	1.04
60°	140°	160°	189	355	478	1.41±0.05	0.96	0.87	1.10	1.43
50°	150°	160°	318	217	487	1.69±0.05	0.91	0.93	0.98	1.25
75°	75°	210°		Forbidden		0±0.05				

lation, show photopeaks near the computed energy (vertical lines) together with evidence for the continuous distribution of Compton electrons. The agreement with expectation is satisfactory in all cases.

In order to prove that our instrument could reliably be used for the study of the rare phenomenon of triplet annihilation in solid media, the energy measurement was repeated with a solid positron absorber as a source. The results, obtained only for the symmetrical configuration, were in agreement with those reported above.

(b) Angular Distribution

In order to measure the angular distribution of the gamma rays, the bands of the pulse-height selectors were opened to accept all the pulses (photo and Compton) due to the appropriate gamma energy; then the number of triple coincidences recorded for any one configuration was compared to that obtained for the symmetrical case. The measured relative intensities are reported in Table I, column *C*.

The data were then corrected for variation in geometrical acceptance and for energy dependence of counter efficiency.

The geometrical acceptances of the different counter arrangements were computed numerically.[12] The errors introduced by correcting the data in this way are no larger than the statistical errors of the measurements themselves.

In order to correct for variation of counter efficiency, it was assumed that the efficiency varies with energy as shown by the curve of Fig. 4. Two points of this curve were experimentally measured. The value 0.38 at 0.51 Mev was obtained from coincidence measurement between the two-quantum annihilation gammas; and the value 0.54 at 0.34 Mev was determined using scattered gamma rays from the annihilation line. The rest of the curve represents an estimate made under the assumption that the efficiency should be unity under 100 kev, and should be proportional to the absorption of the gamma rays at high energy.

Despite our poor knowledge of the efficiency, this correction introduces an error of less than 10 percent in the measurement of angular distribution. This is because all measurements are taken in reference to the symmetrical arrangement, making any error in absolute efficiency at $\frac{2}{3}mc^2$ inconsequential. Furthermore, since the sum of the three energies is constant, the product of the three efficiencies varies much less than any one efficiency, and is in fact constant within 20 percent in all our measurements.

The experimental data corrected for both geometrical and counter efficiency variations are reported in column *D* of Table I. Column *E* shows the theoretical angular distribution, computed by means of a change of variables from the expression for the energy spectrum of the gamma rays given by Ore and Powell.[3] The ratios between the corrected experimental data and the theoretical distribution are shown in column *F* of Table I. The dispersion of these figures is somewhat larger than the purely statistical errors of the measurement, but, considering the errors introduced by the corrections, the disagreements (seldom larger than 10 percent) are within permissible limits.

Column *G* of Table I shows the ratios between the corrected experimental data on angular distribution and a statistical estimate of the distribution, obtained theoretically under the assumption that all matrix elements are independent of energy (and therefore of angle). It can be seen that the experimental data are sufficiently accurate to distinguish between the complete theory and the statistical estimate, favoring the

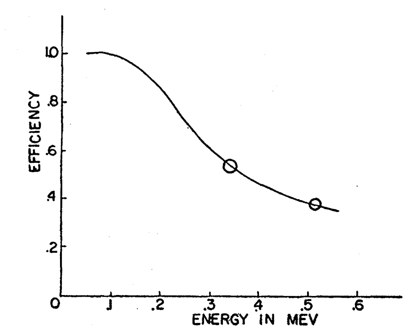

FIG. 4. Assumed efficiency *vs* energy for a 2.5 cm thick sodium iodide crystal. The two points were measured experimentally.

[12] For details of this computation, see R. Siegel, thesis, U. S. Atomic Energy Commission Report NYO-3302 (unpublished).

first in the cases where the two lead to significantly different conclusions.

Since for each angular arrangement the energies of the gamma rays are fixed, it is clear that a measurement of angular distribution is equivalent to a determination of the energy spectrum of the three-quantum annihilation gamma rays. The results of the angular distribution measurements may therefore be interpreted as verifying the energy spectrum from the complete theory rather than the statistical prediction based on the density of final states.

(c) Polarization

Another interesting characteristic of the three annihilation photons is their polarization. After the polarization effects had been theoretically calculated at our laboratory,[13] we proceeded to an experimental investigation of the polarization of one of the gamma rays relative to the plane of emission of the three,[14] since this is the easiest effect to study from the experimental point of view.

The experimental result was in agreement with the theory, which predicts a polarization ratio 3/1 in favor of rays plane-polarized with the electric vector perpendicular to the plane of the three photons. The experiment itself is described in reference 14.

IV. THE NUMBER OF TRIPLET ANNIHILATION EVENTS IN VARIOUS MATERIALS

(a) The Number of Triplet Annihilations in Aluminum

Since it seems justifiable to assume that, within a metal, the annihilation events occur between electron-positron pairs with randomly distributed spins, one is led to expect that the relative probability of three- and two-quantum annihilation should be 1/370, as computed by Ore and Powell. An attempt to verify this is described in what follows.

A sample of Na^{22} was enclosed in an aluminum capsule. With this as a source, the number of triple coincidences in the symmetrical position (120°, 120°, 120°) was measured and found to be, after background subtraction, $C=0.92\pm0.10$ counts/min.

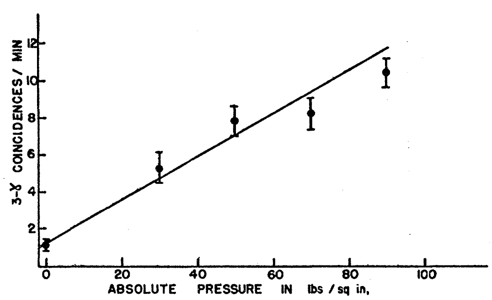

FIG. 5. Rate of 3-γ coincidences *vs* pressure in Freon-12 (CCl_2F_2).

[13] R. Drisko (private communication).
[14] Leipuner, Siegel, DeBenedetti, Phys. Rev. **91**, 198 (1953).

In order to compute the number of coincidences to be expected according to the theory, it is necessary to know the strength of the source, the efficiency of the three counters, and their geometrical acceptance. The geometrical acceptance was calculated as described in reference 12; the efficiency was taken from the curve of Fig. 4; and the strength of the source was obtained by comparison with a source calibrated in Oak Ridge, and also from an annihilation double-coincidence experiment (the same experiment from which the efficiency at 0.51 Mev was computed), the two methods yielding consistent results.

Using the values obtained in these preliminary studies, the expected value of C corresponding to the 1/370 ratio was 1.2 ± 0.16 counts/min. This is not in disagreement with the measured value.

(b) The Number of Triplet Annihilations in Gases

The number of triplet annihilations is considerably enhanced in gases whose chemical properties favor the formation of positronium.[11] In order to verify this effect, we performed the following experiment. The aluminum container described in Sec. III (a) was filled with Freon, and the three-quantum coincidence rate was observed as a function of pressure. The increase in coincidences with pressure (Fig. 5) indicates that ortho-positronium is formed when positrons stop in the gas rather than in the metal walls of the container.

TABLE II. Relative positronium yields in various gases.

Material	Pressure (psia)	Relative electron density	Relative 3-γ counting rate (reduced to equal electron densities)
Freon-12	75	1.0	1.00
SF_6	62	1.0	0.79 ± 0.04
He	215	0.099	1.15 ± 0.15
H_2	215	0.099	0.96 ± 0.25
N_2	212	0.68	0.84 ± 0.06
O_2	122	0.46	0.006 ± 0.018

The container was also filled with other gases in order to study relative positronium formation. An estimate of the amount of positronium formed per positron stopped in the gas, was obtained by dividing the triple-coincidence counting rate by the relative electron density of the gases at the pressures used. The results are shown in Table II.

It is noteworthy that all the gases examined (except O_2, which is known to catalyze the conversion of triplet positronium to singlet) give a yield of positronium per stopped positron which is the same within ±25 percent. This average yield has been estimated from the source strength and counter geometry to be about 12 percent, in fair agreement with results of Deutsch and Pond.[15]

Figure 6 shows a graph of 3-γ counting rate (i.e. amount of orthopositronium formed) *vs* density of SF_6

[15] T. A. Pond, Phys. Rev. **85**, 489 (1952).

in the gas container. Because of the high density of SF_6 attainable at room temperature, it was possible to follow the 3-γ rate into a density region in which the quenching becomes measurable. The decrease in 3-γ rate in SF_6 between 0.11 and 0.25 gm/cc yields a quenching cross section of $\sigma(SF_6)=9.7\times10^{-22}\,\text{cm}^2$, with an estimated accuracy of about 15 percent. This result has been confirmed by Wheatley and Halliday.[16]

One concludes from the magnitude of this cross section that the spin-exchange process which probably causes the large quenching effect in oxygen $[\sigma(O_2)=4.0\times10^{-19}\,\text{cm}^2]$[17] is much less effective in the case of SF_6.

(c) Miscellaneous Experiments with Solids and Liquids

An attempt was made to investigate the presence of positronium due to positrons stopped in liquefied nitrogen or Freon. The source of positrons was deposited on the inside bottom of a small Dewar flask and covered with a thin plastic film. A considerable fraction of the positrons emitted traversed the film, as was verified by observing an increase in triplet annihilation when *gaseous* Freon at atmospheric pressure was poured into the Dewar. However, the number of triplet annihilations observed when either *liquid* nitrogen or *liquid* Freon filled the Dewar was of the same order as that expected from an equal number of positrons stopping in a solid metal.

Thus, in the liquid state, even substances like nitrogen or Freon whose chemical properties are known to favor the formation of positronium do not show the presence of triplet positronium in appreciable amounts. This could be due to either of two reasons: (1) either positronium is not formed at all in these liquids because of lack of space between the molecules of a condensed material, or (2) positronium is formed but it is rapidly

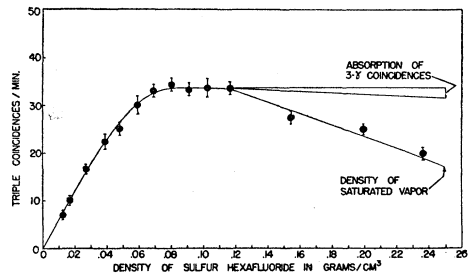

FIG. 6. Rate of 3-γ coincidences *vs* density in SF_6.

converted from triplet to singlet in molecular collisions (an effect already evident in the vapors at high pressure).

Recent studies on the half-lives of positrons in condensed materials[18] seem to favor the second of these explanations. If this second view is accepted, one can compute from the information on half-lives that the number of triplet annihilations in certain substances should be somewhat larger than in metals (3:1 for Teflon/Al; 1.6:1 for quartz/Al). In order to test this idea experimentally, it was necessary to measure the triplet annihilation rate with far greater accuracy than in the above experiments with liquefied gases.

Thus,[19] two Na^{22} sources of nearly equal intensity were prepared, one of them being deposited on and surrounded by aluminum, the other by Teflon, and the number of triplet annihilations was measured in a series of alternate runs. After reduction to the same source strength the observed ratio Teflon/Al was 1.94 ± 0.21. A similar experiment yielded 1.58 ± 0.28 for the ratio quartz/Al.

These results are in agreement with similar work performed at other laboratories[20] and confirm the presence of positronium in solids, at least in the case of certain insulators.

[16] J. Wheatley and D. Halliday, Phys. Rev. **88**, 424 (1952).

[17] M. Deutsch, Massachusetts Institute of Technology Progress Report of the Laboratory of Nuclear Science and Engineering, August 31, 1951 (unpublished).

[18] R. E. Bell and R. L. Graham, Phys. Rev. **90**, 644 (1953).

[19] The experiment was performed by Mr. L. Leipuner, whom we thank for permission to present his unpublished results.

[20] A. T. Stewart (private communication); T. A. Pond, Phys. Rev. **91**, 455 (1953).