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A Direct Measurement of the Ratio of the Reaction Cross Sections for Two-Photon and Three-Photon Positron Annihilation in Aluminum and Silver

Robert Maurice Heavenrich

A DIRECT MEASUREMENT OF THE RATIO OF THE REACTION CROSS SECTIONS FOR TWO-PHOTON AND THREE-PHOTON POSITRON ANNIHILATION IN ALUMINUM AND SILVER

by

Robert Maurice Heavenrich, Jr.

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INTRODUCTION

The probability of annihilation of a positron with its antiparticle, an electron, is spin dependent. Since both particles have spin component one half, there are two possible relative orientations of their spins : they can either be aligned (parallel) in the ^3S, triplet, state with total spin component $|S_z| = \frac{1}{2}\hbar - \frac{1}{2}\hbar = 1\hbar$ or they can be opposite (antiparallel) in the ^1S, singlet, state with $|S_z| = \frac{1}{2}\hbar - \frac{1}{2}\hbar = 0$. To conserve energy and angular momentum, annihilation from the triplet state must result in the creation of an odd number of photons -- predominately three. Similarily, annihilation from the singlet state must result in the creation of an even number of photons -- predominately two.

If the center of mass of the positron and electron is at rest when annihilation occurs, energy-momentum conservation requires, in the two-photon case, that the photons be emitted in opposite directions with equal energy $m_o c^2$ where m_o is the rest mass of the electron and c is the velocity of light. In the three-photon case, the photons are constrained to be in the same plane, but may have many combinations of energies and orientations in that plane provided no photon has an

energy greater than m_0c^2 and no more than two photons are in the same half plane.

Prior to annihilation a positron can capture an electron to form a bound hydrogen-like system called "positronium." Annihilation from an excited state of positronium is unlikely since in such a state the positron and electron wave functions do not overlap sufficiently. Accordingly, positronium formed in an excited state must de-excite through radiative emission or collision to either the ^3S, triplet, state (orthopositronium) or the ^1S, singlet, state (parapositronium) before annihilation can occur. Since orthopositronium can be expected to form three times as often as parapositronium, 75% of the positronium "atoms" would annihilate with the emission of three photons provided the ratio of the spin states was maintained. Through collisions with atoms or molecules in a medium, or in the presence of a magnetic field, orthopositronium can be converted (quenched) to parapositronium. Thus, while positronium formation tends to enhance the three-photon yield, the ratio of three- to two-photon events from positronium may be reduced by quenching.

Positronium formed in a solid will have a distorted wave function, due to the ionic fields, and cannot be expected to behave as if it were in free space. The

smallest Bohr radius for positronium is about an angstrom, while the lattice spacing in a crystal is on the order of a few angstorms. Positronium formation in metals, moreover, has long been considered unlikely. Since the free (ie. conduction) electron density is large, the positron interacts simultaneously with several electrons without becoming bound to any one of them. Therefore, the positrons annihilating in metals can be regarded as unbound and hence, the ratio of two- to three-photon annihilation rates should be that predicted for annihilation with free electrons.

The first theoretical calculations of the reaction cross section for free three-photon annihilation of an electron-positron pair with small relative velocity were made in 1948 by Lifshitz[1], and Ivaneko and Sokolov[2], and in 1949 by Ore and Powell[3]. Although all these authors used the same physical assumptions -- that is, they all used time-dependent perturbation theory, neglected Coulomb binding, and used plane wave functions -- they obtained different results. The various values of σ_2/σ_3, the ratio of the reaction cross sections for two-photon to three-photon free

annihilation and of λ_o the three-photon decay rate are :

	$\frac{\sigma_2}{\sigma_3}$	λ_o
Ivaneko and Sokolov	1670	0.16×10^7 sec^{-1}
Lifshitz	235	1.12×10^7 sec^{-1}
Ore and Powell	370	7.20×10^7 sec^{-1}

Ore and Powell's calculation was later repeated by Radcliffe[4] and by Drisko[5]. Their value of the decay rate was first verified in 1951 by Deutsch[6] who measured, as a function of pressure, positron lifetimes in freon. By extrapolation to zero pressure, he obtained a value for the decay rate of $(6.8 \pm 0.7) \times 10^7$ sec^{-1} or in terms of the ratio of the reaction cross sections $\sigma_2/\sigma_3 = 398 \pm 40$.

The detection of singlet (two-photon) positron annihilation is relatively simple. Since the two photons are emitted in nearly opposite directions with equal energy (511 keV), the rate of coincidence from two detectors, placed on a common axis with the sample at the center would give a measure of the two-photon annihilation rate.

Difficulties arise in detecting triplet (three-photon) free annihilation, not only because (according to Ore and Powell) it occurs 1/370 times as often as

two-photon annihilation, but also because there is a distribution in the energies the photons may have. The problem can, however, be simplified by placing the detectors symmetrically around the sample and coplanar with it. The annihilations observed are those where the photons emerge 120^{o} apart with equal energy ($2/3\ m_{o}c^{2}$).

The first direct observation of three-photon positron annihilation was reported in 1950 by Rich[7] who placed three anthracene and napthalene scintillation detectors symmetrically around a ^{64}Cu source enclosed by a sufficient thickness of aluminum to stop all the positrons. Rich attempted to determine σ_2/σ_3 from the two- and three-photon rates taking into account the detector efficiencies and the detectable fraction of the two- and three-photon events, but was unable to discriminate among any of the theoretical results.

In 1952 deBenedetti and R. Siegel made a preliminary report[8] of an experiment in which three symmetrically placed NaI(Tl) scintillation detectors were used to measure the three-photon coincidence rate of positrons from a ^{22}Na source annihilating in aluminum. At that time their knowledge of the source strength and the absolute detector efficiencies was too meager to permit more than a rough comparison with theory. In 1954, having made more accurate measurements of both the

source strength and detector efficiencies, they were able to report[9,10] that their measured value of .92 ± .10 counts/min was not "inconsistent" with a value predicted from Ore and Powell's work of 1.20 ± .16 counts/min.

In 1954 Graham and Stewart[11] reported an experiment in which an arrangement similiar to deBenedetti and R. Siegel's was used to measure three-photon annihilation rates in various substances. They found that within experimental uncertainty (about 33%) the six metals studied (Li, Be, Al, Cu, Au, and Pb) had "the same low counting rate " while insulators such a fused quartz, Polystyrene, and Teflon had annihilation rates several times higher. Graham and Stewart did not correct their data for the loss of photons due to scattering and absorption in the sample or for annihilations in the source material and did not attempt a comparison with theory.

In 1965 Bertolaccini et al.[12] reported a similar experiment in which they measured three-photon positron annihilation lifetimes and yields in various metals and insulators. Bertolaccini et al. made a relative measurement and compared the yield for each substance with that of aluminum. While the three-photon yields for ten of the metals (Be, V, Fe, Cu, Zn, Mo, Pd, W, Ir, and Au) were in good agreement with the aluminum data, the

yields for the other five metals (Ni, Ag, Cd, Pt, and Pb) were ten to sixteen percent higher than that of aluminum, a difference larger than the reported experimental uncertainty (about two or three percent). As in Graham and Stewart's experiment, the yields for the insulators were considerably higher than that of aluminum.

The source used by the Bertolaccini group (about 40 microcuries of ^{22}Na sealed in a thin Moplefan foil) was put in a "composite sandwich of four targets, two of which, ie. those of Al, operated as standard targets." They measured the three-photon coincidence rate with the "specimen under investigation inside and the Al outside and conversely," and then assumed that the loss of photons due to absorption and scattering was the same with both arrangements. To minimize such losses, their samples had the same size and thickness (200 mg/cm^2). To compare the three-photon yield in each substance with that of aluminum, the data had to be corrected for backscatter of the positrons and for annihilations in the Moplefan foil.

Although Bertolaccini et al. did not measure σ_2/σ_3, they did compare their data with a simple model based on Ore and Powell's theory. From their measurements of the three-photon yields and lifetimes in insulators they found the positrons not forming positronium were as

effective in producing three-photon annihilations as those annihilating in aluminum. The three-photon yield for aluminum, moreover, turned out to be in "very good agreement" with what could be expected theoretically assuming annihilations with free electrons.

The results of the Bertolaccini experiment are similar to those reported in 1956 by Telegdi[13] who measured three-photon annihilation yields for aluminum, Teflon, and fused quartz in the presence of an external magnetic field. While the three-photon yield for aluminum was independent of the field intensity, the three-photon yields for the insulators were quenched in the presence of the field. The Telegdi experiment thus demonstrated :

(1) In metals all, or most positrons do not form bound systems.

(2) In insulators, a certain fraction of the positrons form positronium, the rest annihilate directly.

(3) Orthopositronium formed in insulators can be converted to parapositronium in the presence of an external magnetic field.

The most significant direct measurement of $\frac{\sigma_2}{\sigma_3}$ and the first accurate enough to distinguish among the various theoretical results was made in 1954 by Basson[14].

Like deBenedetti and R. Siegel, he used three NaI(Tl) detectors symmetrically placed around a ^{22}Na source in an aluminum container thick enough to stop all the positrons. From the three-photon coincidence rate, absolute source strength, absolute detector efficiencies, and the calculated fraction of the three-photon events detected, Basson obtained an experimental value for the ratio of the reaction cross sections of 402 ± 50. This compares favorably with Ore and Powell's theoretical value of 370 and Deutsch's measurement of 398 ± 40.

In calculating the fraction of all threefold annihilations which could be detected, Basson considered his sample as a point source. He did not, however, report the details of his sample. The correction for finite geometry, which he neglected, may have been appreciable, depending on the size of the sample. There is, moreover, no evidence that he corrected his data for the loss of photons due to scattering and absorption in his sample. If the sample thickness had been the minimum to stop all the positrons, the correction would have been about six percent and would bring his value for $\frac{\sigma_2}{\sigma_3}$ into closer agreement with that predicted by Ore and Powell.

In their calculation of the triple coincidence rate deBenedetti and R. Siegel corrected for the loss of

photons due to scattering and absorption, but in determining the geometric acceptance, they considered their sample as a point source. In both the deBenedetti-R. Siegel experiment and in Basson's experiment the random three-fold coincidence rate was measured with one of the detectors rotated 45^{o} out of the plane defined by the source and the other detectors. Thus, the background measurement corrected for coincidences caused by the 1.27 MeV gamma ray which accompanies the positron in the decay of ^{22}Na, but not for annihilations in the source material.

In 1969 J. Siegel[15] reported a measurement of σ_2/σ_3 in aluminum in which he used a geometry that shielded the detectors from the 1.27 MeV gamma ray and from annihilations in the source material. The present work is an extension of J. Siegel's and reports a measurement of σ_2/σ_3 in aluminum and silver using further improvements in the apparatus and technique.

APPARATUS AND PROCEDURE

J. Siegel used NaI(Tl) scintillation detectors to measure the two- and three-photon coincidence rates and then determined $\frac{\sigma_2}{\sigma_3}$ from :

$$\frac{\sigma_2}{\sigma_3} = \frac{N_2^{ij}\, e_i(340)\, e_j(340)\, e_k(340)\, C_3\, A_3}{N_3\, e_i(511)\, e_j(511)\, C_2\, A_2}$$

where :

N_2^{ij} = two-photon coincidence rate using detectors i and j

N_3 = three-photon coincidence rate

$e_i(340)$ = efficiency at 340 keV for detector i

$e_i(511)$ = efficiency at 511 keV for detector i

C_3 = three-photon solid angle factor

C_2 = two-photon solid angle factor

A_3 = three-photon absorption/scattering correction

A_2 = two-photon absorption/scattering correction

Since $e_i(340)/e_i(511)$ is just the relative efficiency for detector i for 340 and 511 keV photons, J. Siegel needed to know the relative efficiency for two of the detectors and the absolute efficiency of the third. In

Basson's experiment and also in the deBenedetti - R. Siegel experiment, the two-photon annihilation rate was not used directly in determining $\frac{\sigma_2}{\sigma_3}$ so that it was necessary to know the source intensity and the absolute efficiency at 340 keV for each of the three detectors.

J. Siegel's positron source consisted of about two millicuries of ^{22}Na deposited in the center of a one inch diameter stainless steel mount and covered by a .0002 inch stainless steel foil. ^{22}Na has a half life of 2.60 years and decays about 90% of the time by emitting a positron with E_{max} = .544 MeV. About .05% of the time, a positron of E_{max} = 1.8 MeV is emitted[16].

^{22}Na does have one distinct disadvantage in this type of experiment. The transition from the first excited state to the ground state of ^{22}Ne occurs in less than 10^{-11} seconds; hence, the 1.27 MeV gamma and the positron can be regarded as having been emitted simultaneously. Since the positron and electron annihilate within a very short period of time, the resulting photons may be in coincidence with the 1.27 MeV gamma ray. To shield the detectors against 1.27 MeV gamma and also against annihilations in the source material, J. Siegel's source mount was recessed one cm. into a 2.7 cm. diameter hole in a lead cylinder, 16 cm. high and 19 cm. in diameter.

J. Siegel's sample consisted of three .010 inch aluminum disks, each one inch in diameter. The disks were annealed at 550^{o} C for 24 hours to remove strains and defects, and then fastened together using three small dots of contact cement "between each surface at the edge." The sample was then attached to a plexiglass rod and located directly over the source, equidistant (15.0 cm.) from the detector faces. The plexiglass rod was supported by glass rods which were attached to the detector mounts. Whenever a detector was moved, the support rods had to be moved and the sample realigned.

In determining the three-photon annihilation rate, J. Siegel followed conventional lines and limited his measurement to the symmetrical case where the photons emerge 120^{o} apart and share the total energy of $2\ m_{o}c^{2}$ equally. The geometry used by J. Siegel (Figure 1A) did not have the spherical symmetry used by the previous investigators, but rather had cylindrical symmetry. Accordingly, the stray rate for the three-photon coincidence rate could not be measured by the conventional method of "rotating a detector out of the plane," but had to be measured by inserting delay lines.

With J. Siegel's geometry, the detectors were not shielded from annihilations in the air around, or under the sample. Since positronium formation is possible in

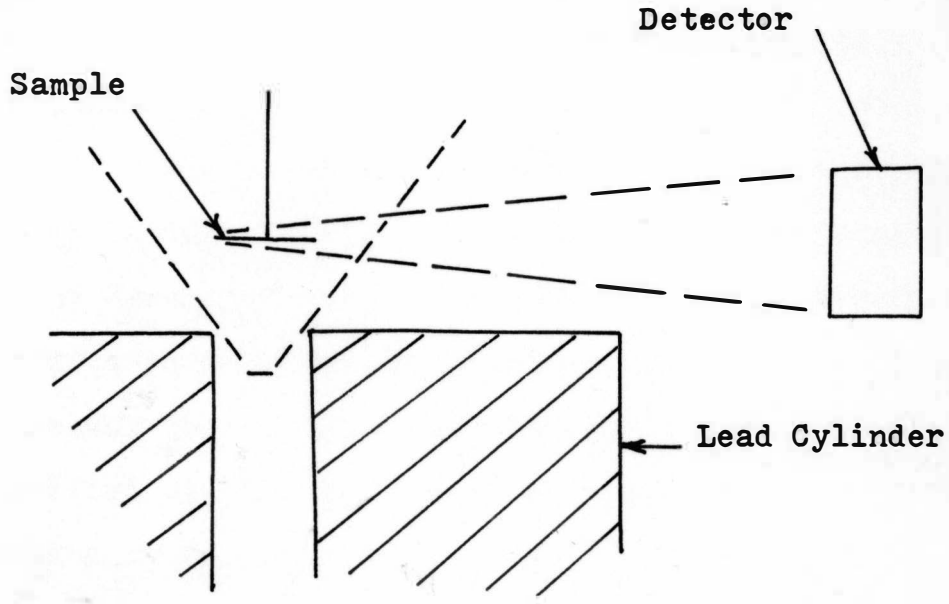

Figure 1A. Cross sectional view of J. Siegel's geometry.

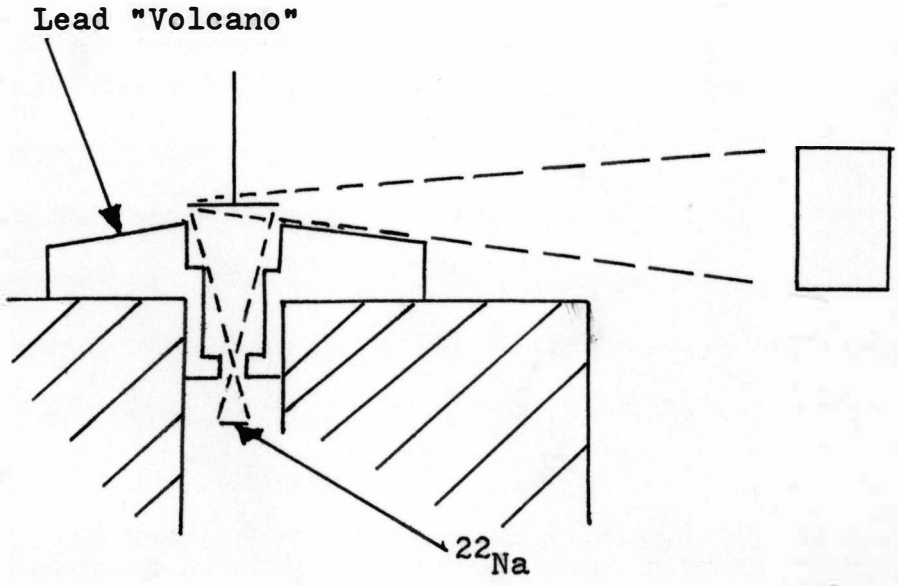

Figure 1B. Cross sectional view of lead "volcano" geometry.

air, such an experimental arrangement might have an enhanced three-photon yield. Accordingly, in the present work, the experiment was first attempted using a lead "volcano" (Figure 1B) to collimate the positron beam and shield the detectors from as many of the remaining annihilations in air as practical. To reduce spurious coincidences caused by Compton scattering between detectors, each detector was provided an additional lead shield. The supporting system for the sample was also modified so that each detector could be moved without having to realign the sample.

The source used in the present work consisted of about ten millicuries[1] of ^{22}Na deposited in the center of a one inch diameter stainless steel mount and covered by a .006 inch Be foil. As in J. Siegel's experiment, the source mount was recessed into a lead cylinder to shield the detectors from the 1.27 MeV gamma ray and from annihilations in or very near the source. Even with the lead volcano design, about twice as many three-photon events could be observed from the air for each one in the sample, making it impossible to determine the difference between the three-photon

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[1] As measured by the manufacturer, New England Nuclear, on Nov. 1, 1970. The data for the present work was taken during the first four months of 1972.

rates in aluminum and silver. The experiment, therefore, had to be performed with the sample under vacuum.

A cross sectional view of the vacuum chamber is shown in Figure 2 and a top view in Figure 3. The aluminum base plate was centered on the lead cylinder and held in place by three screws. To collimate the positron beam, the base plate extended, as shown, into the 2.7 cm. diameter hole in the lead cylinder. In order that the .006 inch Be foil that enclosed the ^{22}Na not be exposed to vacuum, the source was below (outside) the chamber. Those positrons that were headed towards the sample entered the chamber through a .001 inch Kapton foil epoxied to the base plate. Approximately 80% of the incident positrons were transmitted through the foil, the remainder either backscattered or annihilated in it. Annihilations in the ^{22}Na source, collimator, and Kapton foil did not contribute to the true two- and three-photon coincidence rates since they occurred well below the detectors. The singles background from such annihilations and from the 1.27 MeV gamma ray was, moreover, reduced by attenuation in the lead cylinder and the additional (movable) lead shielding shown in Figures 2 and 3.

The vacuum chamber walls were made from a piece of Lucite tubing that had an outer diameter of five inches,

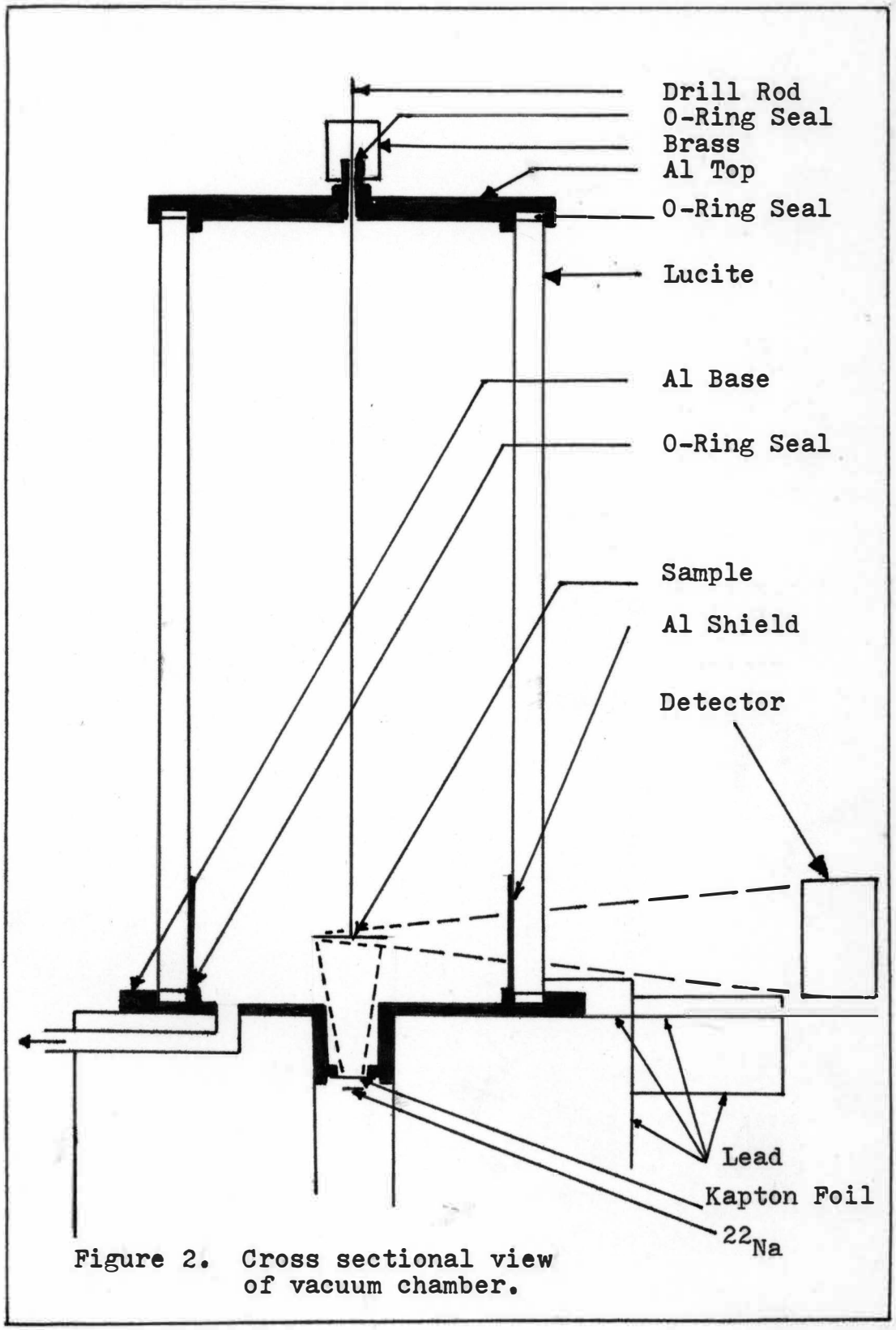


Figure 2. Cross sectional view of vacuum chamber.

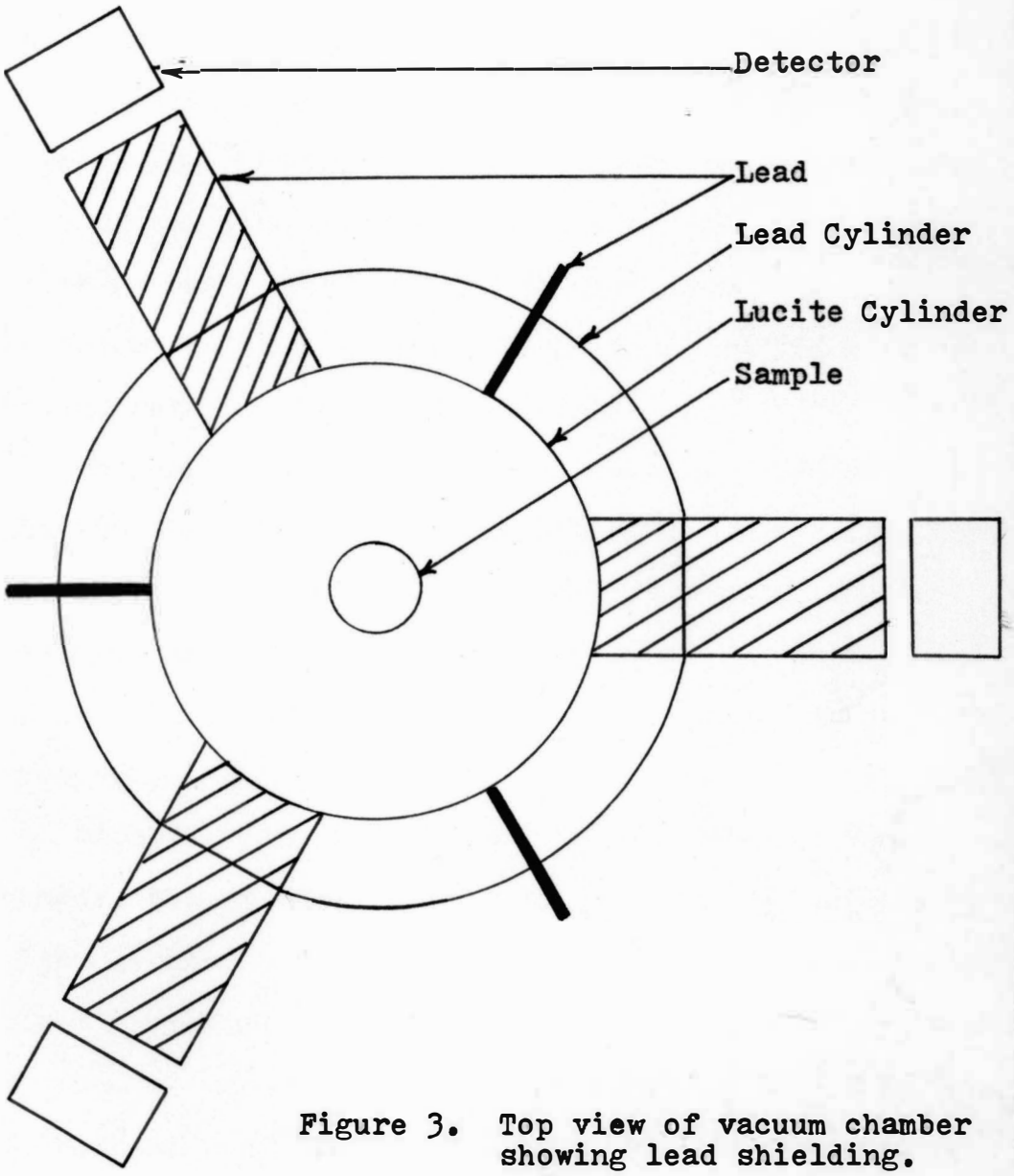


Figure 3. Top view of vacuum chamber showing lead shielding.

a thickness of three-eights inch, and a height of ten inches. This height was selected so that any 511 keV photons which Compton scattered in the aluminum top and headed towards a detector would not be in the three-photon energy range (270-400 keV). Since any positrons entering the Lucite might form positronium, an aluminum "shield" was placed inside the Lucite cylinder. Any positrons scattered towards the chamber walls, within the height of the detector, annihilated in the metal shield, rather than in the Lucite.

The one inch diameter metal samples were glued to a Lucite disk .950 inch in diameter, and about .030 inch thick. An eleven inch long, one-eighth inch diameter drill rod was attached to the center of the Lucite disk and held in place at the center of the aluminum top by a brass nut.

With the aluminum positron shield removed from the chamber, the sample was aligned in the plane defined by a line scribed on the outer surface of the Lucite at a height corresponding to the center of the detector faces. The chamber was evacuated with a forepump; the alignment of the sample checked, and if necessary, adjusted. When the sample had been properly aligned, the system was let up to atmospheric pressure, the aluminum positron shield put in place, and the chamber evacuated again. The two-

and three-photon coincidence rates were not measured until the pressure, as measured with a thermocouple gauge, stabilized : typically at about 50 microns.

To measure the three-photon annihilation rate, the three detectors were placed 120^{o} apart with each detector face 15.0 $\pm$ .1 cm. from the center of the sample. The two-photon rate was measured for each of the three possible pairs of detectors. Each pair of detectors -- one and two, two and three, one and three -- in turn, were placed on a common axis with the distance from each detector to the sample 15.0 $\pm$ .1 cm. Whenever a detector was moved from one position to another, the distance from it to the sample was adjusted so that the singles counting rate, and hence, the solid angle it subtended remained the same. For the three-photon case (Figure 3) additional lead shielding was again used to reduce the spurious coincidences caused by Compton scattering between detectors. The detector positions for the two- and three-photon cases are shown in Figures 4A and 4B. For clarity, only the detectors, lead cylinder, metal sample, Lucite cylinder, and aluminum top are shown.

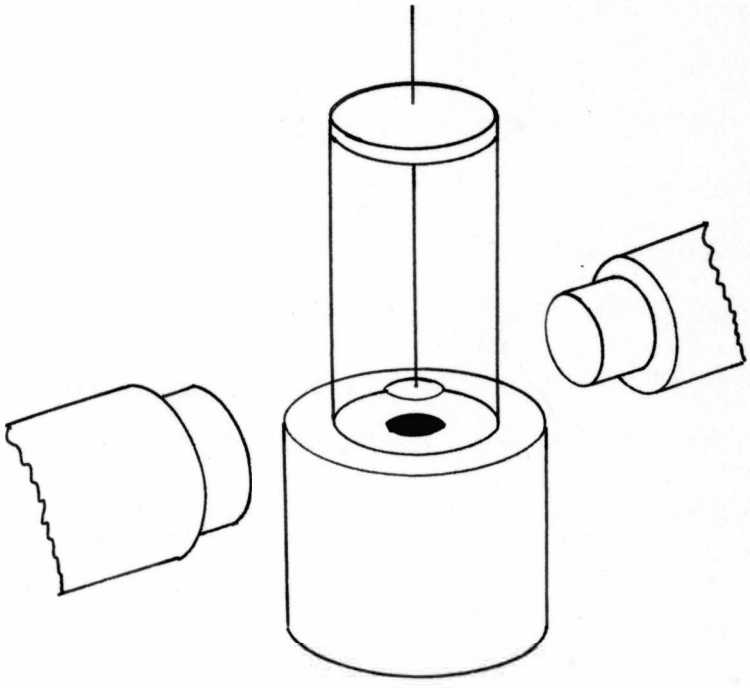


Figure 4A. Two-photon detector position.

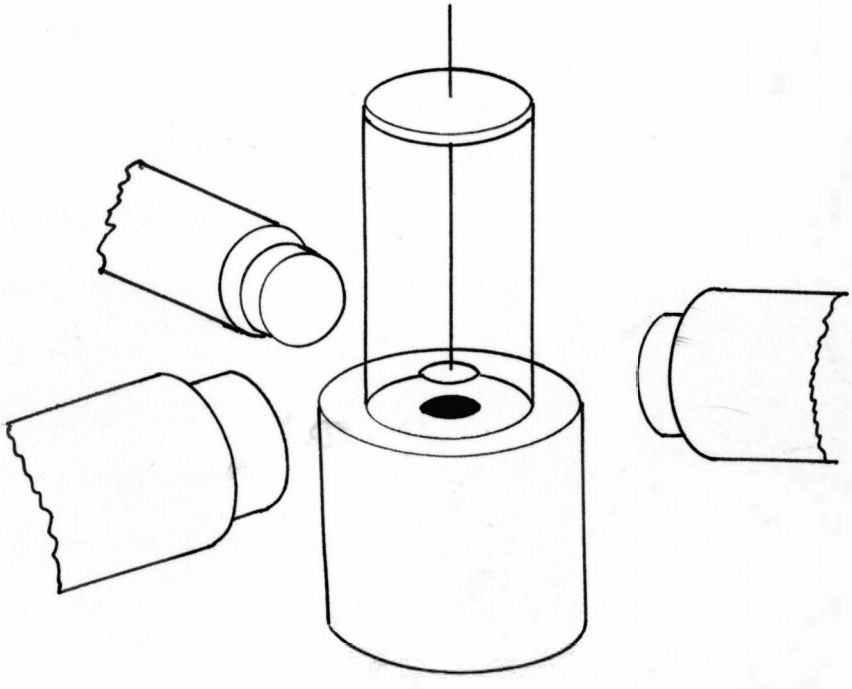


Figure 4B. Three-photon detector position.

SAMPLE PREPARATION

The sample thicknesses were chosen so that essentially all the .544 MeV positrons which entered each sample were stopped within it. The minimum thicknesses (200 mg/cm^2) for aluminum and silver are 0.027 and 0.0075 inch respectively. For convenience, sample thicknesses of 0.030 and 0.010 inch were selected. As in J. Siegel's experiment, high purity aluminum (99.99%) of this thickness was unavailable and the aluminum sample consisted of three 0.010 inch disks. Because the presence of an insulator between the layers of the sample might enhance the three-photon yield, the aluminum sample used in the present work was assembled without using any glue. One of the aluminum disks had three tabs approximately one-eighth inch by one-eighth inch that could be folded "up and over" the other two disks and thus hold the sample together. The silver sample consisted of one high purity (99.999%) disk, 0.010 inch thick.

To remove strains and extended defects which might cause appreciable deviation from the free electron case, the metal samples were annealed. The silver sample was first cleaned with distilled water and acetone, and then placed on a piece of Vylor (fused quartz) in an oven.

After the sample had been heated (in air) at 900° C for approximately five hours, the oven was turned off and allowed to cool to room temperature. The silver sample was then removed and glued to the sample holder with a minute amount of contact cement.

The heat treatment of the aluminum sample followed a similar procedure, except the three disks that made up the sample were first cleaned in carbon tetracloride before being heated at 600° C for five hours.

The surface of aluminum oxidizes very rapidly. Once, however, the initial oxide film is formed, the metal is protected from further attack.[17] Hunter and Fowle[18] measured the oxide films of a number of aluminum samples that had been stored in air at room temperature for periods varying from one week to several years. For each specimen they studied, a definite oxide film approximately 10 Å thick was observed. The thickness of the film that forms when aluminum is heated is both time and temperature dependent. After being heated at 600° C for several hours, the oxide film can be 2000 to 4000 Å thick.[19] The presence of such an oxide layer could, of course, enhance the three-photon yield. To remove the oxide layer that formed when the Al disks were annealed, they were etched for a few seconds in a 10% NaOH bath that was heated to 70° C, and then rinsed with a liberal

amount of distilled water. The aluminum sample was then assembled and glued to the sample holder such that the disk with the tabs was on the bottom, and the tabs themselves were on the top, next to the Lucite. As soon as the glue dried, the sample was placed under vacuum.

ELECTRONICS

The electronics used in the present work was identical to that used by J. Siegel. Harshaw Model 6S/4 Scintillation Detectors with a 1.5 by 1.0 inch NaI(Tl) crystal and an integrally mounted R.C.A. Photomultiplier Tube were used in conjunction with the electronics shown in Figure 5. The resolution of the detectors for the ^{137}Cs 662 keV peak was 7.8% or better.

A Nuclear Data Model ND 180 FM 512 Channel Analyzer and an Ortec Model 204 Precision Pulse Generator were used to set the single channel analyzer windows. The relationship between energy and channel number for the multichannel analyzer is linear and was calibrated using the following known sources and energies : ^{57}Co, 122 keV; ^{22}Na, 511 keV and ^{137}Cs, 662 keV. The channel numbers corresponding to the three-photon energy range (270-400 keV) were determined from the calibration curve; the pulser connected to the input of the linear amplifier and adjusted so that the pulse height corresponded to the desired channel number, and hence, energy. The single channel analyzer windows were then adjusted so that only pulses corresponding to events within the desired energy range could be counted. The two-photon windows were set a little above and below the 511 keV

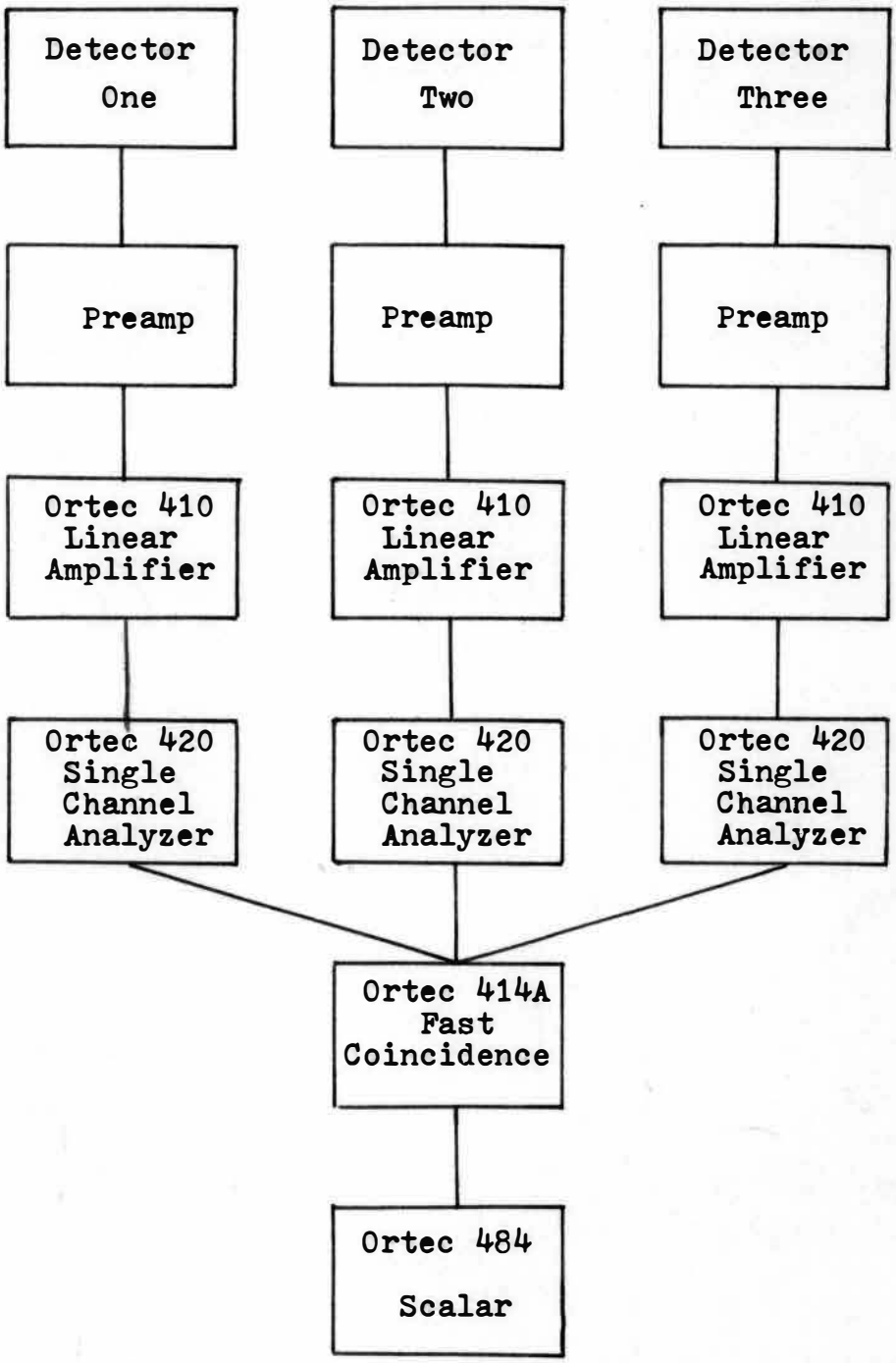


Figure 5. Schematic of the electronics.

annihilation photopeak. A spectrum from the silver sample with the two- and three-photon windows indicated is shown in Figure 6. During each data run, the gains on the linear amplifiers were checked every few hours, and adjusted (when necessary) so that the 511 keV peak remained in the same channel. The position of the peak rarely drifted more than two or three channels.

Since the coincidence circuit is triggered only when pulses arrive "at the same time," the time for the pulses to travel from the detectors to the coincidence circuit had to be equalized. The resolving time was set at the lowest setting and the single channel analyzer windows set to observe two-photon events. Each pair of detectors, in turn, was placed 180^{o} apart and the internal delay in the single channel analyzers systematically varied until the two-photon counting rate reached a maximum.

The coincidence circuit will accept two or more events as being simultaneous if the time interval (if any) between them is less than the resolving time of the circuit. To minimize the number of coincidences, which by chance, occur "at the same time," the resolving time should be as short as possible without rejecting any true coincidences. After the delays had been set, the resolving time was increased until the true two-photon counting

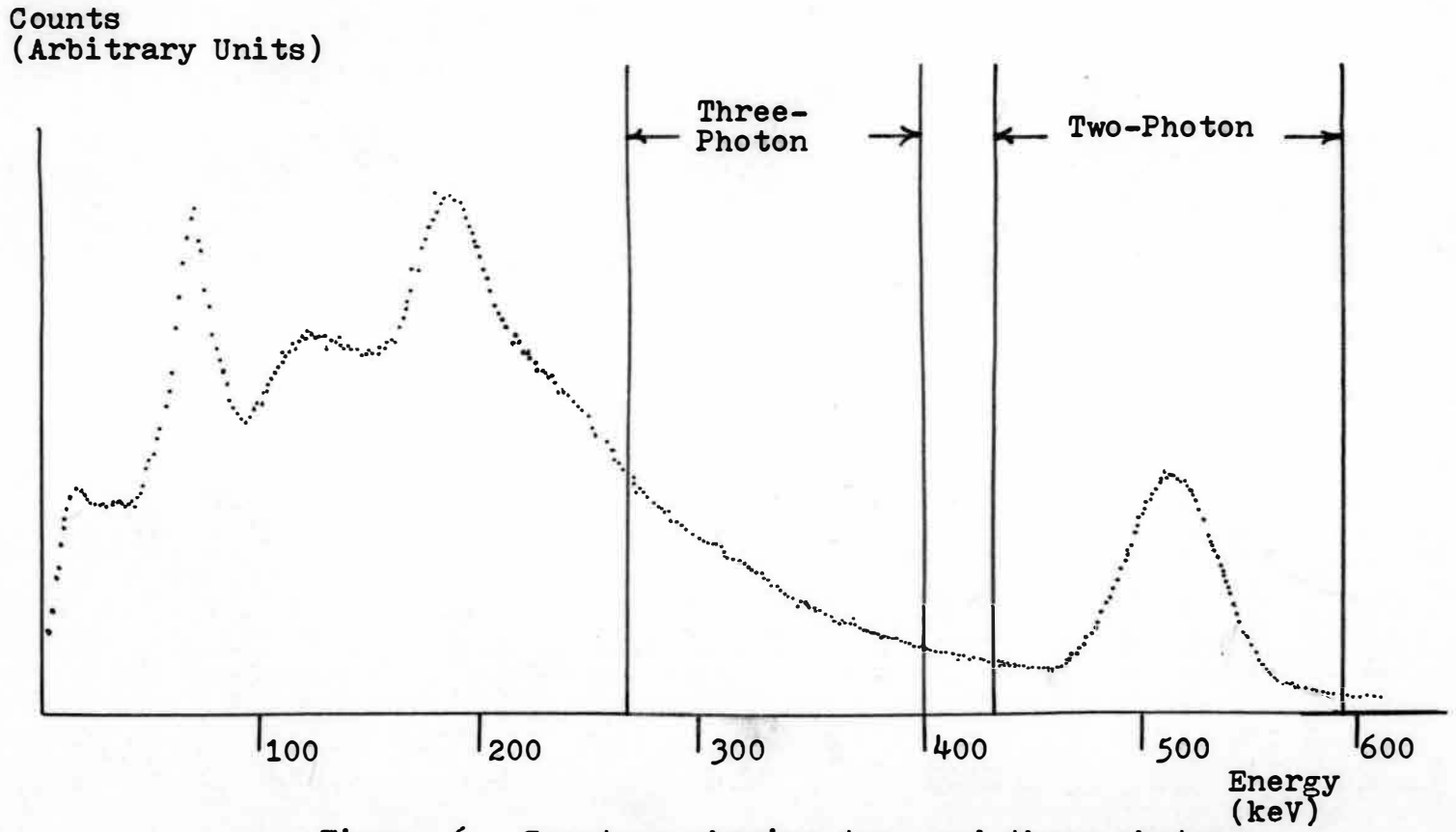


Figure 6. Spectrum showing two- and three-photon single channel analyzer window settings.

rate reached a maximum. The lowest possible dial setting to do this was "30 nsec", later found to correspond to an average resolving time of $2T = 40.62 \pm .50$ nsec. The settings for the delays, two- and three-photon single channel analyzer windows, and the resolving time were checked periodically throughout the course of the experiment with no significant variation observed.

DATA AND RESULTS

In the present work, as in J. Siegel's, the problem of determining σ_2/σ_3 for silver and aluminum may be divided into separate calculations and measurements :

(1) The calculation of the correction for the loss, due to scattering and absorption of 340 keV and 511 keV photons in each metal.

(2) The calculation of the solid angle factors.

(3) The measurement of the relative efficiency e(340)/e(511) and the absolute efficiency e(340) for each of the detectors.

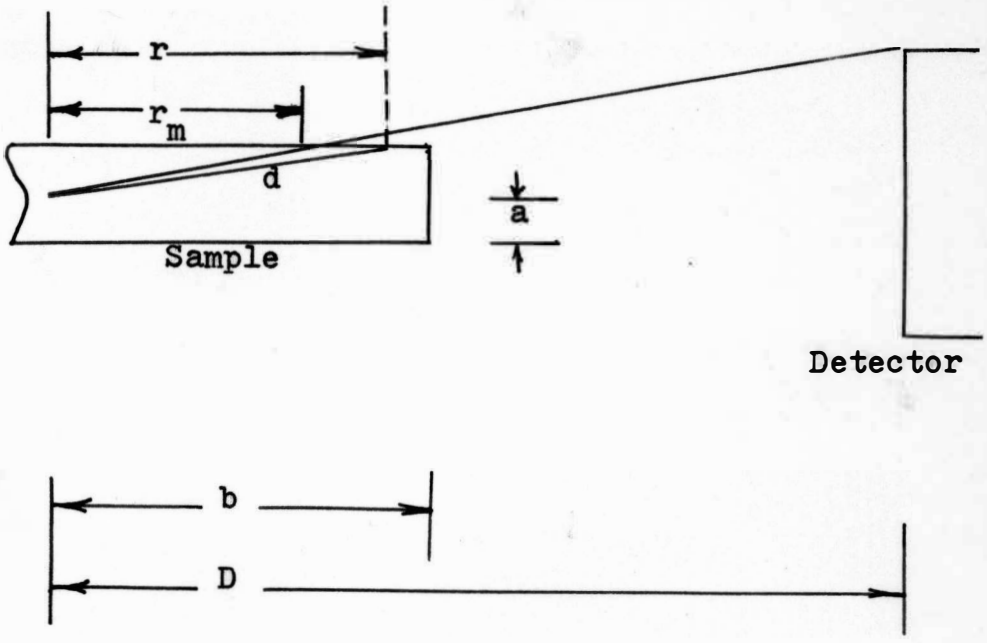
(4) The measurement of the two- and three-photon counting rates for each metal.

Correction for the Loss of Photons in the Sample

The photons that result from a positron annihilating in the sample must travel a considerable distance in the one inch diameter metal disks, and hence, some loss due to scattering and absorption will occur. The calculation of the probability of the loss of photons was made assuming, on the average, that the annihilation occurs at the center of the sample. First those photons which exit through the edge of the sample, headed for a detector, are considered; and then those which pass through the bottom or top of the sample headed for a detector are considered.

The notation used in the calculation is shown in Figure 7. The probability that a photon heads out of the sample in a solid angle subtended by an area on the edge, top, or bottom is the ratio of that solid angle to the total solid angle subtended by that edge, top, or bottom. The total solid angle subtended by the sample edge is :

$$\Lambda_e = \frac{2\pi\, bt}{b^2} = \frac{2\pi\, t}{b}$$



t = sample thickness

a = t/2

b = sample radius

D = distance from sample center to detector

R = radius of detector

$d = (a^2 + r^2)^{\frac{1}{2}}$

Figure 7. Notation for calculation of loss of photons in sample.

The solid angle subtended by an element of area in the sample top is :

$$d\Lambda_t = \left(\frac{a}{d}\right)\left(\frac{2\pi r\ dr}{d^2}\right) = \frac{2\pi ar\ dr}{(a^2 + r^2)^{3/2}}$$

The solid angle subtended by that area of the top through which the photon can strike the detector is :

$$d\Lambda_t = \int_{r_m}^{b} \frac{2\pi ra\ dr}{(a^2 + r^2)^{3/2}}$$

$$= 2\pi \left[\frac{1}{(1 + D^2/R^2)^{\frac{1}{2}}} - \frac{1}{(1 + b^2/a^2)^{\frac{1}{2}}}\right]$$

where $r_m = Da/R$ is the minimum value of r such that the photons can strike the detector.

The probability of the photon getting out of the sample is $e^{-\mu b}$ where μ is the linear attenuation coefficient for (photons of) the energy of interest and d is the distance the photon travels in the sample. The probability of a photon emerging through an edge is :

$$P_e = \frac{2\pi\, t/b\ e^{-\mu b}}{4\pi} = (t/2b)\ e^{-\mu b} = (a/b)\ e^{-\mu b}$$

The probability of a photon emerging through the top or bottom such that it can strike a detector is :

$$P_{tb} = 2\left(\frac{1}{2\lambda_t}\right)\int_{r_m}^{b}\left(\frac{2\pi r\ dr}{d^2}\right)\left(\frac{a}{d}\right)e^{-\mu d}$$

$$=\left(\frac{2\pi a}{\lambda_t}\right)\int_{r_m}^{b}\frac{r\ dr\ e^{-\mu\,(a^2+r^2)^{\frac{1}{2}}}}{(r^2+a^2)^{3/2}}$$

The average probability of a photon of energy E emerging from the sample is :

$$P(E) = P_{tb} + P_e$$

The values of the linear attenuation coefficient were obtained from Storm and Israel's tabulation.[20] Their value of the narrow beam linear attenuation coefficient was used because this is what is measured when both the source and detector are highly collimated. Since P(E) is an average probability, the correction factors for two- and three-photon annihilation are :

$$A_2 = P(511)^2 \quad ; \quad A_3 = P(340)^2$$

These correction factors for aluminum and silver are presented in table 1.

Table 1

Absorption and Scattering Correction

Metal	E	μ	P(E)
Al	340 kev	.268 cm^{-1}	.881
Al	511	.226	.903
Ag	340	1.47	.698
Ag	511	.983	.781

Metal	A(340)	A(511)	A(340)/A(511)
Al	.685	.815	.840 $\pm$ .029
Ag	.339	.609	.557 $\pm$ .036

Solid Angle Factors

Two solid angle factors are needed in the present work. To determine the absolute detector efficiencies, it is necessary to know the average solid angle subtended by one detector from a disk. In determining $\frac{\sigma_2}{\sigma_3}$, it is necessary to know the fraction of the two- and three-photon events which are intercepted by the detectors.

Ferrari[21] has calculated $\Delta\Lambda$, the average solid angle subtended by one detector for the case of two-photon coincidence detection. In the following discussion, his technique is applied to the one detector case. The coordinate system used by Ferrari is shown in Figure 8. The calculation for both the one and two detector cases was made assuming the annihilation or decay occurs in a most probable plane, namely Z = 0. The sample has radius R and is centered at the origin (0,0,0); the detector face has radius a and is centered at (0,d,0). The X' and Z' axes are parallel to the X and Z axes respectively and are a distance d from the origin. The annihilation or decay occurs at some point in the sample (x,y,0) and the photon or gamma ray enters the detector at some point (x',d,z'). Thus :

$$\underset{\sim}{r} = (x'-x)\,\hat{\underset{\sim}{i}} + (d-y)\,\hat{\underset{\sim}{j}} + (z')\,\hat{\underset{\sim}{k}}$$

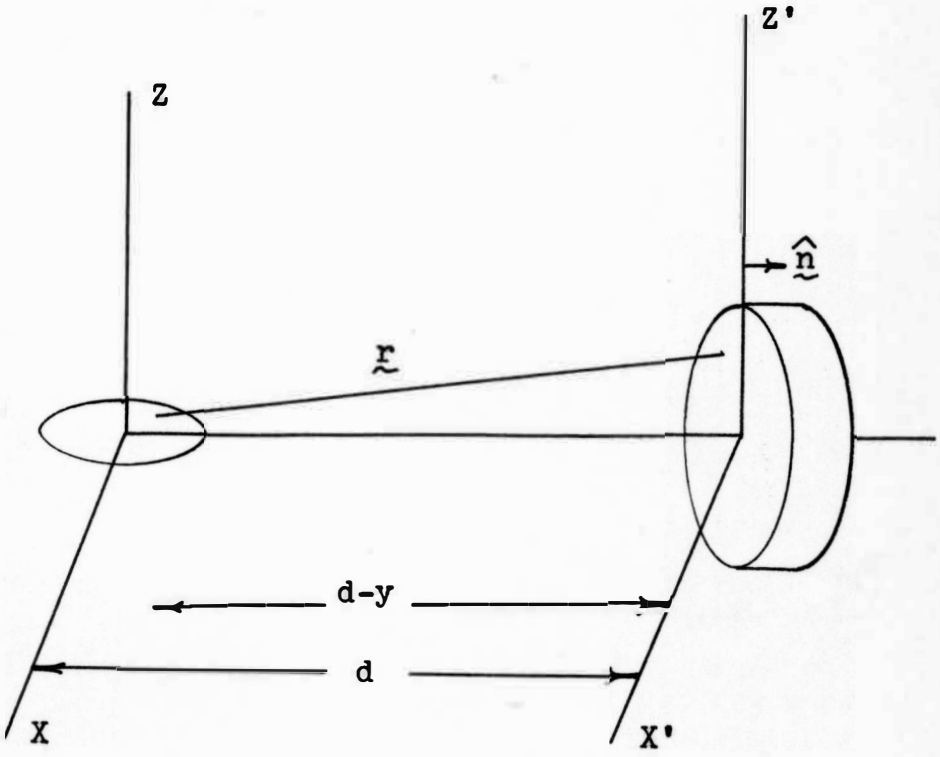


Figure 8. Notation for solid angle calculation.

The element of solid angle is :

$$d\Lambda = \frac{\underline{r} \cdot \underline{n}\, dS'}{r^3} = \frac{(d-y)\, dx'\, dz'}{\left[(x'-x)^2 + (d-y)^2 + z'^2\right]^{3/2}}$$

In the one detector case, the solid angle subtended by a point in the sample can be found by integrating d over the limits :

$$z' = \pm (a^2 + x'^2)^{\frac{1}{2}}$$
$$x' = \pm a$$

Taking the average over the entire disk gives the total solid angle :

$$\Lambda = \frac{1}{\pi^2 R} \iiiint \frac{(d-y)\, dx\, dy\, dx'\, dz'}{\left[(x'-x)^2 + (d-y)^2 + z'^2\right]^{3/2}}$$

where the additional limits of integration are :

$$x = \pm (R^2 + y^2)^{\frac{1}{2}}$$
$$y = \pm R$$

Except for a change in the limits, this integral is the same as one evaluated by Ferrari; its evaluation followed

his method and the same approximations were used. For a one inch diameter disk, and a detector face of 1.5 inch, 15 cm from the center of the disk :

$$\Delta\Lambda = .0502 \pm .0005 \text{ str.}$$

For a point source located at the origin :

$$\Delta\Lambda = \iint \frac{d \, dx' \, dz'}{x^2 + d^2 + z'^2 \quad 3/2}$$

where the limits of integration are :

$$z' = \pm (a^2 - x^2)$$
$$x' = \pm a$$

Evaluation of this integral by Simpson's rule gives :

$$\Delta\Lambda = .0505 \pm .0005 \text{ str.}$$

An approximate method, assuming $\underset{\sim}{r} \cdot \hat{\underset{\sim}{n}} = r$ gives for the one detector case :

$$\Delta\Lambda = \frac{a^2}{d^2} = .0507 \pm .0005 \text{ str.}$$

Ferrari's result for the present (extended source) geometry for the average solid angle subtended by one detector for the case of two-photon coincidence detection was :

$$\Delta\Lambda = .0321 \pm .0003 \text{ str.}$$

For a point source and the present geometry, the average solid angle subtended by one detector for the case of two-photon coincidence detection was :

$$\Delta\Lambda = .0507 \pm .0005 \text{ str.}$$

Thus, while there is essentially no difference in the value of $\Delta\Lambda$ for a point and extended source in the <u>one</u> detector case, there is a substantial difference in the <u>two</u> detector, coincidence detection, case.

The three-photon solid angle correction C_3 had not yet been determined for the present (extended source) geometry. Basson has, however, calculated C_3 for a point source. In terms of $\Delta\Lambda$, his result is :

$$C_3 = 21.3 \left(\Delta\Lambda / 4\pi \right)^{5/2}$$

It is possible to estimate C_3 for the present geometry if one assumes :

(1) The solid angle subtended by one detector is approximately the same for two- and three-photon detection.

(2) The functional form (but not the value) of Basson's calculation of C_3 applies to the present geometry.

Under these assumptions, the extended source estimate is :

$$C_3 = 21.3 \left(\frac{.0321}{4} \right)^{5/2} = 7.027 \times 10^{-6}$$

compared to a point source value of :

$$C_3 = 21.3 \left(\frac{.0507}{4} \right)^{5/2} = 22.00 \times 10^{-6}$$

Using the extended source estimate of C_3, the estimated solid angle factor needed to obtain σ_2/σ_3 is :

$$\frac{C_3}{C_2} = \left(\frac{21.3}{2} \right) \frac{(\Delta\Lambda/4\pi)^{5/2}}{(\Delta\Lambda/4\pi)} = 13.76 \times 10^{-4}$$

For the point source case, $C_3/C_2 = 27.29 \times 10^{-4}$.

Efficiencies

A convenient way to measure the relative efficiency e(340)/e(511) would be to measure the relative counting rates from a source which decays by emission of 340 and 511 keV photons of known relative intensity. Since such a source was unavailable, J. Siegel used ^{181}Hf which has a half life of about 45 days and which emits 482 and 346 keV gamma rays. J. Siegel placed the ^{181}Hf source -- a solution in a thin-walled, Lucite container shaped after the aluminum sample -- in the same position relative to the detectors as the sample, and then obtained spectra, less background, for each detector.

The number of counts under each peak in the spectrum is proportional to the source strength, the intensity of the gamma ray, the solid angle subtended by the detector, and the absolute efficiency of the detector at that energy. Accordingly, the ratio of the number of counts under the ^{181}Hf 346 and 482 keV peaks is proportional to the relative intensity of the two gamma rays and to e(346)/e(482).

J. Siegel determined the area under each peak in two different ways : first by simply summing the number of counts in each channel under the peak; and second by fitting a normal curve to the top third, top half, and

top two-thirds of each peak by the method of curvilinear regression. The values he obtained for the area under each peak, for the two methods, were in agreement within experimental uncertainty. Since the log of the photopeak efficiency e(E), when plotted against the log of energy E gives[22,23] a straight line with slope n, J. Siegel was able to obtain e(340)/e(511) by extrapolation from e(346)/e(482).

J. Siegel also used calibrated ^{137}Cs and ^{133}Ba sources to measure the absolute efficiencies e(662) and e(357) for each detector and then found e(340), e(511), and e(340)/e(511) by extrapolation. The sources, disks 2.37 cm in diameter, were placed in the same position relative to the detectors as the aluminum samples and a spectrum of each source, less background, obtained for each detector. As with the ^{181}Hf data, J. Siegel fitted a normal curve to each peak. While J. Siegel's results for the two methods were consistent within experimental error, the values obtained for e(340)/e(511) using ^{181}Hf were about three percent higher than those obtained using the ^{137}Cs and ^{133}Ba sources. There is a possibility that the ^{181}Hf source he used was contaminated with ^{175}Hf, an isotope which has a half life of about 70 days and which emits a 343 keV photon. The presence of such a contaminant could account for the discrepancy.

The ^{137}Cs and ^{133}Ba sources were used in the present work to measure the detector efficiencies. The principle activity of the ^{137}Cs and ^{133}Ba sources had been measured by Tracerlab to within one percent in March 1969 and had to be corrected for the decay which had occurred since then. The values of the half lives used in making the corrections were 29.901 ± 0.045 years for ^{137}Cs and 10.352 ± 0.040 years for ^{133}Ba.(24)

In the present work the efficiency of each detector was measured three times : about a year before, about a month before, and about a month after the two- and three-photon coincidence rates were measured. For the first two runs, the calibrated sources were placed on top of a small Lucite cylinder in the same position relative to the detectors as the samples. For the third run, the calibrated sources were taped to a sample holder and placed in the vacuum chamber with the aluminum positron shield in place, but with the pump turned off. The results of all three runs were consistent within experimental uncertainty (about two or three percent). The data for the third run were, however, slightly lower than the previous two due to absorption and scattering in the vacuum chamber walls and aluminum positron shield. Since the results from the third run (table 2) best represent the actual experimental conditions, they were used in the calculation of σ_2/σ_3 .

Table 2

Detector Efficiencies

	Detector One	Detector Two	Detector Three
Slope	-1.31 ± .04	-1.29 ± .05	-1.38 ± .04
e(662)	.130 ± .002	.137 ± .002	.123 ± .002
e(357)	.291 ± .007	.304 ± .008	.288 ± .006
e(511)	.182 ± .004	.191 ± .004	.176 ± .004
e(340)	.311 ± .007	.324 ± .008	.308 ± .007
$\frac{e(340)}{e(511)}$	1.70 ± .03	1.69 ± .03	1.76 ± .03

Two- and Three-Photon Data

Two-photon coincidence rates

The silver sample was aligned three times and the aluminum sample twice. For each sample position, both the two- and three-photon coincidence rates were measured. The average two-photon "singles" rates for each of the detectors are presented in table 3 and the total (chance plus true) coincidence rates in table 4.[1] The time for each of the singles and two-photon coincidence was at least 100 seconds and was measured with an electric lab timer.

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[1]The data in these tables and in tables 5 and 6 have been corrected for the decay of ^{22}Na which occurred over the course of the experiment.

Table 3

Two-Photon Singles Rates
(Counts/Sec.)

Sample	Run	Detector One	Detector Two	Detector Three
Ag	1	3999 ± 75	4285 ± 95	3977 ± 108
Ag	2	3861 ± 40	4183 ± 40	3954 ± 42
Ag	3	3866 ± 26	4205 ± 37	3954 ± 31
Ag	Avg	3909 ± 78	4224 ± 54	3962 ± 40
Al	1	4052 ± 47	4487 ± 199	4104 ± 63
Al	2	4156 ± 27	4470 ± 21	4263 ± 42
Al	Avg	4104 ± 52	4479 ± 100	4170 ± 132

Table 4

Total Two-Photon Counting Rates
(Counts/Sec.)

Sample	Run	Detectors One and Two	Detectors Two and Tnree	Detectors One and Tnree
Ag	1	80.4 ± 2.7	82.0 ± 1.4	79.1 ± 2.5
Ag	2	79.0 ± 2.0	79.3 ± 1.8	77.7 ± 0.7
Ag	3	81.1 ± 0.8	81.8 ± 0.9	80.0 ± 1.2
Al	1	150.7 ± 1.9	152.4 ± 9.1	143.4 ± 1.4
Al	2	153.2 ± 3.1	148.9 ± 1.9	146.5 ± 1.1

Two-photon stray rate

In the type of coincidence circuit used in the present work, each input can, according to Chase[25], be thought of as occupying a time interval equal to the average resolving time T of the circuit. When the time intervals associated with the input signals overlap, a coincidence is recorded. The measured coincidence rate, thus, includes both true and chance coincidences, ie. :

$$N_{total} = N_{true} + N_{chance}$$

For two inputs, A and B, which receive signals at respective rates Na and Nb, the coincidence circuit will accept as simultaneous all signals from input A which are separated from a B signal by a time interval less than the average resolving time of the circuit. For a chance coincidence to be recorded, the A signal must occur within a time interval 2T which symmetrically brackets the B signal. If Nt is the true coincidence rate, the rate for pulses not associated with a true coincidence is (Na - Nt) for channel A and (Nb - Nt) for channel B. The fraction of the total data collection time during which a chance coincidence may occur is (Na - Nt) (2T). Multiplying this by (Nb - Nt) gives the chance

coincidence rate :

$$N_{calc} = (Na - Nt)\ (Nb - Nt)\ (2T)$$

If Nt is much smaller than Na and Nb, the chance rate is to a good approximation :

$$N_{calc} = Na\ Nb\ (2T)$$

One way to determine the true two-photon rate, therefore, would be to measure the singles rates Na and Nb and then use :

$$N_{calc} = (Na - Nt)\ (Nb - Nt)\ (2T)$$

Since in the present experiment, the two 511 keV photons emerge 180^{o} apart, a second way to determine the chance coincidence rate would be to measure the coincidence rate with the detectors at an angle where they cannot detect the two photons in coincidence. In the present work, this was done with the detectors 120^{o} apart. A third way to measure the two-photon stray rate would be to leave the detectors 180^{o} apart and insert delays so that pulses from a true annihilation cannot reach the coincidence circuit simultaneously. Typical results for the two-photon chance rate are given in table 5. The electric

Table 5

Two-Photon Stray Rate

(Typical Data, Counts/Sec.)

Metal	Detector Pair	N_{120^o}	N_{elect}	N_{calc}
Ag	12	$.87 \pm .03$	$.66 \pm .03$	$.71 \pm .02$
Ag	23	$.82 \pm .03$	$.66 \pm .03$	$.64 \pm .01$
Ag	13	$.81 \pm .03$	$.61 \pm .03$	$.64 \pm .01$
Al	12	$.86 \pm .02$	$.59 \pm .02$	$.65 \pm .02$
Al	23	$.78 \pm .02$	$.66 \pm .02$	$.71 \pm .02$
Al	13	$.76 \pm .02$	$.71 \pm .02$	$.66 \pm .02$

delay rates (N_{elect}) and the calculated rate (N_{calc}) are in agreement, but N_{120^o} is about 25% higher than the other two. There are at least two possible explanations of this discrepancy. A 1.27 MeV gamma ray could have "tunneled through" the lead shielding and then Compton scattered in the NaI(Tl) crystal. An annihilation may have occurred in the aluminum positron shield. In any event, the discrepancy is small compared to the uncertainty in the total two-photon counting rates.

The calculated stray rate (N_{calc}) was used to determine the true two-photon coincidence rates which are presented in table 6.

Table 6

True Two-Photon Counting Rates
(Counts/Sec.)

Sample	Run	Detectors One and Two	Detectors Two and Three	Detectors One and Three
Ag	1	80.7 ± 2.7	81.4 ± 1.4	78.5 ± 2.5
Ag	2	78.4 ± 2.0	78.7 ± 1.8	77.1 ± 0.7
Ag	3	80.5 ± 0.8	81.2 ± 0.9	79.4 ± 1.2
Ag	Avg	79.9 ± 2.3	80.2 ± 1.8	78.2 ± 1.9
Al	1	151.7 ± 9.1	150.1 ± 1.9	142.8 ± 1.4
Al	2	148.2 ± 1.9	152.6 ± 3.1	145.9 ± 1.1
Al	Avg	150.0 ± 4.6	151.4 ± 1.8	144.4 ± 1.4

Consistency of two-photon data

As in J. Siegel's experiment, the singles counting rate, the true two-photon coincidence rates, and the absolute detector efficiencies at 511 keV can be used to check the consistency of the two-photon data. The true two-photon coincidence rate for any pair of detectors is proportional to the efficiency of each detector at 511 keV. Thus, the ratio N_{ij}/N_{ik} should equal $e_j(511)/e_k(511)$. Since the singles rate is proportional to the efficiency of the detector, the ratio of the singles rates from two different detectors should also equal the ratio of their efficiencies. The ratio of the singles rates, coincidence rates, and the absolute efficiencies are presented in table 7.

Table 7

Detector Ratios

Ratio	One/Two	Two/Three	One/Three
From Efficiencies	.95 ± .02	1.09 ± .03	1.04 ± .03
From Two-Photon Rates			
Aluminum	.95 ± .02	1.04 ± .03	.99 ± .03
Silver	.97 ± .03	1.02 ± .04	.99 ± .03
From Singles Rates			
Aluminum	.92 ± .02	1.07 ± .03	.98 ± .02
Silver	.93 ± .03	1.07 ± .03	.99 ± .03

Comparison of silver and aluminum two-photon data

The difference between the two-photon rates for silver and aluminum is attributable to the absorption and scattering of the photons in the samples and to the backscatter of the incident positrons. According to Bisi and Braicovich,[26] the backscattering coefficient as a function of atomic number Z is :

$$P_t(Z) = (0.0593 \pm 0.0019)\, Z^{\frac{1}{2}}$$

For the silver sample the fraction of the positrons that backscatter is .4065 $\pm$ .0130 ; for aluminum the fraction backscattering is .2162 $\pm$ .0069 . Combining the backscattering coefficients with the correction for the loss of photons in each sample, one might expect 1.77 $\pm$ .08 times as many two-photon events to be observed in aluminum for each one in silver. The actual values obtained were :

Detector Pair	$\frac{\text{Two-photon Events in Al}}{\text{Two-photon Events in Ag}}$
One and Two	1.88 $\pm$ .06
Two and Three	1.88 $\pm$ .03
One and Three	1.84 $\pm$ .02

The agreement of the predicted and observed ratios indicates that the approximations made in calculating the correction for absorption and scattering of the positrons in the samples are reasonable.

Three-photon coincidence rates

Several three-photon counting runs were taken for each metal. Since the counting rates were small, long observation times were required. The observation times were limited by the possibility of drift in the single channel analyzer windows and delays. A power failure terminated one run (with the silver sample) prematurely.

Every few hours, during the three-photon run, the gains on the linear amplifiers were checked, and the total number of counts and total elapsed time recorded. The elapsed time was measured by an electric clock and checked against the "dial the time service" provided by the local telephone company. The length of time for the runs varied from about ten to ninety-three hours. The uncertainty in measuring the time is negligible compared to the other uncertainties.

The time for each run, the total number of counts, and the counting rate per hour are reported in table 8. The error given in the counting rate was determined from $N_i^{\frac{1}{2}}$ where N_i is the number of counts for each run. The results of a weighted least squares fit to $Y = MX$ are also given, but since the model used in making the fit is invalid for this type of data, it can only be used to check the consistency of the counting rate for each run.

Table 8

Three-Photon Coincidence Runs

Sample Position	Run	Day	Time Hours T	Counts N	Rate/Hour N/T	Rate/Hour Least Squares
Silver						
1	1	2	10:10	18	1.77 ± .41	---------
1	2	3	43:37	83	1.90 ± .21	2.01 ± .02
1	3	7	58:00	122	2.10 ± .19	2.10 ± .02
1	4	14	45:26	84	1.85 ± .20	1.91 ± .04
2	1	27	61:00	133	2.18 ± .19	2.26 ± .03
2	2	40	66:00	131	1.98 ± .17	2.26 ± .02
3	1	45	40:00	67	1.68 ± .20	1.61 ± .02
3	2	47	66:00	125	1.89 ± .17	1.93 ± .01
3	3	50	92:00	180	1.96 ± .15	2.00 ± .02
Aluminum						
1	1	66	84:00	235	2.80 ± .18	2.80 ± .01
1	2	69	90:00	244	2.71 ± .17	2.82 ± .02
2	1	75	90:00	255	2.83 ± .18	2.66 ± .02
2	2	77	16:30	42	2.55 ± .39	---------
2	3	80	93:00	269	2.88 ± .18	2.78 ± .01

Three-photon stray rate

In a three-fold coincidence experiment, four types of events can result in a chance coincidence :

(1) All three detectors observe random photons.

(2) Detector one sees a random photon, detectors two and three see photons from a true coincidence.

(3) Detector two sees a random event, detectors one and three see a true event.

(4) Detector three sees a random event, detectors one and two a true event.

For (case one) three uncorrelated input signals 1, 2, and 3 with respective singles rates N_1 , N_2 and N_3 the two-fold chance coincidence rate for inputs 1 and 2 is $N_1 N_2 (2T)$. The "overlap" time interval for these coincidences has some length between 0 and T. Since the signals are uncorrelated, all overlap times are equally likely. On the average, the overlap interval is T/2 . If a signal from input 3 is received during such an overlap interval, or precedes such an interval by a time less than T, a three-fold chance coincidence will occur. Thus, for a period T + T/2 = 3/2 T a random signal from detector 3 can cause a three-fold chance coincidence.

The fraction of the data collection time during which this may occur is :

$$N_1 N_2 (2T) (3/2) T = 3 N_1 N_2 T^2$$

Multiplying this fraction by N_3 gives the three-fold coincidence rate due to random pulses in all three inputs :

$$R_1 = 3 N_1 N_2 N_3 T^2$$

If N_{23} is the true two-fold coincidence rate between inputs 2 and 3, and if the true three-fold coincidence rate is small, the chance rate due to a true coincidence in inputs 2 and 3 and a random pulse in input 1 is (case 2) :

$$R_2 = N_{23} N_1 (2T)$$

The rates for the other two cases are :

$$R_3 = N_{13} N_2 (2T)$$

$$R_4 = N_{12} N_3 (2T)$$

Thus, the chance rate is :

$$N_{calc} = R_1 + R_2 + R_3 + R_4$$
$$= 3\, N_1 N_2 N_3 T^2 + (N_{12}N_3 + N_{13}N_2 + N_{23}N_1)(2T)$$

As in the two-photon case, the three-photon stray rate can be measured by inserting extra delays. The various possibilities are presented in table 9. If, for example, a delay is inserted in channel one, true coincidences between all three detectors and between detector one and either of the other two detectors are eliminated. Random coincidences between all three detectors (case one) and also random coincidences in which (case two) detectors two and three see a true event would, however, still be observed. If two different delays are inserted in two channels, only three-fold random coincidences can be observed. If $N_{1,2,3}$ is the observed coincidence rate with delays inserted in two channels and if $N_{1,23}$, $N_{2,13}$ and $N_{3,12}$ are the coincidence rates when delays are inserted in channels one, two, and three respectively, the stray rate will be given by :

$$N_{stray} = N_{1,23} + N_{2,13} + N_{3,12} - 2\,(N_{1,2,3})$$

Table 9

Events Observed With Various Delays Inserted

	No Delay	One Delay In :			Two Delays
		Detector One	Detector Two	Detector Three	
Event :	N_{total}	$N_{1,23}$	$N_{2,13}$	$N_{3,12}$	$N_{1,2,3}$
All 3 Random	Yes	Yes	Yes	Yes	Yes
All 3 True	Yes	No	No	No	No
12 True 3 Random	Yes	No	No	Yes	No
23 True 1 Random	Yes	Yes	No	No	No
13 True 2 Random	Yes	No	Yes	No	No

The electronic delay method was used by J. Siegel to determine the three-photon stray rate, but is impractical when the coincidence rates are small. In the present work, the three-photon stray rate was determined from N_{calc} ; and the electric delay method used only as a check.

A second check is to measure the three-fold coincidence rate with all three detectors in the same half plane where no three-photon events can be observed. To do this, two of the detectors were placed 120^{o} apart and the third halfway between these with sufficient lead shielding to eliminate spurious coincidences due to Compton scattering. The singles rates for each detector were about the same as when the three-photon coincidence rates were measured. The true two-fold coincidence rates for the detectors 120^{o} apart were also the same as normal; but for the detectors 60^{o} apart, the true two-fold coincidence rate was somewhat smaller than normal. The calculated three-fold coincidence rate for the half plane position was $.98 \pm .06$ counts/hour compared to a measured rate of $.96 \pm .10$ counts/hour.

To determine N_{calc}, the singles rates N_1 , N_2 and N_3 for detectors one, two, and three; and the total and delayed two-fold coincidence rates for detector pairs one and two, two and three, and one and three were measured.

The true two-fold coincidence rate for each detector pair was determined from :

$$N_{ij} = N_{ij}^{t} - N_{ij}^{d}$$

where N_{ij}^{t} is the total two-fold coincidence rate and N_{ij}^{d} is the delayed two-fold coincidence rate. The stray rate was found from :

$$N_{stray} = 3\, N_1 N_2 N_3 + (N_{12}N_3 + N_{13}N_2 + N_{23}N_1)(2T)$$

The resolving time of the coincidence circuit was determined for each pair of detectors from :

$$2T = N_{ij}^{d} / (N_i N_j)$$

for each set of measurements of N_{ij}^{d} , N_i and N_j. The average values for each detector pair were :

Detector Pair	Resolving Time, 2T
One and Two	42.16 $\pm$ 1.06 nsec
Two and Three	38.58 $\pm$ .66
One and Three	41.14 $\pm$ .82

The average resolving time for all three pairs of detectors, $2T = 40.62 \pm .50$ nsec, was used in calculating the three-photon stray rate. The average value for each detector pair was used in determining the two-photon stray rate for that detector pair.

Each set of singles and true two-fold coincidence data was corrected for the ^{22}Na which had occurred since the first data had been taken; their average values found and used to determine N^i_{stray}, the stray rate for each three-photon run. The true three-photon rates (table 11) were determined from :

$$N_3 = \sum_i (T_i/T)(N^i_{total} - N^i_{stray})/T_i \; e^{-(D_i/365)(.693/2.60)}$$

where :

T_i is the observation time for run "i"

T is the total observation time

N^i_{total} is the total number of counts observed for run "i"

D_i is the number of days since the first data was taken.

Table 10

Three-Photon Stray Rate Data
(Counts/Sec.)

Event	Silver	Aluminum
N_1	5050.4 ± 86.6	5074.3 ± 76.1
N_2	5459.1 ± 73.2	5635.5 ± 21.8
N_3	5033.4 ± 53.0	5024.8 ± 47.4
N_{12}	.345 ± .039	.364 ± .057
N_{13}	.351 ± .057	.422 ± .029
N_{23}	.351 ± .059	.393 ± .037
N_{calc}*	1.411 ± .080	1.542 ± .044

*Counts/Hour

Table 11

Three-Photon True Rate

Metal	N_{true} (Counts/Hour)
Silver	.621 ± .098
Aluminum	1.513 ± .092

COMPARISON OF $\frac{\sigma_2}{\sigma_3}$ FOR AL AND AG

The ratio of the reaction cross section ratios for two- and three-photon positron annihilation in silver and aluminum is given by :

$$R = \frac{\left(\sigma_2/\sigma_3\right)_{Al}}{\left(\sigma_2/\sigma_3\right)_{Ag}}$$

Since :

$$\frac{\sigma_2}{\sigma_3} = \frac{N_2^{ij}\ C_3\ A_3}{N_3\ C_2\ A_2}\ e_i(340/511)\ e_j(340/511)\ e_k(340)$$

R_{ij} for each of the three pairs of detectors is :

$$R_{ij} = \frac{(N_2^{ij}/N_3)_{Al}\ (A_3/A_2)_{Al}}{(N_2^{ij}/N_3)_{Ag}\ (A_3/A_2)_{Ag}}$$

The results for each of the three detector pairs were :

Detector Pair	R_{ij}
One and Two	1.16 ± 0.22
One and Three	1.14 ± 0.21
Two and Three	1.17 ± 0.22

The average value of R_{ij} , 1.16 ± 0.15 , is consistent

with both a value of 1.0 and with Bertolaccini et al's value of 1.13 ± 0.02 .

The estimated value of C_3/C_2 would give the following values of $\frac{\sigma_2}{\sigma_3}$ in aluminum and silver :

Detector Pair	Aluminum	Silver
One and Two	365 ± 30	314 ± 55
Two and Three	385 ± 33	330 ± 58
One and Three	385 ± 30	337 ± 59
Average	378 ± 18	327 ± 33

The uncertainties in the estimates of $\frac{\sigma_2}{\sigma_3}$ include all uncertainties except that in C_3 . The results for aluminum appear to be in good agreement with Ore and Powell's theoretical value of 370 ; the results for silver are somewhat lower. If the point source estimate of C_3 had been used, the estimates of $\frac{\sigma_2}{\sigma_3}$ would have about twice as large. Clearly, further comparison of the experimental values of $\frac{\sigma_2}{\sigma_3}$ with theory must await a better calculation of C_3 .

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