# Direct Quantitative Observation of the Three-Photon Annihilation of a Positron-Negatron Pair*

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Three-photon annihilation of the positron with a negatron has been determined quantitatively as well as qualitatively by the simultaneous observation of the emitted photons with scintillation counters. The ratio of the reaction cross sections for two- and three-photon annihilation has been determined as $\sigma_{2k}/\sigma_{3k}=402\pm50$. This is in agreement with the theory of Ore and Powell but definitely differs from the theoretical values obtained by Lifshitz and by Ivanenko and Sokolov.

## INTRODUCTION

THE possibility that an appreciable part of positron-negatron reactions might result in annihilation with the radiation of three photons, was first theoretically postulated by Lifshitz[1] and by Ivanenko and Sokolov[2] and a short while later by Ore and Powell.[3] They all used the time-independent perturbation theory to compute the cross section for three-photon annihilation. The influence of Coulomb binding was neglected and plane wave functions were assumed for the initial and final states of the positron-negatron system. Similar results were obtained but with different numerical values.

When the positron and negatron meet in free space they can be considered to form a bound system similar to that of the hydrogen atom, as suggested by Wheeler.[4] The triplet or singlet state is formed depending on whether the spins of the positron and negatron are parallel or antiparallel. These states are called respectively *ortho*- and *para*-positronium. Transitions between the two are strictly forbidden.[3]

The singlet state is annihilated with the emission of two quanta, while the triplet state has to emit three quanta in order to conserve the total spin. Because of the multipole nature of the last process, *ortho*-positronium has a lifetime ($1.4\times10^{-7}$ sec[3]) markedly longer than that of *para*-positronium ($1.25\times10^{-10}$ sec[4]), and threefold annihilation has a correspondingly smaller chance of occurring.

The optical spectrum of positronium has not yet been observed,[5] but the formation of the triplet atom in gases has been observed experimentally by Deutsch[6] with $Na^{22}$ as the positron emitter. In a gas at normal temperature and pressure, the positronium atom will undergo thermal collisions at the rate of the order of $10^{12}$ per second. This may result in the de-excitation of the triplet state to the singlet state, with resulting two- instead of three-photon annihilation, in a gas (such as NO) where electron exchange takes place easily. The number of delayed ($\sim10^{-7}$ sec) coincidences between the emission of the gamma quantum from the decay of the $Na^{22}$ nucleus and the appearance of an annihilation quantum when the positron is brought to rest in the gas, has been measured by Deutsch in different gas mixtures. In the case of nitrogen, for example, the number of delayed coincidences—due to the formation of *ortho*-positronium—is markedly decreased by the addition of a few percent of NO. The electrons from the positronium atom are easily exchanged during a collision with an unpaired electron (from the NO) with opposite spin. Furthermore, by observing the number of delayed coincidences from positron capture in freon (where this exchange is almost nonexistent) as a function of the pressure and extrapolating to zero pressure, Deutsch found for the lifetime of the *ortho*-positronium a value in good agreement with the theoretical value of Ore and Powell.

When the positrons are brought to rest in a solid no positronium is formed. The smallest Bohr radius of the positronium atom is about one angstrom,[7] while the lattice distances in a crystal are of the order of only a few angstroms. Because of the intense perturbing influence of the ionic and electronic fields that it would experience, a positronium atom could not exist in a solid. The positron moves through the substance till it is brought to rest and then it is annihilated by a negatron without any real binding by any one of them. (According to Heitler,[8] the chance that a positron with an energy of 0.55 Mev, from $Na^{22}$, will be destroyed before being brought to rest, is about 3 percent.) The particles which are annihilated can therefore be regarded as quite free and the ratio of two- to three-photon annihilation therefore equals the ratio of the reaction cross sections, i.e., $\sigma_{2k}/\sigma_{3k}$.

* This work forms part of a thesis submitted in August, 1953, to the University of Pretoria in partial fulfillment of the requirements for the degree of D.Sc.

[1] E. M. Lifshitz, Doklady Akad. Nauk. S.S.S.R. **60**, 207 (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. A. Wheeler, Ann. N. Y. Acad. Sci. **48**, 219 (1946).
[5] J. D. Fox and J. Korringa, Ned. Tijdschr. Natuurk. **18**, 304 (1952).
[6] M. Deutsch, Phys. Rev. **82**, 455 (1951); **83**, 866 (1951).
[7] The Bohr radius of the hydrogen atom is $a=\hbar^2/me^2=0.525$A. The effective mass of the electron in the positronium atom is half of that in the hydrogen atom. Its Bohr radius is therefore 1.05A.
[8] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1944), p. 231.

The first direct observation of this three-photon process was made by Rich,[9] who obtained threefold coincidences from three scintillation counters spaced around a $Cu^{64}$ source of positrons. He did not obtain quantitative results accurate enough to discriminate between the various theoretical results. With the aid of NaI(Tl) counters, De Benedetti and Siegel[10] measured the three-photon disintegration rate of $Na^{22}$ positrons, stopped in aluminum. Owing to lack of knowledge of source strength and counter efficiencies, they could only report a correspondence within a factor of two with the theory of Ore and Powell.[10a]

It was therefore considered worthwhile to make an accurate, direct determination of $\sigma_{2k}/\sigma_{3k}$, since it would, apart from its relation to positron-negatron annihilation, yield a method of testing the theoretical foundation of the computations.

## CALCULATIONS

The different ways in which a positron-negatron pair can be annihilated are determined by the laws of energy and momentum conservation. In the case of three-photon annihilation these laws reduce to

$$\text{energy conservation: } k_1+k_2+k_3=2m; \tag{1}$$

$$\text{momentum conservation: } \mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3=0. \tag{2}$$

Figure 1 gives the spectrum of the individual photons according to Ore and Powell. The type of annihilation with the greatest probability is that in which one quantum has an energy $m$ and the other two are emitted in the opposite direction. This form of annihilation is impractical to observe due to the intense background of two-photon annihilation. In this work a symmetrical

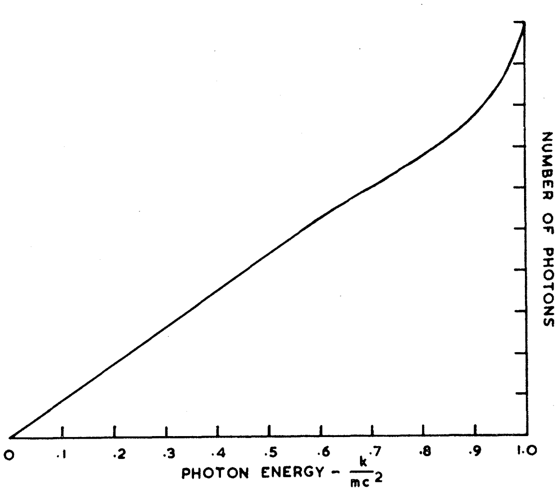


FIG. 1. Energy spectrum of individual photons from the three-photon annihilation of an electron and a positron—Ore and Powell.

[9] J. A. Rich, Phys. Rev. **81**, 140 (1951).
[10] S. De Benedetti and R. Siegel, Phys. Rev. **85**, 371 (1952).
[10a] For references to further work on this subject, see M. Deutsch in *Progress in Nuclear Physics*, edited by O. R. Frisch (Academic Press, New York, 1953), Vol. 3, p. 131.

120° setup has been chosen as it also simplifies the calculation of the fraction of all threefold annihilations observed with our experimental setup. In this case the three photons have the same energy, i.e., $\frac{2}{3}m=340$ kev.

Equation (2) of Ore and Powell[3] gives the probability of three-photon annihilation in terms of the matrix $H_{FA}$ (which gives the relation between the initial and final states of the system) and $\rho_F$ (the phase density of the final states per unit energy interval)

$$dP=2\pi|H_{FA}|^2\rho_F. \tag{3}$$

Substituting the values of $|H_{FA}|^2$ and $\rho_F$ from their Eqs. (4) and (5) gives

$$dP=\frac{2e^6\kappa^3}{3\pi^2m^4}[(1-\cos\alpha)^2+(1-\cos\beta)^2+(1-\cos\gamma)^2]\times dk_1dk_2d\Omega_1,$$

where $\alpha$, $\beta$, and $\gamma$ are the angles between $k_2$ and $k_3$, $k_3$ and $k_1$, and $k_1$ and $k_2$, respectively, and $1/\kappa$ is twice the Bohr radius. The term $dP$ is the probability that the positron and negatron are annihilated to give three photons such that the energy of photon 1 lies between $k_1$ and $k_1+dk_1$, with direction in the small solid angle $d\Omega_1$ and the energy of photon 2 between $k_2$ and $k_2+dk_2$. These three conditions completely determine the manner of annihilation.

To normalize, we have to divide $dP$ by $P$ the total probability of threefold annihilation which, according to Ore and Powell, is

$$P=\frac{1}{{}^3\tau}=\frac{16e^6\kappa^3}{9\pi m^2}(\pi^2-9).$$

This gives $dW$, the fraction of all threefold annihilation within the limits $dk_1$, $dk_2$, and $d\Omega_1$,

$$dW=dP/P$$

$$=\frac{3}{8\pi m^2(\pi^2-9)}[(1-\cos\alpha)^2+(1-\cos\beta)^2+(1-\cos\gamma)^2]dk_1dk_2d\Omega_1. \tag{4}$$

Resolving (2) into its $X$ and $Y$ components (with the $X$ axis in the direction of $k_1$ (see Fig. 2) gives

$$k_1+k_2\cos\gamma+k_3\cos\beta=0,$$

$$k_2\sin\gamma-k_3\sin\beta=0.$$

(All the $Z$ components disappear as the $X$-$Y$ plane is chosen so that the three photons fall in it.) Furthermore $\alpha+\beta+\gamma=2\pi$. Therefore

$$k_1=\frac{2m\sin\alpha}{\sin\alpha+\sin\beta+\sin\gamma},$$

$$k_2=\frac{2m\sin\beta}{\sin\alpha+\sin\beta+\sin\gamma},$$

$$k_3=\frac{2m\sin\gamma}{\sin\alpha+\sin\beta+\sin\gamma}.$$

This gives $k_1$ and $k_2$ as functions of $\alpha$ and $\beta$. Equation (4) can now be transformed by means of the Jacobian,

$$\frac{\partial(k_1,k_2)}{\partial(\alpha,\beta)}=\frac{4m^2\sin\alpha\,\sin\beta\,\sin\gamma}{(\sin\alpha+\sin\beta+\sin\gamma)^3},$$

and becomes

$$dW=\frac{3}{2\pi(\pi^2-9)}[(1-\cos\alpha)^2+(1-\cos\beta)^2+(1-\cos\gamma)^2]\times\frac{\sin\alpha\,\sin\beta\,\sin\gamma}{(\sin\alpha+\sin\beta+\sin\gamma)^3}d\alpha d\beta d\Omega. \quad (5)$$

Since $d\alpha$, $d\beta$, and $d\Omega$ are small angles in this experimental setup, the probability can be assumed to be constant over the range $d\alpha$, $d\beta$, and $d\Omega$ and equal to that at $\alpha=120°$ and $\beta=120°$. Substituting in (5) gives

$$dW=\frac{3}{8\pi(\pi^2-9)}d\alpha d\beta d\Omega. \quad (6)$$

Now $\alpha$ and $\beta$ can vary from 0 to $2\pi$ and $\Omega$ from 0 to $4\pi$. Therefore the fraction of the total $\alpha,\beta,\Omega$ phase space observed is

$$B=\frac{1}{16\pi^3}d\alpha d\beta d\Omega. \quad (7)$$

This gives the fraction of all random threefold annihilations which can be observed with our setup, assuming the probability to remain constant within its limits. Furthermore, the three photons are emitted in the same plane. This factor $B$ is now calculated by means of elementary coordinate geometry as follows:

A random plane intersecting all three counters is given by

$$lx+my+nz=0, \quad (8)$$

where $l$, $m$, and $n$ are the directional cosines of its orthogonal. Also,

$$l^2+m^2+n^2=1. \quad (9)$$

The equations for the circular front surfaces of the counters are:

Counter 1:

$$x=R,\quad y^2+z^2=r^2, \quad (10)$$

where $R$ equals the distance from source to counter and $r$ the radius of each counter;

Counter 2:

$$x-\sqrt{3}y+2R=0,\quad x^2+y^2+z^2=R^2+r^2; \quad (11)$$

Counter 3:

$$x+\sqrt{3}y+2R=0,\quad x^2+y^2+z^2=R^2+r^2. \quad (12)$$

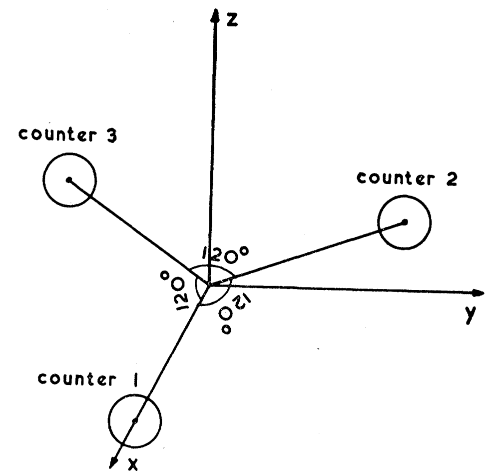


FIG. 2. Positions of front surfaces of scintillation counters with respect to our coordinate system.

The random plane now intersects the three counters in lines $AB$, $CD$, and $EF$. Solving for $AB$ from Eqs. (8), (9), and (10) gives

$$AB=4\left(r^2-\frac{l^2}{1-l^2}R^2\right).$$

Similarly,

$$CD=4\left[r^2-\frac{(l-\sqrt{3}m)^2}{4-(l-\sqrt{3}m)^2}\cdot R^2\right],$$

$$EF=4\left[r^2-\frac{(l+\sqrt{3}m)^2}{4-(l+\sqrt{3}m)^2}\cdot R^2\right].$$

The fraction of all three-photon annihilations, in one specific plane, through the three counters, is

$$6\times\frac{AB}{2\pi R}\times\frac{CD}{2\pi R}\times\frac{EF}{2\pi R},$$

where $AB$, $CD$, and $EF$ are functions only of $l(=\cos\phi)$ and $m(=\cos\theta)$. The factor 6 is introduced as there are 6 possible ways of annihilation giving photons in the three directions. The fraction of all planes with directional angles between $\phi$ and $\phi+d\phi$, and $\theta$ and $\theta+d\theta$ is therefore

$$dB=6\times\frac{AB}{2\pi R}\times\frac{CD}{2\pi R}\times\frac{EF}{2\pi R}\times\frac{d\theta}{2\pi}\times\frac{d\phi}{2\pi}.$$

The total fraction observed by our setup is

$$B=\frac{6}{(2\pi)^5}\int\int\frac{AB\cdot CD\cdot EF}{R^3}d\theta d\phi. \quad (13)$$

This integration takes place over the ranges of $\theta$ and $\phi$ where the plane still cuts all three counters. At the limits the plane touches one or more of the counters.

For Counter 1, these boundaries are given by

$$AB=4\left(r^2-\frac{l^2}{1-l^2}R^2\right)=0.$$

As $l$ and $m$ are the cosines of large angles, $l^2\ll 1\gg m^2$. Also, we make the substitution $r/R=f$. Then for the limits determined by Counter 1,

$$l=\pm f.$$

Similarly, the limits determined by Counter 2 are

$$l-\sqrt{3}=\pm 2f;$$

and the limits determined by Counter 3 are

$$l+\sqrt{3}m=\pm 2f.$$

Now $l=\cos\phi$ and therefore $d\phi=-dl/(1-l^2)^{\frac{1}{2}}\approx -dl$; similarly $d\theta\approx -dm$. Substituting further $a=l/f$ and $b=\sqrt{3}m/f$ in (13) gives

$$B=\frac{12f^5}{\sqrt{3}(2\pi)^5}\int\int\{(1-a^2)[4-(a-b)^2]\times[4-(a+b)^2]\}^{\frac{1}{2}}dbda,$$

where the integration takes place within the limits given by

$$a=\pm 1,\quad a-b=\pm 2,\quad a+b=\pm 2.$$

Numerical integration gives

$$B=\frac{12f^5}{\sqrt{3}(2\pi)^5}\times 13.8. \tag{14}$$

Substituting (7) and (14) into (6) gives

$$dW=\frac{3}{8\pi(\pi^2-9)}\times 16\pi^3 B$$
$$=0.665f^5.$$

Now the fraction of space subtended by each counter is $\Delta\Omega/4\pi=\pi r^2/4\pi R^2=\frac{1}{4}f^2$. Therefore,

$$dW=21.3(\Delta\Omega/4\pi)^{5/2}.$$

The probable error of this calculation is less than 2 percent.

## EXPERIMENTAL PROCEDURE

Three NaI(Tl) scintillation counters were placed around the $Na^{22}$ source (as NaCl) in a container of aluminum which was thick enough to stop all the positrons. The 2.5-cm×4-cm diameter crystals were mounted directly on the 5819 photomultipliers with silicone grease and MgO as reflector. Jordan-Bell amplifiers were used with shorted delay-line shaping of the input pulses and connected to conventional differential discriminators. The output pulses from the discriminators were fed into a threefold coincidence circuit. It was found that the resolving time could be decreased to $4.5\times 10^{-7}$ sec before losing any genuine coincidences.

The number of genuine coincidences, observed with this setup, is given by

$$K_3=\frac{\sigma_{3k}}{\sigma_{2k}+\sigma_{3k}}Ne_1e_2e_3dW, \tag{15}$$

where $N$=disintegration rate of $Na^{22}$ source, $e_1$=efficiency of Counter 1, $e_2$=efficiency of Counter 2, and $e_3$=efficiency of Counter 3. Substituting the value that we obtained for $dW$ gives

$$\frac{\sigma_{2k}}{\sigma_{3k}}=\frac{N\times e_1e_2e_3}{K_3}\times 21.3\left(\frac{\Delta\Omega}{4\pi}\right)^{5/2}-1.$$

Each parameter on the right-hand side of this equation was determined separately.

### Determination of $N$

The disintegration rate of the $Na^{22}$ was determined with a $4\pi$ counter as $0.436\pm 0.013$ rutherford.

### Determination of $\Delta\Omega/4\pi$

The solid angles subtended at the source by the different counters were first made equal. Two counters were placed on opposite sides of the source. Counter 1 was fixed at about 9 cm distance while the position of counter 2 could be read on a scale. The windows of both discriminators were adjusted on the 510-kev annihilation peak, and twofold coincidences were observed for various distances from Counter 2 to source. When the solid angle of Counter 2 is bigger than that of Counter 1, the coincidence counting rate is determined by Counter 1 and is constant. As soon as Counter 2 is moved farther away from the source, the coincidence counting rate is determined by the solid angle of Counter 2 and the counting rate drops. Counter 2 could therefore be adjusted to a position with solid angle equal to that of Counter 1. Now the individual counting rates are given by

$$N_1'=2Ne_1'\Delta\Omega/4\pi,$$
$$N_2'=2Ne_2'\Delta\Omega/4\pi,$$

where $e_1'$ is the efficiency of Counter 1 for 510-kev photons and $e_2'$ is the efficiency of Counter 2 for 510-kev photons. The coincidence rate is

$$K_{12}=2Ne_1'e_2'\Delta\Omega/4\pi,$$

as every photon into Counter 1 is accompanied by one in the direction of Counter 2. This gives

$$\Delta\Omega/4\pi=N_1'N_2'/2NK_{12}.$$

Now Counter 2 is moved to the 120° position such that its counting rate, and therefore its solid angle, remains the same. The accidental twofold coincidence rate is determined in this position. The expression $\Delta\Omega/4\pi$ was found to be $1.41\times10^{-2}$.

The same procedure was repeated with Counters 1 and 3, giving $\Delta\Omega/4\pi=1.38\times10^{-2}$. The average value was taken as $(1.40\pm0.04)\times10^{-2}$.

### Determination of $e_1$, $e_2$, and $e_3$

The efficiencies of the three counters for 340-kev radiation were determined with an $I^{131}$ source emitting 364-kev photons instead of the $Na^{22}$. The $I^{131}$ source had been calibrated with the $4\pi$ counter. It emits the following gamma rays with intensities as indicated:[11] 80 kev—2.17 percent; 163 kev—<0.5 percent; 284 kev—5.3 percent; 364 kev—80.0 percent; 637 kev—9.3 percent; 722 kev—2.8 percent.

With the window of the differential discriminators adjusted to the region from 200–450 kev, the 80-kev and 163-kev photons could be neglected. The scintillation spectra of the 722-kev and 637-kev photons were extrapolated from the high-energy region to 200 kev in analogy with the spectrum of the 662-kev photons from $Cs^{137}$ and with due regard to their relative intensities. Only the photopeak of the 284-kev photons falls inside the discriminator window and this peak was drawn from the knowledge of its intensity and the resolution of the scintillation counter (see Fig. 3). The area of these three curves falling inside the region 200–450 kev was subtracted from the total gamma spectrum, giving the fraction due to the 364-kev photons as 0.88. Now the efficiency of the $n$th counter is given by

$$e_n=\frac{0.88N_n}{0.80D\times\Delta\Omega/4\pi},$$

where $N_n$ is the counting rate of scintillation counter $n$ and $D$ is the disintegration rate of the $I^{131}$ source.

The efficiencies of the three counters were found to be $e_1=0.357$, $e_2=0.305$, and $e_3=0.369$ (6 percent probable error). It was then assumed that the efficiencies for 340-kev photons would be near enough to these values—after adjusting the bias of the discriminator 24 kev lower.

### Determination of $K_3$

Threefold coincidences were determined with the discriminator bias adjusted to the 340-kev photopeak—the window width remaining the same. The random coincidence rate was determined with one of the counters lifted out of the plane but with exactly the same individual counting rate. We obtained a total counting rate (coplanar) of

$$K_3+T_3=2.93\pm0.12 \text{ per minute,}$$

[11] R. E. Bell and R. L. Graham, Phys. Rev. **86**, 212 (1952).

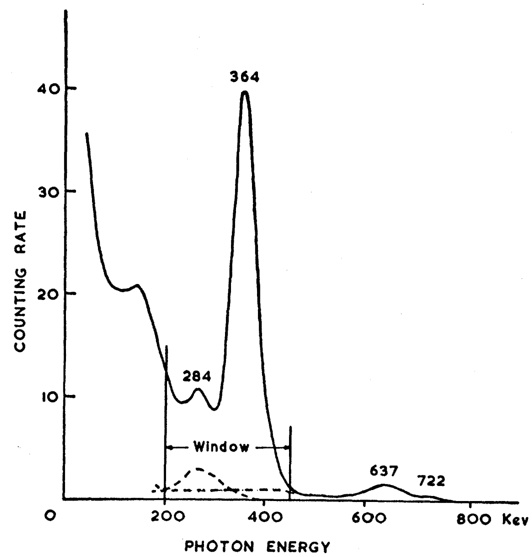


FIG. 3. Pulse distribution of photons from $I^{131}$ showing the window of the differential discriminator as well as the extrapolated 284-, 637-, and 722-kev spectra.

and a random counting rate (out of plane) of

$$T_3=1.65\pm0.09 \text{ per minute.}$$

Therefore, the true threefold coincidence rate is

$$K_3=1.28 \text{ per minute.}$$

Substituting these values in (15) gave

$$\sigma_{2k}/\sigma_{3k}=402\pm50.$$

### DISCUSSION

The difference in threefold coincidence counting rate with one counter in or out of the plane cannot be ascribed to anything else than the emission of three simultaneous quanta by the source. The annihilation of the positron by a negatron, with emission of three quanta instead of the usual two, has therefore been quantitatively confirmed by direct observation of the three quanta.

The following values for the ratio $\sigma_{2k}/\sigma_{3k}$ between the cross sections for two- and three-photon annihilation have been obtained previously:

#### Theoretically

| | |
|---|---|
| Ore and Powell[3] | 370 |
| Lifshitz[1] | 235 |
| Ivanenko and Sokolov[2] | 1670 |

#### Experimentally

Rich[9] and De Benedetti and Siegel[10] observed the three-photon annihilation directly, but could only state that the order of magnitude of the effect corresponded with the value of Ore and Powell.

Deutsch[6] determined the lifetime of *ortho*-positronium as observed in freon. He obtained

$$1/{}^3\tau = (6.8\pm0.7)\times10^6 \text{ sec}^{-1}.$$

Therefore,

$$\sigma_{2k}/\sigma_{3k} = \tfrac{1}{3}\,{}^3\tau/{}^1\tau = 398\pm40,$$

which is in agreement with our value.

The theory of Ore and Powell for the three-photon annihilation of a positron-negatron pair has therefore been directly confirmed. The numerical results of Lifshitz and Ivanenko and Sokolov are definitely in disagreement with our experimental results.

The author would like to express his indebtedness to the South African Council for Scientific and Industrial Research for permission to publish this paper, and to Dr. S. J. du Toit, head of the Nuclear Physics Division, for suggesting this problem.

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# Fission Yield Fine Structure in the Mass Region 99–106*

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Fission yield measurements were carried out in the mass region 99 to 106 to investigate previously postulated fine structure related to nuclear closed shells. Radiochemical data were obtained in this mass region in the fission reactions $U^{235}(n,f)$, $U^{238}(\gamma,f)$, $U^{235}(d,f)$, and $U^{238}(d,f)$ for the yields of $Mo^{99}$, $Mo^{101}$, $Mo^{102}$, $Ru^{103}$, $Ru^{105}$, and $Ru^{106}$. Fine structure spikes were found in all reactions. It was found that the mass position of the spikes can be explained by the assumption that primary fission fragments with 82 neutrons or 50 protons and their complements have an enhanced yield as a result of selectivity in the fission process itself. Agreement was also found with two previously known reactions: $U^{233}(n,f)$ and $Cm^{242}$ spontaneous fission. Fine structure is also predicted for several other fission reactions.

## I. INTRODUCTION

SINCE the discovery of fission in 1939, a great deal of work has been done by nuclear chemists to investigate the problem of the distribution of mass between the fission fragments. It was found quite early that fission is asymmetric, and that the distribution of mass could be described,[1] at least approximately, by a smooth curve with two broad maxima. This fact has been verified many times, for many fission reactions, and although the shape and mass position of the yield-mass curve may change somewhat, all cases of low- and medium-energy fission seem to follow the same type of mass distribution.

Several significant departures from the smooth distribution curve have been discovered, however, and in certain mass regions the smooth curve is found to be totally inadequate. The earliest indication of anomalous yields was reported by Thode and Graham,[2] who found that the measured yields of $Kr^{84}$ and $Xe^{134}$ were about 35 percent too high to lie on any continuous smooth curve. Stanley and Katcoff[3] found the cumulative yield of $I^{136}$ in the thermal-neutron fission of $U^{235}$ to be 3.1 percent, while the next member of the same chain, $Xe^{136}$, whose independent fission yield was estimated[4] to be about 0.4 percent, had been found by Thode and Graham[2] to have a cumulative yield of 6.1 percent. This indicated a much higher independent yield for $Xe^{136}$ than could easily be explained. Still another anomalous fission yield was found by Macnamara, Collins, and Thode,[5] in that the fission yield of $Xe^{133}$ was found to be 6.3 percent instead of the expected 5.0 percent. Recent studies[6,7] have led to somewhat higher absolute values for the xenon fission yields.

These so-called fine-structure phenomena, as known up to that point, were explained satisfactorily by Glendenin.[8] He postulated that in all cases where a primary fission fragment contains one neutron in excess of a closed shell (i.e., contains 51 or 83 neutrons) this extra loosely bound neutron is immediately lost. This neutron loss, considered to be entirely independent of

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Presented in partial fulfillment for the Ph.D. degree in the Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts, November, 1953. The findings of this study were reported briefly at the Hauptversammlung of Die Deutsche Bunsen Gesellschaft, Bayreuth, Germany, May 28–29, 1954; to be published in Z. Electrochemie.

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1 See, for example, C. D. Coryell and N. Sugarman, editors, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Vol. 9, Div. IV.

2 H. G. Thode and R. L. Graham, Can. J. Research **A25**, 1 (1947).

3 C. W. Stanley and S. Katcoff, J. Chem. Phys. **17**, 653 (1949).

4 Glendenin, Coryell, and Edwards, Paper 52 of reference 1.

5 Macnamara, Collins, and Thode, Phys. Rev. **78**, 129 (1950).

6 S. Katcoff and W. Rubinson, Phys. Rev. **91**, 1458 (1953).

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