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THE THREE-PHOTON ANNIHILATION OF POSITRONS AND ELECTRONS

by

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I. ABSTRACT

The three-photon annihilation of positrons and electrons has been investigated experimentally by means of a triple coincidence technique. Three NaI(Tl) scintillation counters in coincidence were used to study the occurrence of the triplet annihilation process when positrons annihilate against the electrons within solids, liquids, and liquefied gases. The annihilation rates in aluminum, liquid nitrogen, and liquid Freon-12 (CCl_2F_2) were compared with the theoretical predictions made under the assumption that free positron-electron pairs produced all annihilation events; experiment and theory agreed within the experimental error. The formation of stable ortho-positronium in Freon-12, SF_6, N_2, H_2, and He was verified and its comparative stability in these gases measured. The spectra of the annihilation rays from ortho-positronium were measured to demonstrate that energy and momentum are conserved in the three-photon annihilation process.

II Introduction and History of the Problem

Among the first to consider the possibility that an appreciable number of positron-electron pairs might annihilate with the emission of three photons instead of the usual two were Lifshitz[1] and Ivanenko[2] in Russia and Ore and Powell[3] in this country. These authors all used time-dependent perturbation theory to calculate the probability per unit time for the three-quantum annihilation of a free positron-electron pair with small relative velocity. They neglected the effects of Coulomb binding upon this probability, and assumed plane wave functions for the electron in its initial (positive energy) and final (negative energy) states. In spite of the similarity of approach to the problem, all three calculations yielded different numerical results. Ore and Powell's value for the relative frequency of occurrence for three-photon and two-photon annihilation was $\frac{\sigma_{3\gamma}}{\sigma_{2\gamma}} \approx \frac{1}{370}$, and this value has since been obtained theoretically also by Radcliffe and by R. Drisko at this institution[4],[5]. Lifshitz and Ivanenko found results in considerable disagreement with the above ratio. (Ore and Powell's value for the lifetime of positronium in the ^3S state has also been verified experimentally and will be discussed later.) All these authors calculated the cross-sections to the lowest non-zero order for the process, corresponding to the type of Feynman diagram shown in Figure 1(a), with all possible permutations of k_1, k_2, k_3 included in the sum over intermediate states. Another result of the various theoretical calculations is that the triplet state (spins parallel) do not contribute to the two-quantum annihilation, a conclusion consistent with selection rules derived by Yang[6] and Landau[7]. Further, the singlet state of the electron-positron system does not contribute to the matrix elements for the three-quantum process. Wolfenstein and Ravenhall[8] have shown that this latter statement will be true to all orders of calculation if the principle of invariance under charge conjugation is accepted.

Figure 1(b) shows the energy spectrum for one photon from three-quantum annihilation obtained by Ore and Powell, with its peak at $E = mc^2$. It is apparent from momentum conservation that no one of the three photons can carry off more than mc^2 of the total $2mc^2$ available; in this case the other two photons may share the other mc^2, their directions being at 180° with the direction of the first photon. The sharp peak in the otherwise almost linear energy spectrum indicates that this arrangement of the three gamma-rays has a relatively large probability of occurrence. Unfortunately it is experimentally most difficult to detect, because the two-quantum annihilation background will be at a maximum.

Although at the time work on the experimental detection of the effect was started here the three-photon annihilation had not been found experimentally, the preliminary work of J. A. Rich[9] showed that there might be a measurable triplet annihilation rate when positrons were stopped in solids. He used an apparatus similar in principle to that to be described, and found an effect consistent with any of the cross-section calculations discussed above. Therefore it seemed to be of interest to check more closely the theoretical value of $\frac{\sigma_{3\gamma}}{\sigma_{2\gamma}}$, as well as to study the shape of the energy spectrum, which is equivalent to an angular distribution for the three photons.

There is an important aspect of positron-electron interaction which we have not yet considered. If a positron and electron meet in free space, one would expect that there would be definite energy levels which the electron could assume with respect to the positron, thus forming a bound system similar to the hydrogen atom. Wheeler(10) was among the first to speculate on the existence of such a system, as well as those consisting of two positrons and one electron, etc. He called these systems "polyelectrons," using the symbol P^{+-} for the electron-positron system which is now commonly called positronium, as suggested by Ruark. Wheeler proposed that the existence of the positronium atom might be verified by observation of its emission of a characteristic optical spectrum. This spectrum would be similar to that of hydrogen except for a displacement towards the infra-red by a factor of two in wavelength. This follows from the fact that the reduced mass of P^{+-} is $\sim \frac{m_e}{2}$, while for the hydrogen atom it is $\sim m_e$. Thus the binding energy of the positronium atom is 6.77 volts and its energy-level spacings are one-half those in hydrogen. Wheeler also calculated the binding energy of P^{++-} as 6.96 volts, so that this system is stable by only .19 volts against disintegration into a positron and a positronium atom.

The optical spectrum of positronium has not yet been observed, but the existence of the atom in its ^3S-state has been detected by measurement of the characteristic lifetime of this state and its emission of the three-photon gamma spectrum upon annihilation(11)(12). (As noted above, the triplet states must annihilate with the emission of three photons, the singlet state with two.) If a positronium atom is in an excited state, annihilation is rather improbable because of the small value of the electron wave-function at the position of the positron. Thus, even though positronium in an excited state may be formed when positrons are stopped in a gas under standard condition, one would expect that de-excitation by collision or radiative emission would bring the atom to an S-state before annihilation could occur. The lifetime against annihilation for the ^3S-state is 1.4×10^{-7} seconds, and for the

^1S-state it is 1.25×10^{-10} seconds. There is an energy difference of 8.5×10^{-4} ev between the two states; the ^3S is metastable with respect to radiative transitions to the ^1S level. One therefore expects the ^3S-state to contribute three-fourths of all annihilation events from positronium, and the three-photon spectrum should be clearly visible even if only a small fraction of the positrons stopping in the gas form positronium. Deutsch(11) has in fact detected the existence of ortho-positronium (ortho = triplet, para = singlet) by observing its three-photon spectrum with a scintillation counter. He has also measured its lifetime against annihilation by a delayed coincidence method(12), and found close agreement with the value given by Ore and Powell.

He observed that about ten per cent of all positrons stopping in many gases (including N_2, He, A, CO_2, and CCl_2F_2) form positronium and that certain other gases, notably O_2 and NO, act strongly to destroy the ortho-positronium before it can annihilate. This quenching is attributed to spin exchange between the electron in positronium and a loosely bound electron in the NO (or O_2) molecule, an exchange converting the triplet state to the singlet state, which will annihilate rapidly by two-quantum annihilation. It was this quenching effect which enabled Deutsch to separate the three-photon spectrum from the background of two-photon gammas. He measured the "flipping" (quenching) cross-section of an NO molecule for an incident positronium atom, and found it to be 7.6×10^{-17} cm^2 (13), a rather large value compared to the geometrical cross-section for the molecules of 10^{-15} cm^2 or less. The corresponding cross-section for O_2 was somewhat smaller, about 4×10^{-19} cm^2. The stability of ortho-positronium in other gases indicates much smaller cross-sections than those for NO and O_2.

If the positrons stop in a solid instead of in a gas, it is doubtful whether positronium is formed at all. For the smallest Bohr radius of a positronium atom is 1.05 Å, while the lattice spacing in solid crystals is a few Angstroms or less. Therefore a positronium atom moving through a solid will be strongly perturbed by ionic and electronic fields, so that the electron would rapidly change its spin relative to the positron. We might even conclude that a positronium "atom" cannot really exist within a solid, and that it cannot fit into the lattice structure. In a solid a positron probably moves throughthe crystal lattice until it annihilates against one of the electrons without ever having been very strongly bound to any one of them. If we consider the annihilating particles to be entirely free, then the ratio of 370 mentined above should prevail in favor of two-photon annihilation over three photons. Thus a measurement of the triplet annihilation rate should serve as a check on the theoretical value of $\frac{\sigma_{3\gamma}}{\sigma_{2\gamma}}$.

III. Experimental Procedure

A. Coincidence Measurements of Annihilation Phenomena

The double coincidence method of studying two-quantum annihilation is well known and widely used. Two gamma-ray counters are placed on opposite sides and collinear with a source of annihilation events, and the rate of coincidences between the two counters is a measure of the annihilation rate of positrons in the material surrounding the source. If a thermal positron annihilates with an electron in an atom or with an electron drifting through a solid lattice, the total momentum is very nearly zero compared to that associated with one of the annihilation gamma-rays. Then energy-momentum conservation determines that the directions of the two gammas lie at nearly 180° with each other(14). This strong angular correlation aids study of two-photon annihilation by making the coincidence rate effectively independent of the solid angle subtended by one of the two counters, because one of the counters serves to completely define the beam of annihilation rays.

However, the extension of the coincidence method to three-photon annihilation events involves new difficulties, because the energy-momentum conservation equations do not suffice to determine uniquely the momenta of the three photons. In the case of two-photon annihilation, the energies and momentum components of the two rays constitute a set of eight variables. There are four relations among these variables from the conservation laws and two from the energy-momentum relation ($E^2 = p^2c^2$) for photons. Hence the momenta of the two quanta are completely fixed by assigning a direction in space to one of them, since this involves assigning values to two space angles.

For the three-photon problem the energies and momentum components are twelve in number, three momentum components and an energy for each photon.

Four relations among these variables follow from conservation laws, and three from $E^2 = p^2c^2$. There are then five variables left, and fixing one photon direction removes only two more unknowns. It is necessary to fix the remaining three variables by giving a direction in space to another photon and by fixing the angle between the third photon and one of the other two, or by some equivalent procedure. The third photon must be in a plane with the other two and the source because the positron will annihilate at effectively thermal energy, and the total momentum is therefore practically zero. It also follows that the three photons cannot all be in the same half-plane. The fact that the three-photon rays are only coplanar while the two-photon rays are collinear makes the former effect considerably more difficult to detect.

The plan of the experiments was to mount three scintillation gamma-ray counters in a plane about a source of positrons, and to measure the counting rates of triple coincidences occurring when the positrons are stopped in various materials surrounding the source (Figure 2(a), and A, Figure 2(b)). The background of random triple coincidences was measured with one of the counters rotated 45° out of the plane (B, Figure 2b).

B. Counters and Sources

The scintillation counters consisted of NaI(Tl) crystals 4 cm in diameter and 2.5 cm thick, mounted in front of RCA 5819 photomultiplier tubes. The crystals were surrounded by a one mil aluminum foil reflector except for the face through which light passed to the cathode of the phototube. Each crystal was then enclosed in a cylindrical plastic box filled with mineral oil and sealed to prevent moisture from deteriorating the crystal, which is extremely hygroscopic. That end of the plastic box in contact with the photomultiplier was made of Lucite and was ground to fit the tube face, a thin film of microscope immersion oil serving to complete the optical bond between crystal and multiplier. The entire assembly of crystal, container, and phototube was wrapped with black Scotch tape to insure light tightness.

Each counter was mounted in a holder consisting of an aluminum box containing the voltage divider for the phototube and other necessary connections, and a bakelite cylinder supporting the phototube. A cylindrical lead snout protected the crystal from scattered radiation (Figure 3). The whole counter was mounted about eighteen inches above a circular aluminum base plate, with two of the three counters supported on movable sleds which could be rotated in a horizontal plane about the center post. The base plate was divided into ten degree intervals so that it served as a protractor for positioning the counters around a source mounted on the center column. The third counter could be moved 45° out of the horizontal plane of the source. Photographs of the apparatus are shown in Figures 4 and 5.

Two source holders were used, one in which the positrons were stopped in 1/16 inch aluminum surrounding the source, the other in which some or all of the positrons could be stopped in gas. The two holders are shown in

Figure 6. The aluminum bell (Figure 6(b)) has an inlet and outlet for the various gases which were used at pressures up to 350 pounds per square inch. It was not deemed safe to use greater pressures because of the thin walls of the container. The source used with this holder was Na^{22} in the form of activated NaCl crystals deposited on a thin Zapon (plastic) film. This film was supported on a "ring stand" of fine steel music wire resting on the brass base of the bell. The source was covered with another Zapon film to prevent contamination of the gas container. The two films were very thin, as shown by interference colors from visible light. It is therefore believed that less than one per cent of the positrons were stopped by these films, and that most of them annihilated in the gas or in the walls of the bell.

The active spot of NaCl crystals was less than six millimeters in diameter, and was centered in the bell to within about one millimeter of the central position shown in Figure 6(b). The source atoms were thus on the average about one centimeter from the walls of the bell.

Na^{22} was used as a source of positrons because of its long life ($T_{1/2}$ = 3.0 yrs.) and availability, though the 1.3 Mev gamma-rays coincident with the positrons from this source were a major source of background. The Na^{22} was obtained from Oak Ridge as an aqueous NaCl solution of one millicurie total activity. Portions of this primary source averaging about 10^6 disintegrations per second in activity were withdrawn and used in the three-photon measurements. The end point of the Na^{22} positron spectrum is .575 Mev, corresponding to a range of .16 gm/cm^2 or about .06 cm of aluminum. The one-sixteenth inch walls of the source holders were therefore adequate for stopping all positrons.

C. Electronic Apparatus

A block diagram of the electronic apparatus is shown in Figure 9. It is to be noticed that each 5819 photomultiplier has two outputs, one leading to a linear amplifier and thence to a differential pulse height selector, the other to the fast triple coincidence circuit. The pulses on the plates of the phototubes were used to feed the fast coincidence circuit, a diagram of which is shown in Figure 10(a). It is of the crystal diode type used by De Benedetti and Richings[10], as modified for triple coincidences. The pulses from the photomultipliers are shaped by the RC differentiating circuits at the plates of the tubes instead of by the shorted transmission lines of the above authors, and the pulses were amplified by travelling-wave amplifiers before reaching the coincidence circuit.

The resolving time of this circuit was measured by feeding random pulses from three independent sources into the counters and measuring the resultant coincidence rate. If S_1 is the single counting rate in the first counter, etc., then the triple coincidence rate will be $6S_1S_2S_3\tau^2$, where τ is called the resolving time of the circuit. The distribution of pulse heights in each counter was the same as during the subsequent three-photon experiments. The value of τ measured in this way was 1.0×10^{-7} seconds.

The positive pulses on the last dynode of each multiplier tube were fed to the differential pulse height selectors (DPHS) through model A-1 linear amplifiers. Using these dynode pulses, which were about seventy per cent as large as the negative pulses on the plate, it was possible to study the heights of the coincident pulses without affecting the operation of the fast coincidence circuit. The type of DPHS used is that described by Minton[18]; it has a single channel and a range of one hundred volts.

Selection of a certain band of pulse heights (voltages) corresponds to selecting a certain energy interval for the electrons causing the scintillations in the sodium iodide.

The chain of events leading to an output pulse is as follows: a single gamma-ray is scattered or absorbed in the crystal, thus losing part or all of its energy to an electron by Compton scattering or photo-absorption. This electron will then lose its kinetic energy by exciting molecules of the crystal, these molecules emitting photons which impinge upon the photocathode of the multiplier and initiate electron avalanches, these finally appearing as pulses across the plate resistor. One therefore expects that the output pulse amplitude will be proportional to the energy of the secondary electron produced in a single scattering event within the crystal if the gamma-ray escapes without further collision. However, in the crystals used for these experiments there was appreciable probability that a gamma-ray which had suffered one Compton scattering would be photo-absorbed or scattered again before escaping the crystal. The consecutive scattering or absorption events occurred within a time short compared to the resolution of the circuits, so that the pulses added to give a single pulse as large as that produced when the entire energy loss of the gamma-ray was dissipated in a single collision.

The conclusion that the output pulses were proportional to the energy loss in the crystal is justified only if certain effects are negligible. These include the statistical variation in the number of electrons ejected from the photocathode by a fixed size of light pulse, the variation of optical bonding and efficiency over the crystal, etc. These effects are experimentally shown to be small, as we shall now see.

Sodium iodide is mostly iodine by weight, and the rather high atomic number (53) of this element results in a large photoelectric cross-section in NaI for gamma-rays with energies below $\sim$700 kev. Thus a large, sharply defined photo-peak appears in the spectrum of Figure 8, Curve 2 from 510 kev annihilation gammas. The position of the center of this photo-peak serves to measure the energy of the gamma-rays, and the 1.3 Mev photo-peak from Na^{22} in Curve 1 verifies that the pulse heights vary linearly with electron energy. An additional calibration point is obtained from the photo-peak from 364 kev gammas emitted by I^{131}, and this point is consistent with the energy scale established from Na^{22}.

The resolution of the NaI crystals used here is also demonstrated in Figure 8. In Curve 1 not only the photo-peaks at 510 kev and 1.3 Mev are clearly defined, but also visible are the Compton peaks corresponding to these two primary energies. The 510 kev Compton peak is superimposed on the low-energy pulses from noise and scattering. In Curve 2, the spectrum of mc^2 coincident pulses, the background at low energies has disappeared because scattered radiation and noise pulses rarely contribute to the coincidence rate. The high energy pulses have also decreased greatly in relative intensity, since there is no angular correlation between the nuclear gamma-ray and an annihilation ray. Hence a coincidence between them is less probable by a factor $\frac{\omega}{4\pi}$ (percentage solid angle subtended by one of the two counters) than a two-photon annihilation coincidence. In this case $\frac{\omega}{4\pi} \approx 10^{-2}$, leaving only a small residue of high-energy coincidences.

One disadvantage of NaI is that the light intensity I in a scintillation pulse decreases exponentially ($I = I_o e^{-t/t_o}$) with a time constant t_o = 2.5×10^{-7} seconds, as compared to 5×10^{-9} seconds and less for certain organic phosphors. This means that in NaI there is a lower limit set on the time duration of the pulse even after differentiating or clipping, thus making it difficult to attain resolving times shorter than 10^{-7} seconds. In experiments for which this is not an important limitation, however, the efficiency and energy resolution of large NaI crystals are great advantages.

Finally, it is apparent from the block diagram of Figure 9 that a count will register on the scalar only if (a) a fast triple coincidence occurs between the counter pulses and (b) pulses of the proper height trip all DPHS's simultaneously, i.e. within 2 x 10^{-6} seconds, with the fast coincidence. The pulses entering the DPHS's are identical with those which trip the fast circuit, except that they are taken from the last dynode instead of the plate of each photomultiplier. Thus the change of final counting rate which occurs when the band of pulse heights accepted by DPHS #1 is varied while the settings of the other two DPHS's are kept constant corresponds to taking an energy spectrum of the coincident pulses in Counter #1. This is the method by which the energy spectra of three-photon annihilation events were studied.

IV. Three-Photon Annihilation in Solids

In a first experiment, positrons from an Na^{22} source of $\sim 10^6$ disintegrations/sec. were allowed to stop in aluminum and glass. The symmetric arrangement of counters (Figure 2(a)) was used, the face of each crystal being 12.0 cm from the source. With the pulse height selectors set to accept electron pulses of energy between about 100 kev and 500 kev, the following counting rates were observed:

Counters coplanar	2.42 ± .14 counts/min.
Counters not coplanar	1.37 ± .14 counts/min.
Difference	1.05 ± .19 counts/min.

It seems difficult to account for this difference in any other way than by three-quantum annihilation, for no random effect would display the planar characteristic found here. At the time this measurement was made our knowledge of the source strength and counter efficiencies was too meager to permit a quantitative comparison with the theoretical predictions; however, a rough estimate showed agreement within a factor two with the theoretical value of one triplet annihilation per 370 singlet. More accurate measurements will be described below.

The evidence that this difference between the coplanar and non-coplanar counting rates actually resulted from three-photon annihilation was strengthened by a pulse height analysis of the coincident rays in one of the counters. (Hereafter the difference rate between counters coplanar and counters not coplanar will be called the 3-γ counting rate). A spectrum of coincident pulses in counter #1 was measured with a bandwidth of 200 kev, the region from 0 to 600 kev being covered in three steps. The results are indicated as a bar graph in Figure 11. The same figure shows the spectrum in counter #1 from primary radiation of 340 kev energy, as taken from the three-quantum spectrum in positronium (see Section VI). The statistical errors for the histogram are quite large, as shown by the vertical barred line, but the similarity between the two spectra is nonetheless striking.

In order to make an accurate check of the theoretical estimate of $\frac{\sigma_{3\gamma}}{\sigma_{2\gamma}}$ for free positron electron pairs, we attempted an absolute measurement of the three-photon annihilation rate of positrons stopping in aluminum. The discussion of Section II led us to this method of checking the theory; if the annihilation events within a solid occur in collisions between free positrons and electrons, the theoretical triple coincidence rate will be

$$\text{(a)} \quad C = \frac{1}{371} N \lambda \epsilon_1 \epsilon_2 \epsilon_3 \Gamma ,$$

where N is the number of positrons annihilating per second, λ is a factor compensating for the absorption of the gamma-rays in the material surrounding the source, ϵ_i is the efficiency of the i^{th} counter, and Γ is a factor expressing the relative probability of occurrence of the symmetric arrangement of three-photon annihilation gamma-rays and the geometrical efficiency for detecting such an arrangement of rays. The detailed calculation of Γ is carried out in Appendix A.

Since the symmetric arrangement of counters was used in this experiment, the ϵ's of equation (a) are for gamma-rays of $2/3\ mc^2$ energy. A method of double coincidences was used to determine both the source strengths and the counter efficiencies. If two counters with efficiencies $\epsilon_i^{mc^2}$ and $\epsilon_j^{mc^2}$ are placed around a source of N positrons persecond in the usual way for detecting two-quantum annihilation (Figure 7a), then the coincidence rate between them will be

$$C_{ij} = 2\ N\ \epsilon_i^{mc^2}\ \ \epsilon_j^{mc^2}\ w_i/4\pi$$

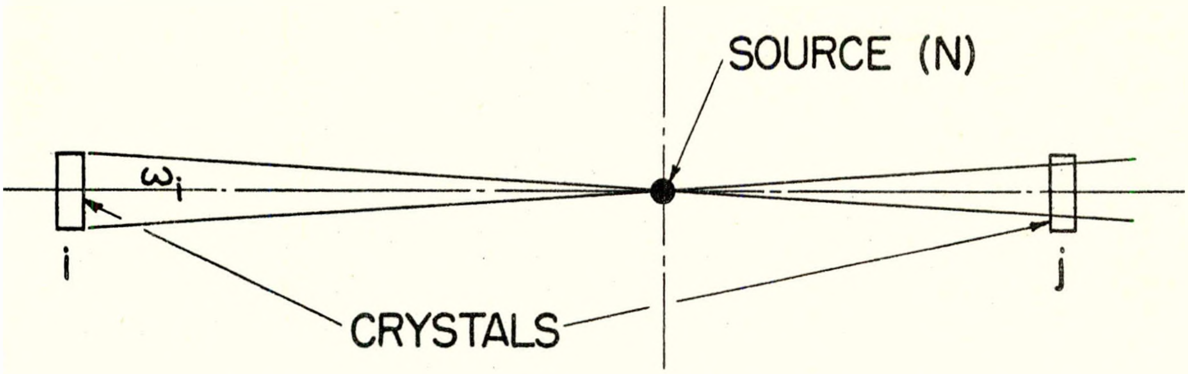

FIGURE 7a

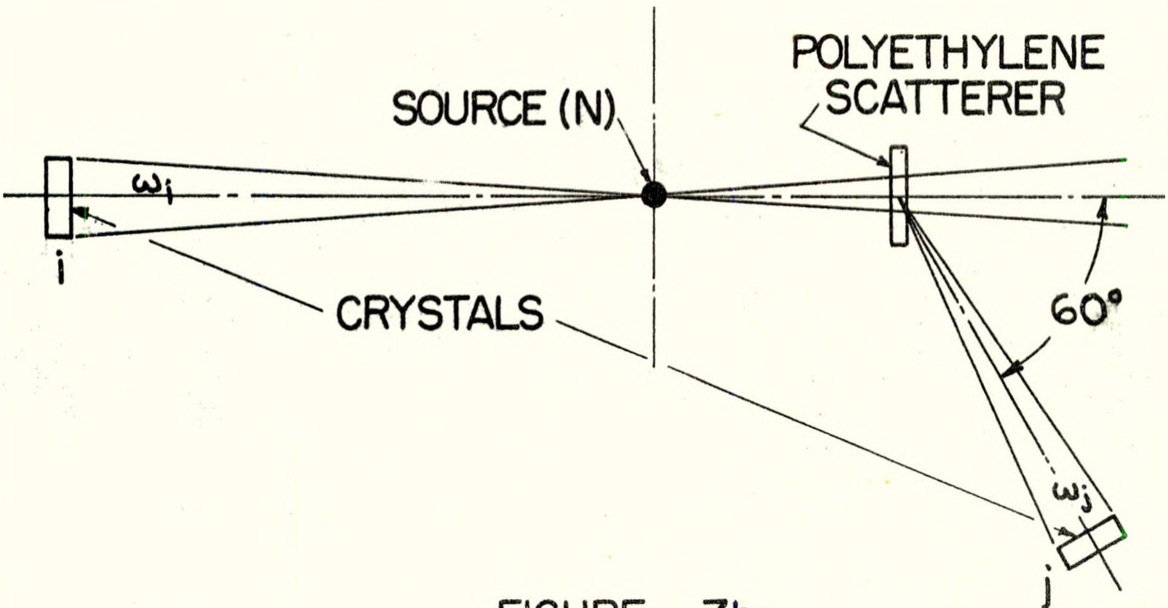

FIGURE 7b

Here w_i is the solid angle subtended by counter i and it is assumed that w_i defines the beam of annihilation rays, i.e. $w_i < w_j$. The single counting rate in counter i from two-quantum events will be $S_i = 2\ N\ \epsilon_i^{mc^2}\ w_i/4\pi$ per second, and similarly for counter j. Then the ratio C_{ij}/S_j will yield a value for $\epsilon_i^{mc^2}$, and substitution of this efficiency in the expression for S_i will give N in disintegrations per second. The source used in measuring the absolute three-photon annihilation rate in aluminum (Figure 6a) was found to have an intensity $N = (9.22 \pm .57) \times 10^5$ dis/sec.* It was also found that the efficiencies $\epsilon_1^{mc^2}$, $\epsilon_2^{mc^2}$, and $\epsilon_3^{mc^2}$ were very nearly equal. When corrected to the efficiency for a parallel beam of gammas (i.e. an infinite source distance), the mean ϵ^{mc^2} for all counters was $.376 \pm .025$. The errors in this value and in the source strength were almost entirely due to uncertainty in S_i. The actual singles rate measured includes not only the contribution from annihilation radiation (S_i) but also many counts from scattered low energy radiation and photomultiplier tube noise. This is apparent from Figure 8, which shows spectrum of single counts from Na^{22} in one of the three counters (Curve 1). Curve 2 shows the spectrum of double coincidences from two-photon annihilation in the same counter. The two spectra are normalized to the same height at the mc^2 photo-peak, so the area between the two curves is a measure of the contribution to the singles spectrum from scattering and tube noise. (See Section III-C for further discussion of Figure 8). The extrapolation of the 1.3 Mev spectrum to low energies is somewhat uncertain, and subtracting the mc^2 spectrum from the total single counts spectrum to find the noise contribution introduces considerable error. As a result the error in S_i as measured by the spread in values of S_i/w_i for the three counters is about six per cent, while the spread in C_{ij}/w_i for various combinations of two counters is less than one per cent. It was this latter fact which led us to believe that all the efficiencies were equal at a primary energy of mc^2 The measured efficiencies

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* This procedure was also used to measure the intensity of a one-millicurie Na^{22} source. The result agreed with a calibration performed at Oak Ridge by other methods.

at 2/3 mc^2 also appeared to be equal.

The $\epsilon_i^{2/3\ mc^2}$ were measured with the two-counter arrangement of Figure 7(b). The beam of two-quantum annihilation rays defined by w_i was partially Compton scattered in the .455 inch polyethylene scatterer through an angle of 60°. The energy of the scattered rays is therefore 2/3 mc^2, and the coincidence rate between counters i and j is now

$$C'_{ij} = 2\ N\ \Sigma\ \lambda\ \epsilon_i^{mc^2}\ \ \epsilon_j^{2/3\ mc^2}\ \ \frac{w_i}{4\pi}\ w_j .$$

Here Σ is the probability that an mc^2 gamma-ray will be scattered through the proper angle, and can be calculated from the Klein-Nishina formula(16). λ is a correction factor for the absorption of the scattered gamma-rays in the scatterer itself, the finite size of the source of scattered rays seen by counter j, etc. The value of $\epsilon^{2/3\ mc^2}$ for a parallel beam of gamma-rays was measured to be .542 $\pm$ .036, the error again arising from the uncertainty in $S = 2\ N\ \epsilon^{mc^2}$ as measured with the geometry of Figure 7(a).

The expected value of C (see Appendix A for outline of the calculations) for our apparatus was 1.24 $\pm$ .16 counts/min., the probable error being the result of inaccuracies in N, ϵ^3, and Γ. The experimental value for C was .92 $\pm$ .10 counts/min. which is in reasonably good agreement with the expected counting rate. The slight discrepancy between the two values is not considered significant, and may be removed by further measurements. In particular, the stability of the electronic apparatus over long counting periods should be improved.

The agreement between the expected and measured counting rates indicates that no bound system of positron and electron can exist within solids, and that the theoretical calculations of Ore and Powell(3) are correct.

V. Three-Photon Annihilation in Gases

In order to confirm the stability of positronium in Freon-12(CCl_2F_2), a source of Na^{22} about equal in strength to that used in the experiment above was deposited between 1/2 mil aluminum sheets and centered in the gas container of Figure 6(b). The bell-shaped container was then filled with Freon at about six atmospheres pressure, so that those positrons escaping the aluminum covering the source would have a large probability of stopping in the gas within the container. Again with the symmetric counter arrangement, and with the DPHS's adjusted with the same bandwidths as above, the following counting rates were observed:

Counters coplanar	11.85 ± .43 counts/min.
Counters not coplanar	1.33 ± .15 counts/min.
Difference	10.52 ± .46 counts/min.

The tenfold increase of the 3-γ rate confirms the stability of positronium in Freon. An analysis of the pulse height spectrum in counter #2 is shown in Figure 13, Curve II. Curve I is the mc^2 spectrum. The photo-peak in Curve II lies at 340 kev = 2/3 mc^2 as expected for this counter arrangement. Also one can see that the relative height of the Compton peak at 340 kev is smaller than that at 510 kev, because the photoelectric cross-section increases much more rapidly with decreasing energy than does the Compton cross-section. In Figure 12 the dependence of the 3-γ coincidence rate upon pressure in Freon-12 is shown. The value at zero pressure is obtained by filling the bell with air at atmospheric pressure. Because oxygen strongly quenches ortho-positronium, the triplet annihilation rate with air in the bell is equal to that with vacuum in the bell, except for possible geometrical effects resulting from the stopping of some positrons within the air rather than in the walls of the bell. These effects are small compared to the statistical errors of this measurement, which shows that the stopping of positrons in the Freon rather than in the aluminum walls is the cause of the increase in 3-γ counts.

The source between aluminum foils was then replaced with one mounted between Zapon films as in Figure 6(b). This source had a strength measured as $(1.36 \pm .08) \times 10^6$ dis./sec., and the 3-γ annihilation rate indicated that about twelve per cent of all positrons stopping in the Freon within the container formed positronium. The triplet annihilation rates in several other gases were compared to that in Freon, with results shown in Table I.

Table I

Material	Pressure (psia)	Relative Electron Density	Relative 3-Counting Rate (corrected to equal electron densities)
Freon-12	75	1.00	1.00
SF_6	62	1.00	$.79 \pm .04$
He	215	.0988	$1.15 \pm .15$
H_2	215	.0988	$.96 \pm .25$
N_2	212	.683	$.84 \pm .06$
O_2	122	.900	$.006 \pm .018$

The values of pressure and relative electron densities given in the second and third columns are those at which the 3-γ rates were actually measured. The last column gives the 3-γ rates corrected to equal electron densities. Since equal electron densities mean only approximately equal stopping powers, the values given in the last column do not give with high accuracy the relative amounts of positronium formed when equal numbers of positrons are stopped in different gases. However, these values are in rough agreement with those given by Deutsch and Pond[19]. The strong quenching of ortho-positronium in oxygen is noteworthy, and is attributed to the exchange mechanism discussed in Section II.

The finding that SF_6 is a favorable substance for positronium formation greatly facilliatated these investigations.* The density (.25 gm/cc) of SF_6 which can be attained at 26°C makes it possible to stop most of the positrons in a centimeter or so of this gas. In fact, Figure 14 indicates that above .10 gm/cc most of the positrons stop in the SF_6, and few reach the walls of the container. The rising portion of the curve at low densities shows that some of the positrons are still reaching the walls of the bell, but increasing the density beyond .11 gm/cm^3 results only in a greater quenching of the ortho-positronium atoms before annihilation can occur. Collisions of positronium atoms with SF_6 molecules cause flipping from ortho-positronium to the para-state, this process competing with the three-quantum annihilation of the ortho-state.

The flipping cross-section per SF_6 molecule is here calculated from the experimental decrease of 3-γ counts occurring when the density of SF_6 varies from .11 to .25 gm/cc. The fraction <u>f</u> of 3-γ counts left at the higher density must satisfy the equation

$$\frac{1-f}{f} = \frac{p}{\lambda_o} = \frac{v\,\sigma\,(N_2 - N_1)}{\lambda_o}$$

where $\lambda_o = 7.2 \times 10^6$ sec^{-1} is the decay constant of ortho-positronium. The

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* Suggestion was originally made by D. Grove of this laboratory that the similar insulating properties of Freon-12 and SF_6 might indicate that both would be favorable gases for positronium formation.

probability of flipping $p = v\,\sigma(N_2 - N_1)$, where v is the mean velocity of a thermalized positronium atom, $(N_2 - N_1)$ is the change in the number of SF_6 molecules per cm^3 between the two densities, and σ is the flipping cross-section of the molecule. The value found for σ is 9.7×10^{-22} cm^2, this including a correction for absorption of the three gamma-rays within the gas at the higher density. Uncertainty in the knowledge of gas density is the primary source of error, which is estimated to be 15 per cent or less. This measurement gives in reality an upper limit upon σ, since part of the effect may have been caused by halogen contamination of the SF_6 used. Deutsch has found that halogens are extremely effective quenchers of ortho-positronium, possibly because of the formation of positron-halogen compounds.

This upper limit on σ in SF_6 is to be compared with the corresponding cross-sections in NO and O_2 of 7.6×10^{-17} cm^2 and 4.0×10^{-19} cm^2 as measured by Deutsch(7). Evidently the exchange process responsible for flipping in NO and O_2 is much less effective in the case of SF_6, and perhaps magnetic interaction is responsible for the quenching effect in this gas.

An attempt was made to detect the formation of positronium when the positrons are stopped in liquefied Freon or nitrogen. The source of positrons was painted on the inside bottom of a small Dewar flask and covered with a thin plastic film. About 4×10^5 positrons per second pierced the film and were stopped in the liquefied gas contained in the Dewar. No 3-γ rate above that to be expected from a similar intensity of positrons stopping in solids was observed with either liquid nitrogen or Freon in the flask. It is therefore concluded that stable positronium cannot be formed in these substances. The measurements will be extended to liquid hydrogen and helium in the near future.

As discussed in Section III-A, the momenta of the three gamma-rays from triplet annihilation which we detect in coincidence should be completely determined by the angles between the three counters. A verification of this prediction was obtained by measuring the energies of the coincident rays in the three counters for three different angular arrangements. The three sets of angles used were $120^\circ - 120^\circ - 120^\circ$, $150^\circ - 150^\circ - 60^\circ$, and $90^\circ - 150^\circ - 120^\circ$, and the resulting spectra are displayed in Figure 15. Each one of the nine graphs shows the spectrum in one of the counters when that counter is placed as indicated by the small figure in the upper right corner of each graph. The spectra in the first column of three graphs were all measured in Counter No. 1, those in the second column in Counter No. 2, etc. For example, the central graph shows the spectrum in Counter No. 2 when that counter is placed opposite a 150° angle in the $150^\circ - 150^\circ - 60^\circ$ arrangement.

The dotted spectra in the top row of graphs are from the mc^2 radiation from two-quantum annihilation, and are shown merely for comparison. They were used to establish the energy scales (abscissae). The position of the photo-peak in each of the nine spectra gives the energy of the coincident rays striking the crystal. The energies predicted in each case by the conservation laws are indicated by the heavy vertical lines in each graph. (In the top row of graphs a line indicating $E = mc^2$ is also shown.) In all graphs the experimental spectra have photo-peaks at the energies predicted by the conservation laws. Slight deviations are not considered significant because of the limited resolution (about 30 kev) attainable with the DPHS bandwidths used.

The three-photon spectra (solid curves) were measured with DPHS channel widths of about 90 kev, the points on the curves being placed at the centers of the corresponding DPHS bands. The maximum counting rate was normalized to a value of ten, and the areas under the curves consequently do not indicate the relative intensities of three-photon annihilation events for the different arrangements. We feel that our knowledge of efficiency versus energy for the counters is not yet dependable enough to justify any conclusion concerning these relative intensities, a measurement of which could serve as a check on the energy distribution of Figure 1(b). However, the counting rates for different angular arrangements were approximately equal, indicating a flat angular distribution in agreement with theory.

The three counters were also placed at intervals of seventy-five degrees with each other, an arrangement of three-photon rays forbidden by conservation of momentum. The observed counting rate of $3-\gamma$ coincidences was zero.

VI. Summary

The following conclusions may be drawn from the experimental work presented in this thesis:

1. The existence of the phenomenon of three-quantum annihilation has been unambiguously demonstrated by the direct method of detecting the three gamma-rays of annihilation in coincidence.

2. It was experimentally verified that the laws of conservation of energy and momentum are satisfied in this process.

3. It was found that three-photon annihilation occurs in condensed materials with an intensity consistent with that expected for random collisions between free positrons and electrons. This observation supports the validity of the theory and shows that no positronium is formed in condensed materials.

4. About twelve per cent of the positrons stopped in certain gases form positronium. In some gases no positronium is observed, probably because ortho-positronium is rapidly converted to the para-state before triplet annihilation can occur.

The work of this thesis could be extended to the investigation of the following problems without serious modification of the equipment used:

a. The polarization of the three gamma-rays.

b. The angular distribution of the three gamma-rays.

c. The chemistry of positronium.

d. Studies of the interactions of positrons in condensed materials.

VII. Appendix A - Computation of the Factor Γ

The calculation of the factors in the equation (a) of Section IV for the coincidence rate from three-photon annihilation will be carried out in detail. The triple coincidence rate for angles α, β, γ between the counters is given by:

$$(1) \quad C = \frac{1}{3!1} N \lambda \epsilon^3 \Gamma(\alpha, \beta, \gamma)$$

N = Source strength in number of positrons/sec.
ϵ = Counter efficiency
λ = Absorption of gamma-rays in material around source.

$$(2) \quad \Gamma = \left[\frac{\text{Intensity of 3-}\gamma\text{ events with }(\alpha, \beta, \gamma)}{\text{Average intensity of 3-}\gamma\text{ events for all angular arrangements}}\right]$$

$$\times \left[\text{Geometrical Efficiency for detecting sets of Three Simultaneous Randomly oriented Coplanar Gamma-Rays}\right]$$

$$\text{or (2a)} \quad \Gamma = \Gamma_1 \times \Gamma_2$$

where Γ_1 and Γ_2 are the two quantities in brackets. These are calculated below.

a. Calculation of Γ_1.

The intensity $I(\alpha, \beta)$ of three-photon events with angles α, β and γ between the rays can be found from the theoretical transition probability per unit time for the triplet annihilation of a free positron-electron pair at low velocity. Expressed as a function of the momenta k_1 and k_2 of two of the photons, this transition probability is:

$$(3) \quad I(k_1, k_2)\, dk_1\, dk_2\, d\Omega_1 = \frac{e^6}{m^2} \times 2 \left[(1 - \cos\alpha)^2 + (1 - \cos\beta)^2 + (1 - \cos\gamma)^2\right] \times \frac{dk_1}{m} \frac{dk_2}{m} \frac{d\Omega_1}{4\pi}$$

where dk_1, dk_2 are the intervals about k_1, k_2 and $d\Omega_1$ is the element of solid angle in which k_1 lies. The energy dependence of I can be expressed in terms

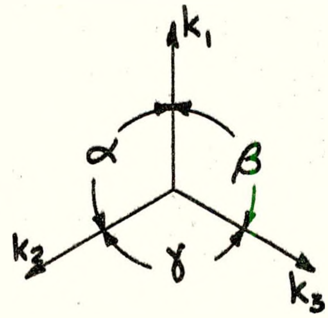
of the angles α, β (see momentum diagram) by inserting the Jacobian $\frac{\partial(k_1, k_2)}{\partial(\alpha, \beta)}$.
The laws of conservation of energy and momentum give relations between k_1, k_2, k_3 and α, β:

$$k_1 + k_2 + K_3 = 2mc^2$$

$$k_2 \sin\alpha = k_3 \sin\beta$$

$$k_1 = -k_2 \cos\alpha - k_3 \cos\beta$$

These simultaneous equations can be solved to give the k's as functions of the angles, and $\frac{\partial(k_1, k_2)}{\partial(\alpha, \beta)}$ can then be obtained.



Momentum Diagram

(4) $$I(\alpha, \beta)\, d\alpha\, d\beta\, d\Omega_1$$

$$= I(k_1, k_2) \frac{\partial(k_1, k_2)}{\partial(\alpha, \beta)} d\alpha\, d\beta\, d\Omega_1$$

$$= \frac{e^6}{m^2} 8 \left[(1 - \cos\alpha)^2 + (1 - \cos\beta)^2 + (1 - \cos\gamma)^2\right] \frac{\sin\alpha \sin\beta \sin\gamma}{(\sin\alpha + \sin\beta + \sin\gamma)^3}$$

$$d\alpha\, d\beta\, \frac{d\Omega_1}{4\pi}$$

Note that I is dependent only on α and β because $\gamma = 2\pi - \alpha - \beta$. From the definition of Γ_1 in equations (2) and (2a) we see that

(5) $$\Gamma_1 = \frac{I(\alpha, \beta)}{\langle I(\alpha, \beta)\rangle}$$ averaged in all directions

$$= \frac{I(\alpha, \beta)}{\frac{1}{2\pi} \cdot \frac{1}{2\pi} \cdot \frac{1}{4\pi} \cdot \iiint I(\alpha, \beta)\, d\alpha\, d\beta\, d}$$

We assume $I(\alpha, \beta)$ constant over the counters, and equal to $I(120^\circ, 120^\circ)$. Then it follows from

(6) $$\iiint I(\alpha, \beta)\, d\alpha\, d\beta\, d\Omega_1 = 8\,(\pi^2 - 9)\,\frac{e^6}{m^2}$$

hat $$\Gamma_1 = \frac{I(120°, 120°)}{\frac{1}{2\pi}\cdot\frac{1}{2\pi}\cdot\frac{1}{4\pi}\cdot\frac{8e^6}{m^2}(\pi^2 - 9)}$$

(7) $$\Gamma_1 = \frac{\pi^2}{\pi^2 - 9}$$

b. Calculation of Γ_2

The factor Γ_1 states how much the (120°, 120°, 120°) arrangement more probable than the average for all arrangements. The factor Γ_2 then the geometrical efficiency for detecting events in which three oplanar gamma-rays are distributed at random over all angles, i. e.

(8) $$\Gamma_2 = \frac{6}{(2\pi)^3}\left\langle \alpha_1\,\alpha_2\,\alpha_3 \right\rangle_{\text{average}}$$

re α_1, α_2, α_3 are the apertures of the counter in the plane of the three andom rays, and the average is to be taken over all such planes intercepted the counters. The diagram on the following page represents one of the ounters as a disk tangent to the unit sphere at point C.

follows with minor approximations that

$\widehat{CP} = \frac{\pi}{2} - \tau$, since $\vec{n}$ is the normal to plane OAB

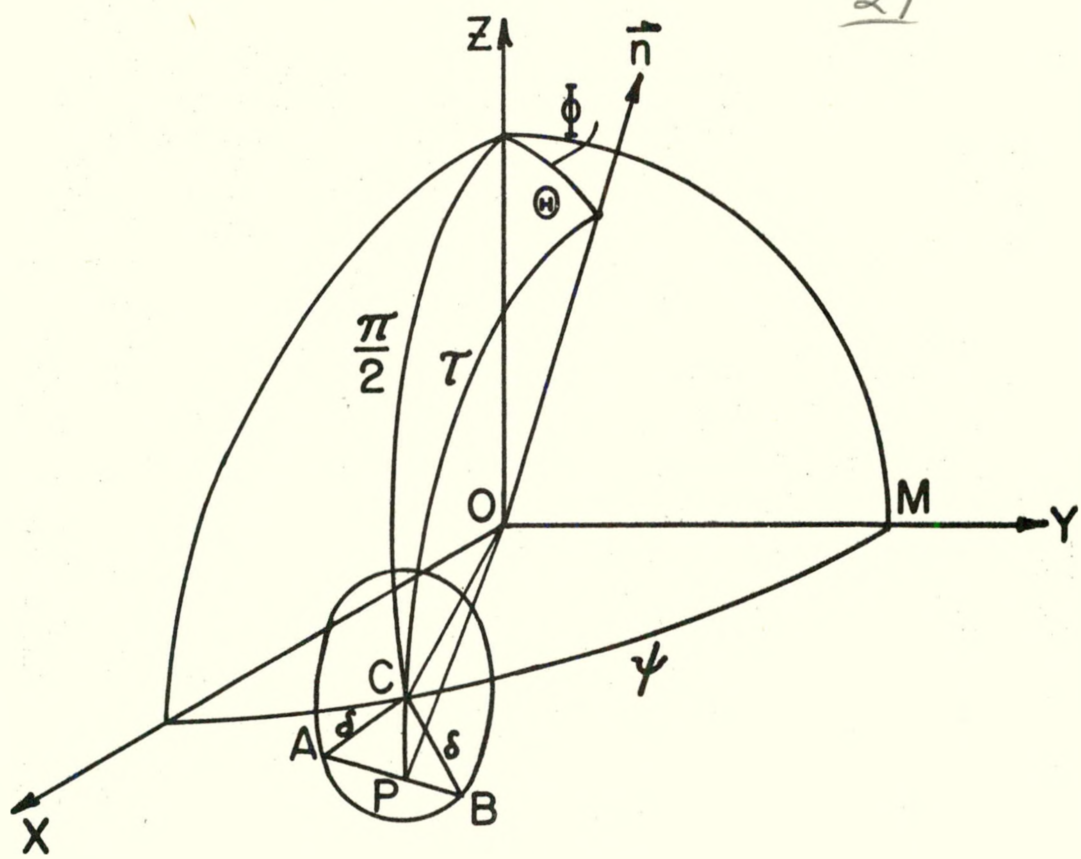
$$\cos\tau = \cos\Theta\cos\frac{\pi}{2} + \sin\Theta\sin\frac{\pi}{2}\cos(\psi - \phi)$$

$$= \sin\Theta\cos(\psi - \phi)$$

r small δ = half-aperture for one counter,

$$\widehat{AB} = 2\sqrt{\delta^2 - (\widehat{CP})^2} = 2\sqrt{\delta^2 - \sin^2\widehat{CP}} = 2\sqrt{\delta^2 - \cos^2\tau}$$

$$= 2\sqrt{\delta^2 - \sin^2\Theta\cos^2(\psi - \phi)}$$



$\vec{n}$ = normal to plane OAB

ψ = angle between C and M

τ = angle between $\vec{n}$ and C

The XY plane is the plane of the counter axes. The plane OAB is the plane of the gamma-rays. AB is the effective aperture of the counter to gamma-rays in the plane OAB.

For three counters at 120° with each other this gives

$$(9)\quad \langle \alpha_1\,\alpha_2\,\alpha_3\rangle_{av} = \frac{8\,\delta^3}{2\pi}\int_0^{2\pi} d\varphi \int_0^{\pi/2} \sin\theta\, d\theta$$

$$\sqrt{\left[1-\frac{\sin^2\theta}{\delta^2}\cos^2\varphi\right]\left[1-\frac{\sin^2\theta}{2}\cos^2(120-\varphi)\right]\cdot\left[1-\frac{\sin^2\theta}{\delta^2}\cos^2(120+\varphi)\right]}$$

The only values of θ which contribute to this integral will be for $\sin\theta \leq \delta$. Since the effective δ for each counter is about .15 radians, $\sin\theta$ may be replaced by θ in (9). Then, if

$$y = \frac{\theta^2}{\delta^2} \approx \frac{\sin^2\theta}{\delta^2}$$

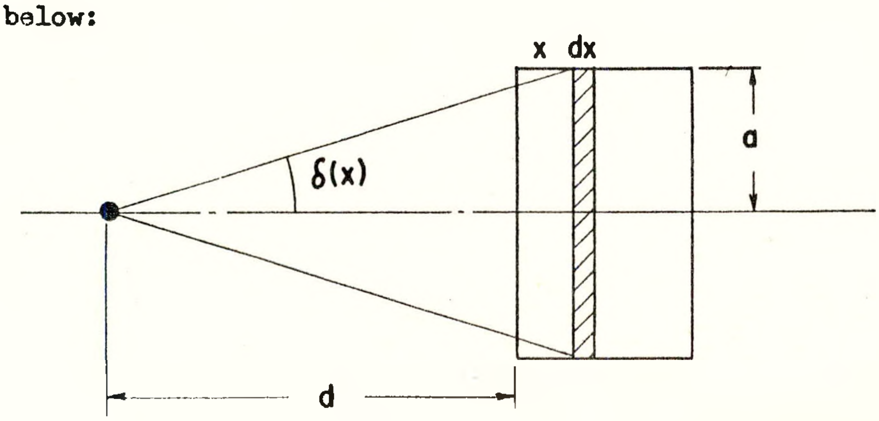
$$(10)\quad \langle \alpha_1\,\alpha_2\,\alpha_3\rangle_{av} = \frac{4\,\delta^5}{\pi}\int_0^{2\pi} d\varphi\int_0^{y_{max}} dy \,\cdot$$

$$\cdot\sqrt{\left[1-y\cos^2\varphi\right]\left[1-y\cos^2(60+\varphi)\right]\left[1-y\cos^2(60-\varphi)\right]}$$

This was integrated graphically to give:

$$(11)\quad \langle \alpha_1\,\alpha_2\,\alpha_3\rangle_{av} = 2.46\,\delta^5$$

The δ^5 which must be inserted is the result of averaging the fifth power of the half-aperture over the depths of the counters, taking into account the absorption of the gamma rays in the counter. The procedure for doing this is shown below:



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$$(12)\ \langle\delta\rangle^5 = \left[\frac{\int_0^t \left(\frac{a}{d+x}\right)^2 e^{-\mu x}\,dx}{\int_0^t e^{-\mu x}\,dx}\right]^{5/2} = .682\ \frac{a^5}{d^5}$$

The value of the absorption coefficient u in NaI was deduced from the efficiency measurements of Section III-B, and error in these measurements contributes to the final error in Γ_2. (Note that the factor ϵ^3 also appears in C). The calculation of the absorption factor λ can easily be carried out from the knowledge of the absorption coefficient τ for 2/3 mc^2 gamma-rays in aluminum. Then

$$(13)\quad \lambda = \frac{I}{I_o} = e^{-3\tau t} \qquad t = \text{thickness of aluminum around source.}$$

Using the theoretical value of $\frac{\sigma_{3\gamma}}{\sigma_{2\gamma}}$ given by Ore and Powell, the experimentally determined value for N, and the results of equations (7), (11), (12), and (13), we find that under the experimental conditions

$$C = 1.24 \pm .16 \text{ counts/min.}$$

VIII. Appendix B - Measurement of Background

As mentioned in Section III-A, the background of random coincidences in the three counters can be measured most conveniently by rotating one of the three counters out of the plane determined by the source and the axes of the other two counters. We will here consider the various types of background and the validity of this method of measuring total background.

Under the conditions of a typical experiment each crystal was about 12 cm from the source, which was about 10^6 disintegrations/sec. In this case the singles counting rate in each counter was about 5,000 counts/sec. The rate of triple coincidences from random coincidences among single counts is $6\ S^3\tau^2 = 7.5 \times 10^{-3}$ counts/sec. or .45 counts/min. This is approximately one-third of the total background measured, and is obviously independent of the coplanarity of the counters.

The large number of two-quantum annihilation events which occurred even when positronium was formed gave rise to much of the background. Since at best only about ten per cent of the positrons formed positronium, well over ninety per cent of all gamma-rays emitted from the source region were either from two-photon annihilations or were the nuclear gamma-rays emitted simultaneously with the positrons. True double coincidences between any pair of counters could thus occur if one of those counters recorded an annihilation gamma-ray from a positron and the other recorded the nuclear ray emitted with that positron. A pulse tripping the third counter within the resolving time of the circuit would then give a triple coincidence. Random triples of this type would be unaffected by rotating one counter about the source. The rate of triple coincidences of this type will be $6DS\tau$, where D is the "true double" rate just discussed. It is given by $D = 2\ N\ \epsilon^{mc^2}\ \epsilon^{1.3\text{ Mev}}\ w^2$, with N the source strength, w the fraction of the total solid angle subtended by each counter, and the ϵ's the efficiencies of the counters at the energies indicated by superscripts. With the efficiency of a counter to 1300 kev radiation estimated as ten per cent with the bandwidths used, one finds a true double rate of $\sim$3.2 counts/sec., a triple rate of .6 counts/min.

Thus about 1.1 counts/min. out of a total of $\sim$1.4 counts/min. of background rate were contributed by triples which were completely independent of the coplanarity of the counters. Other random triples might occur if one of the two photons from an annihilation event were scattered in the material surrounding the source so that this ray entered one counter while the other annihilation ray registered in a second counter. The triple coincidence would be completed by a single ray tripping the third counter. When the symmetric arrangement is being used, rotating one counter out of the plane changes its angle with either of the other counters from 120° to 111°, resulting in a 12.5 per cent change in the triple rate from this type of background. Proper compensation for this effect was made in the measurements reported here. There is also a small amount of background from counter-to-counter scattering which is effectively eliminated by the lead snouts protecting each crystal.

If the three counters are not symmetrically arranged around the source, there may be a significant error in the measured background rate caused by the change in scattering angle when one counter is rotated out of the plane. Since it is difficult to know with certainty the contributions of the various types of background to the total random rate, background rates for non-symmetric arrangements are somewhat inaccurate, but the error has been kept less than the statistical error of all measurements. If positronium is being studied, background can always be measured by quenching with NO.

IX. Acknowledgments

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Mrs. Peggy Lees for drawing the figures.

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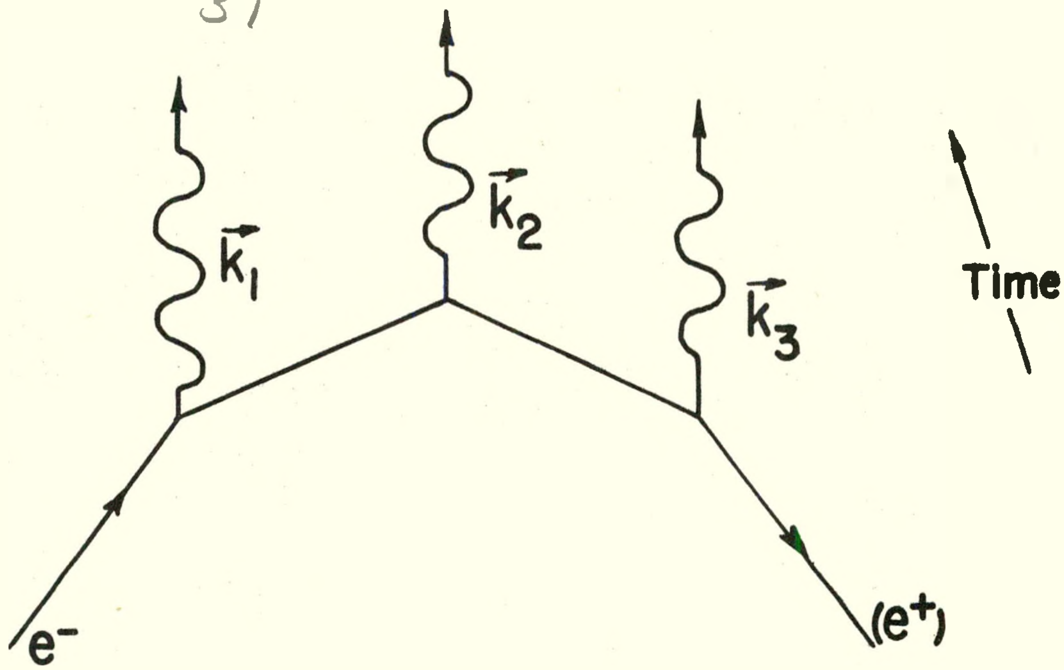


Fig. 1a. A Feynman Diagram for Three-Photon Annihilation

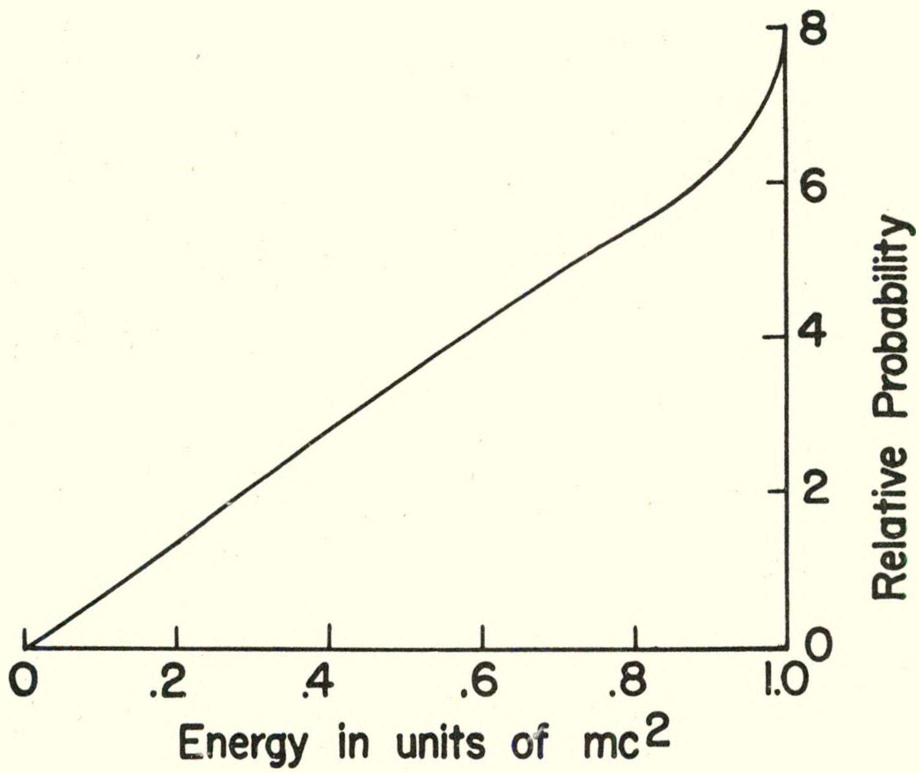


Fig. 1b. Energy Spectrum for One Photon from Three-Photon Annihilation

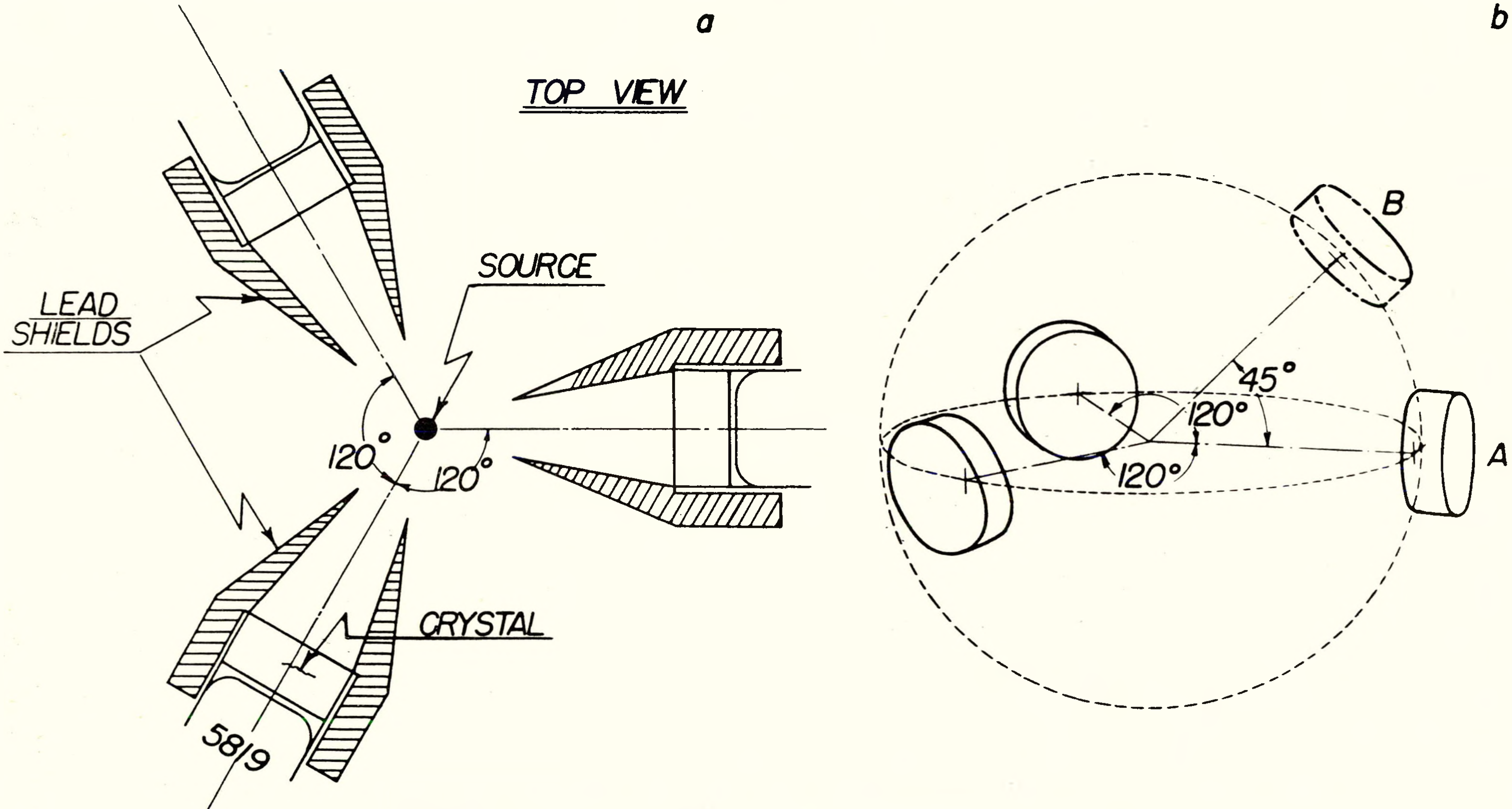


FIGURE 2: ARRANGEMENT OF THE THREE COUNTERS FOR THE DETECTION OF THREE-PHOTON ANNIHILATION.

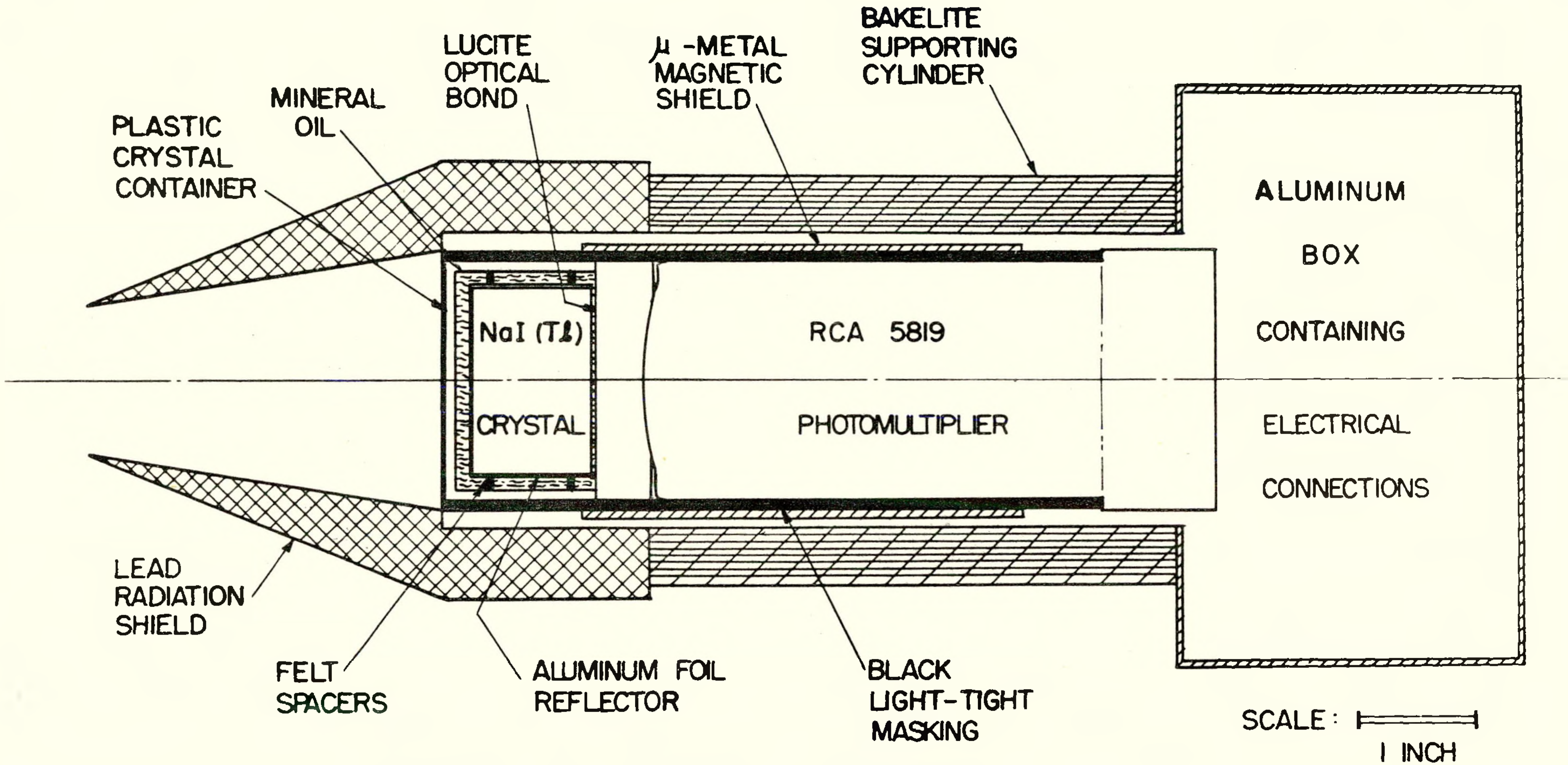


FIGURE 3: SCINTILLATION COUNTER

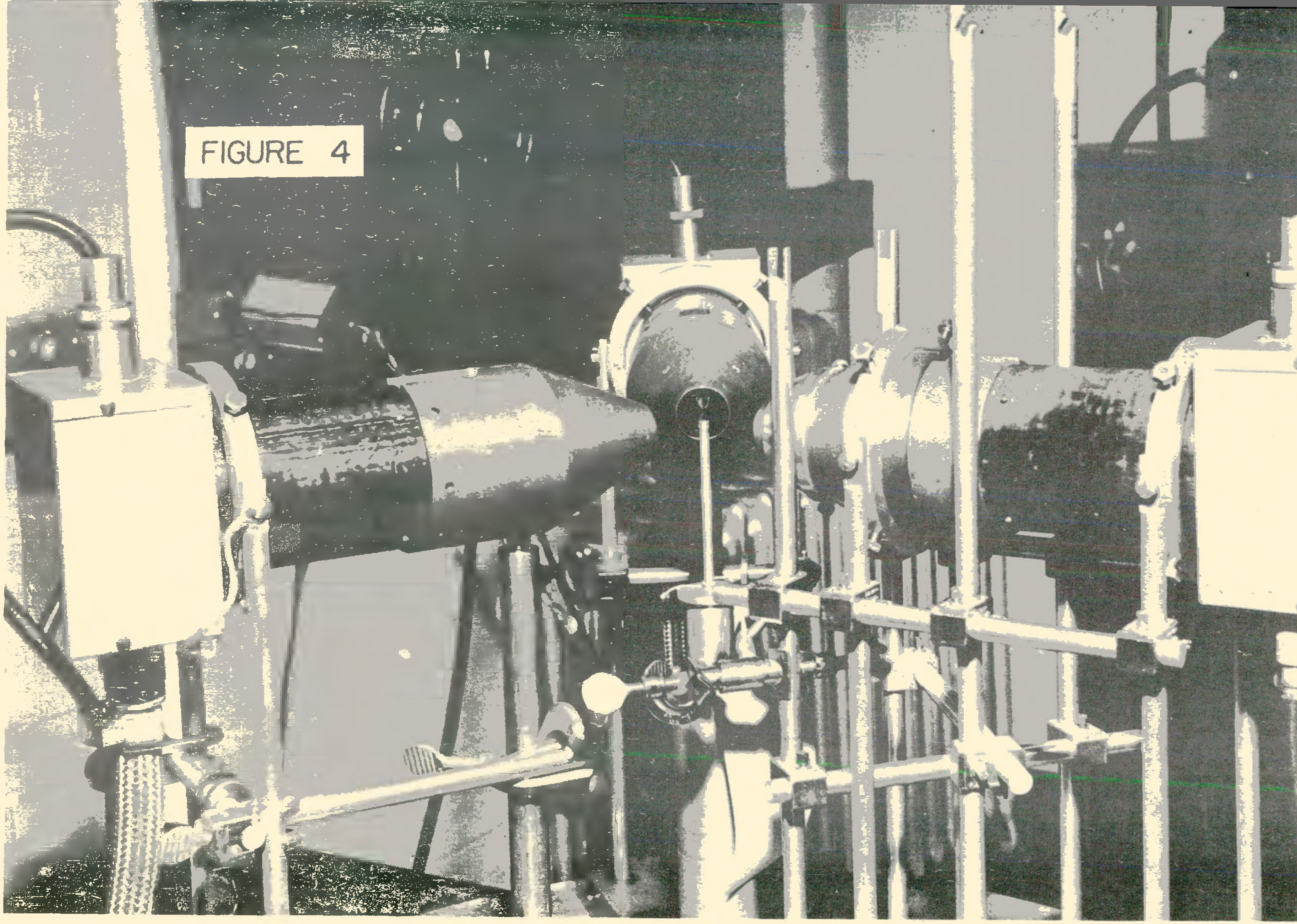


FIGURE 4

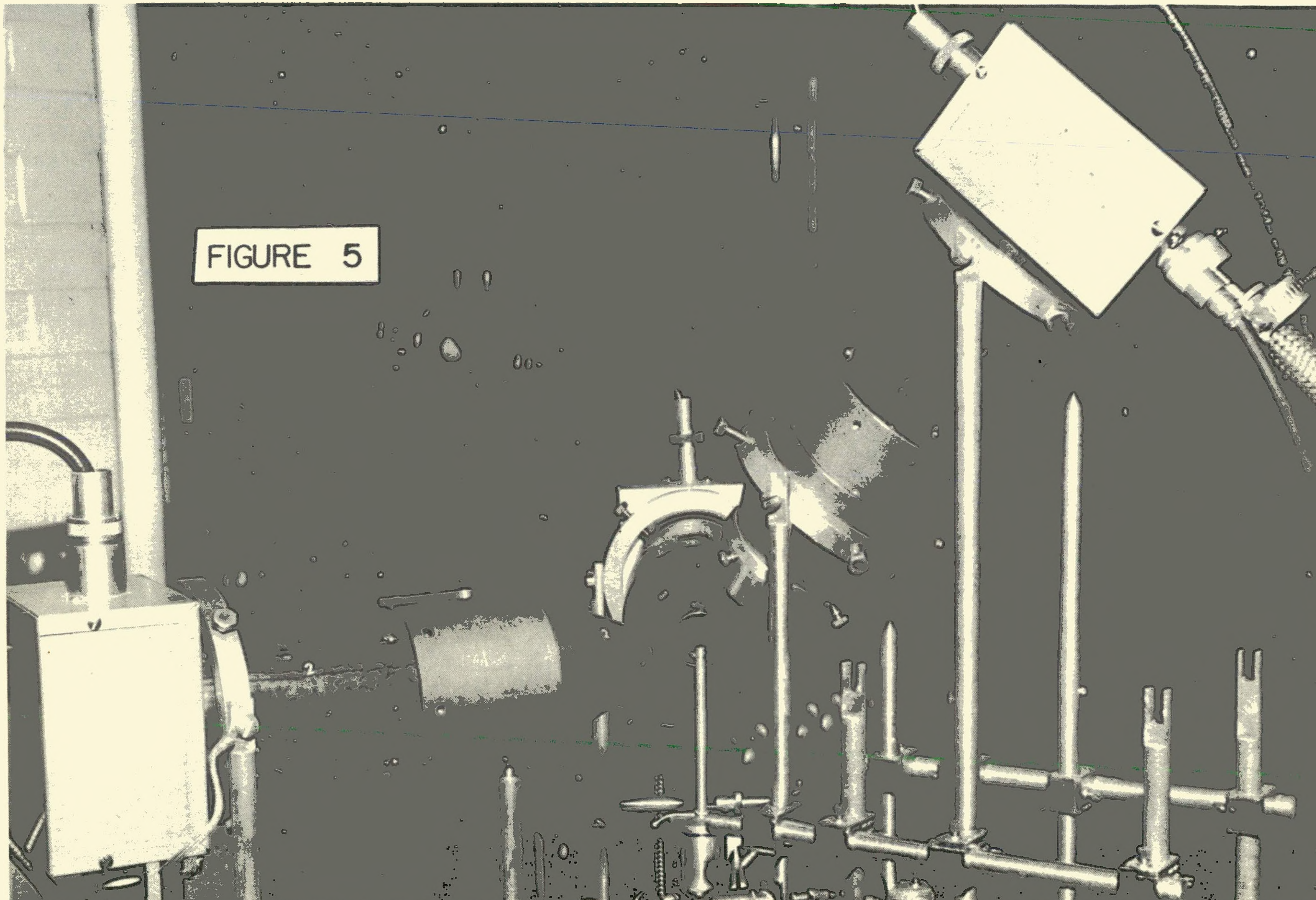


FIGURE 5

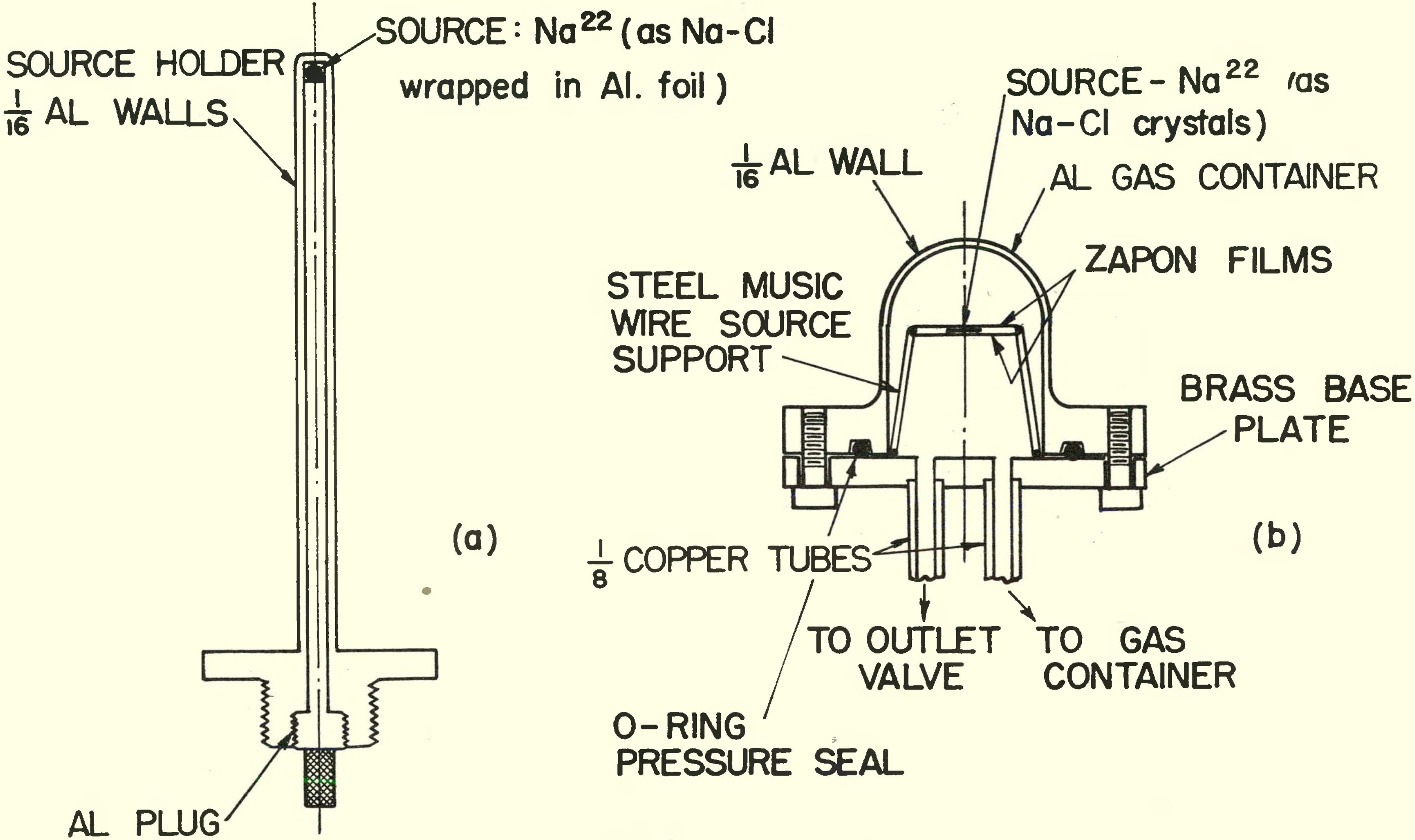


FIGURE 6 - SOURCE HOLDERS FOR STOPPING POSITRONS IN ALUMINUM (a) and GASES (b)

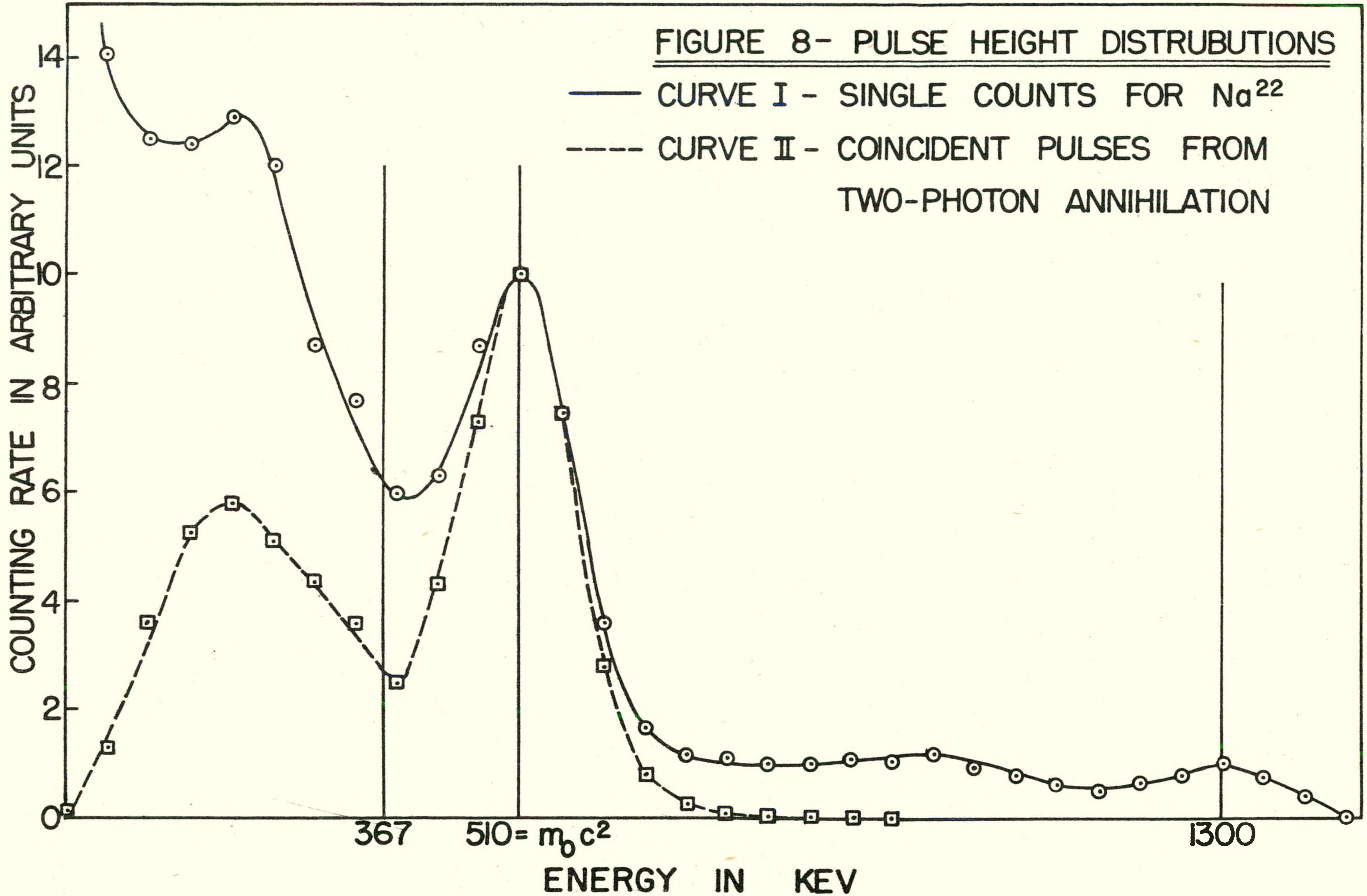


FIGURE 8- PULSE HEIGHT DISTRUBUTIONS
CURVE I - SINGLE COUNTS FOR Na22
CURVE II - COINCIDENT PULSES FROM TWO-PHOTON ANNIHILATION
COUNTING RATE IN ARBITRARY UNITS
0
2
4
6
8
10
12
14
367
510 = m0c2
1300
ENERGY IN KEV

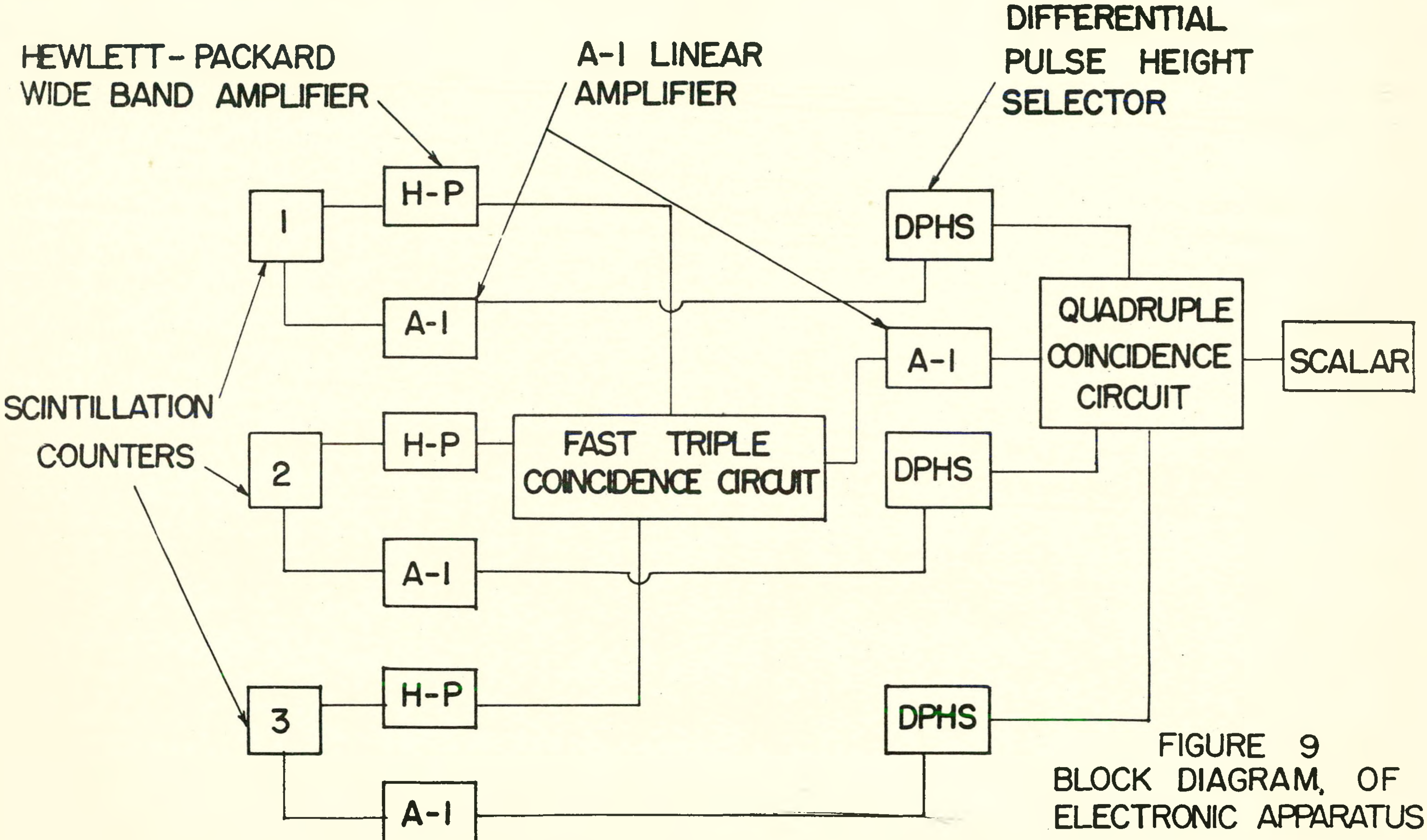


FIGURE 9
BLOCK DIAGRAM OF
ELECTRONIC APPARATUS

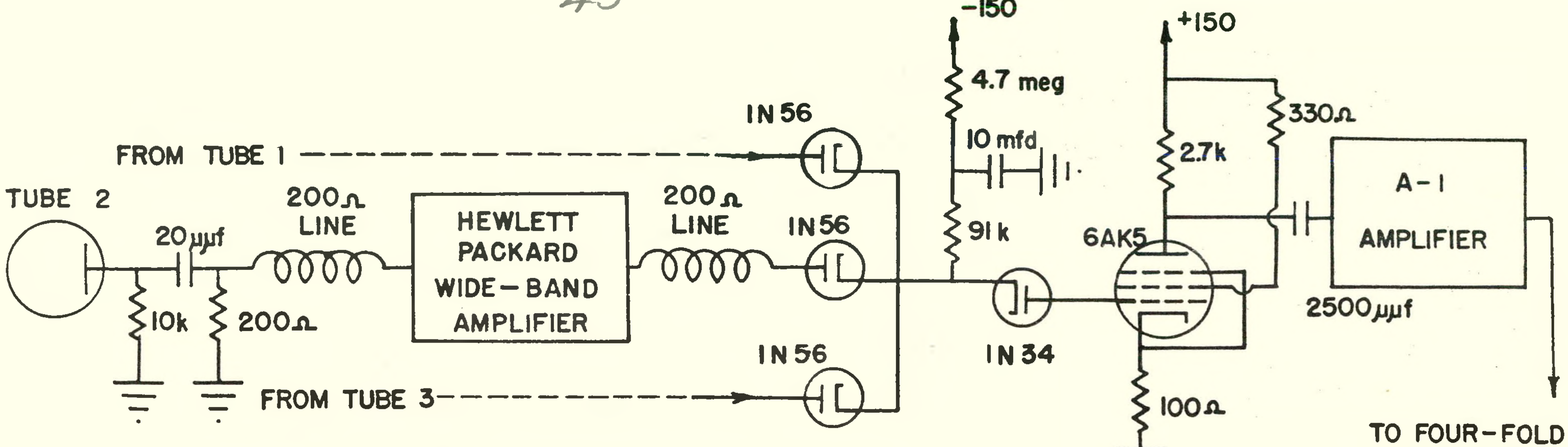


FIGURE 10a. FAST TRIPLE COINCIDENCE CIRCUIT

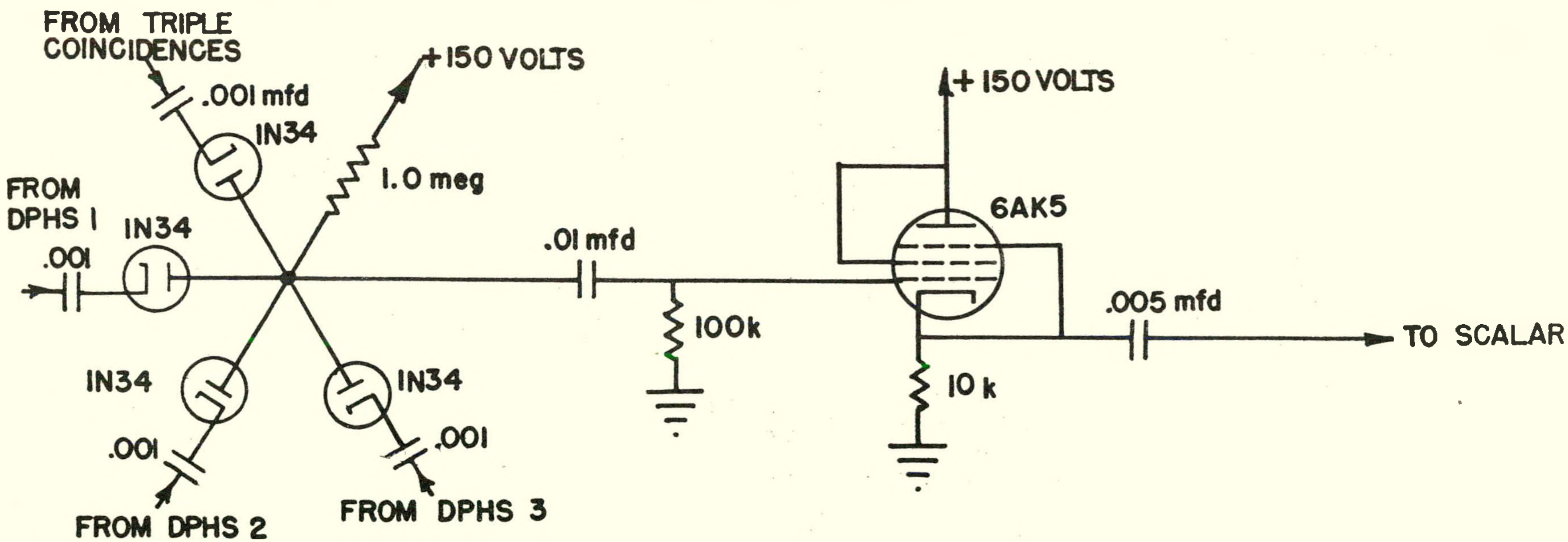


FIGURE 10b. QUADRUPLE COINCIDENCE CIRCUIT

FIGURE 11: 3-γ ANNIHILATION SPECTRUM IN SOLIDS

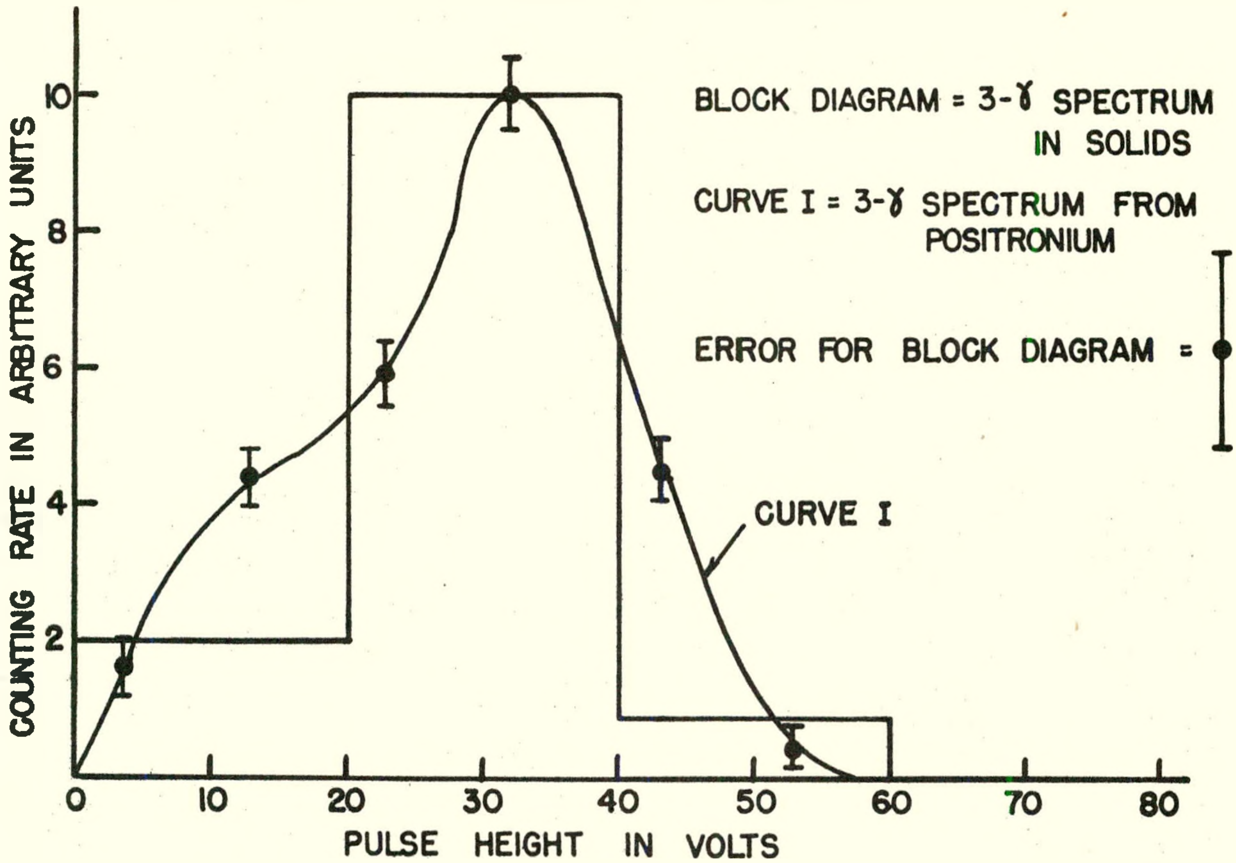


FIGURE 12: 3-γ ANNIHILATION RATE vs PRESSURE IN FREON - 12

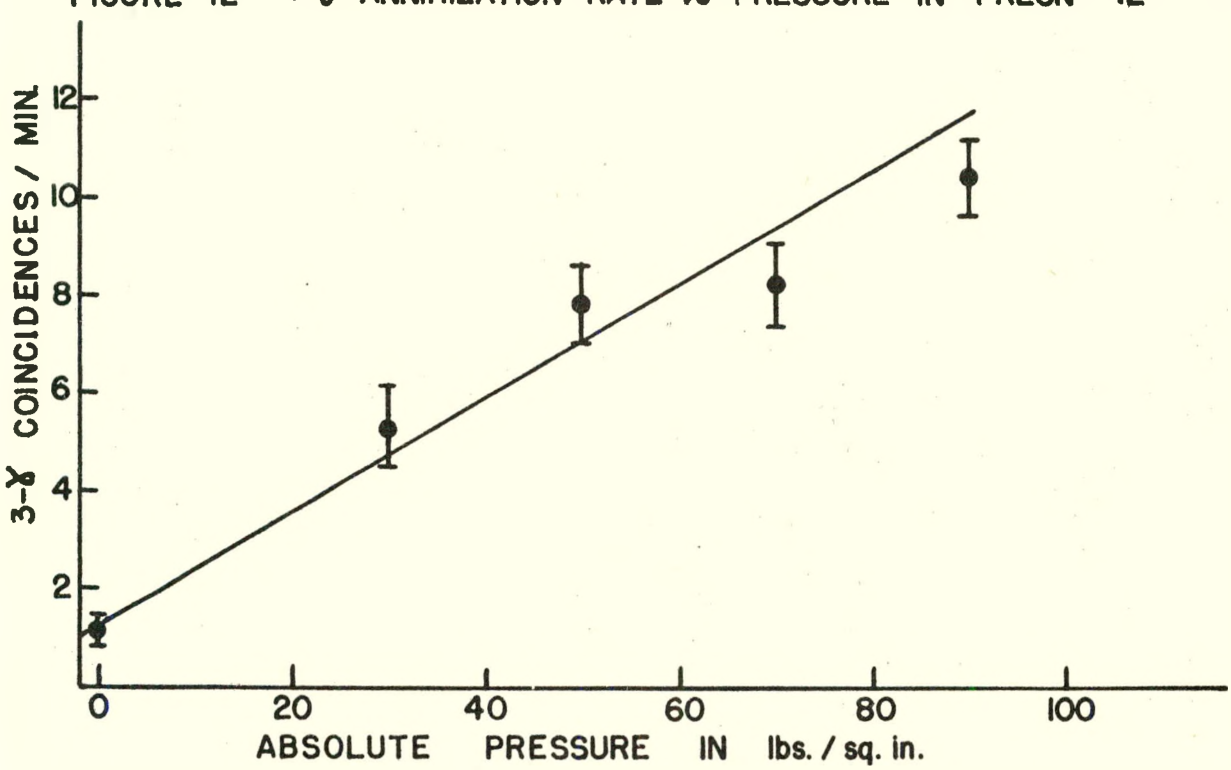


FIGURE 13: THREE-QUANTUM ANNIHILATION SPECTRUM FROM POSITRONIUM

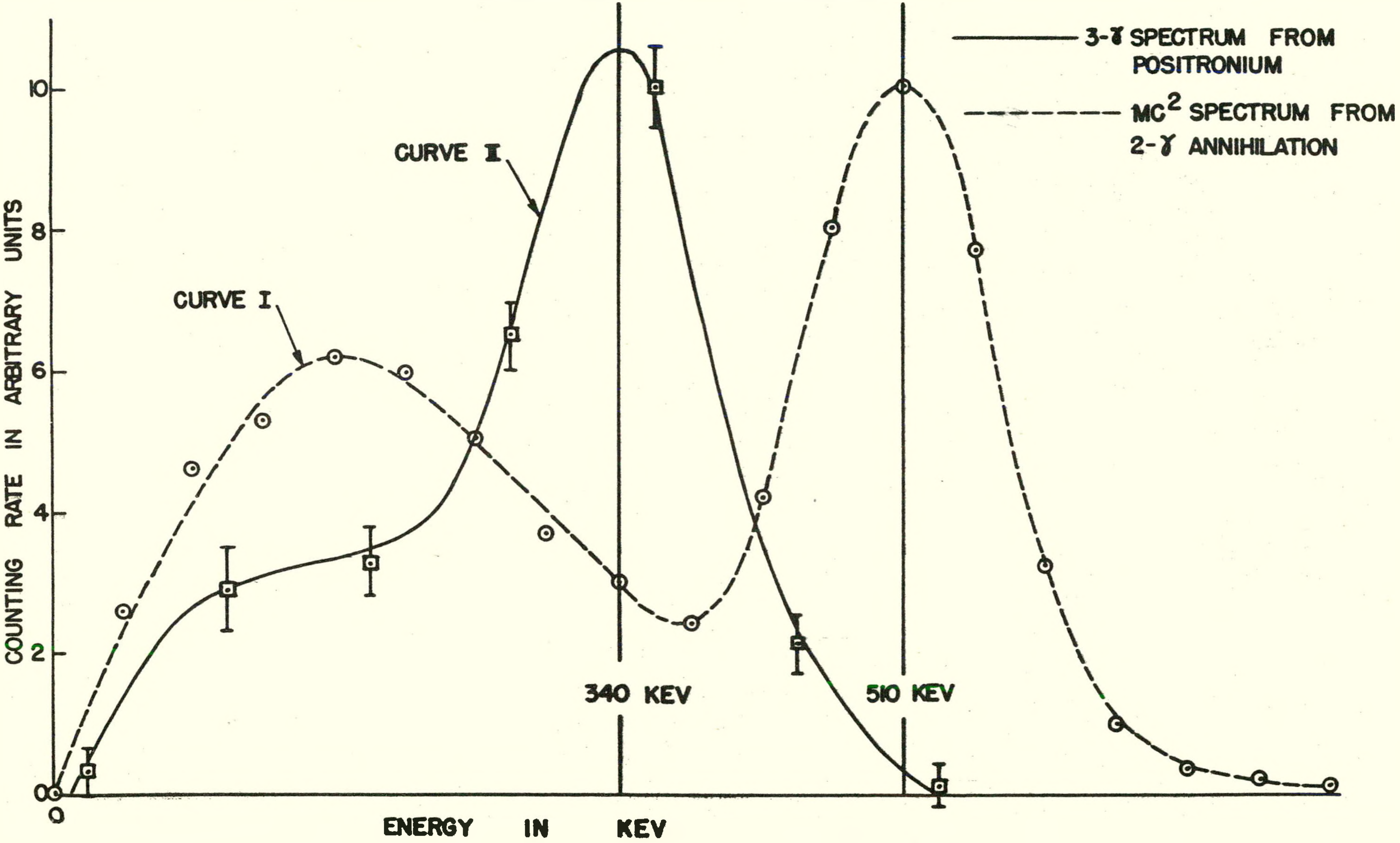
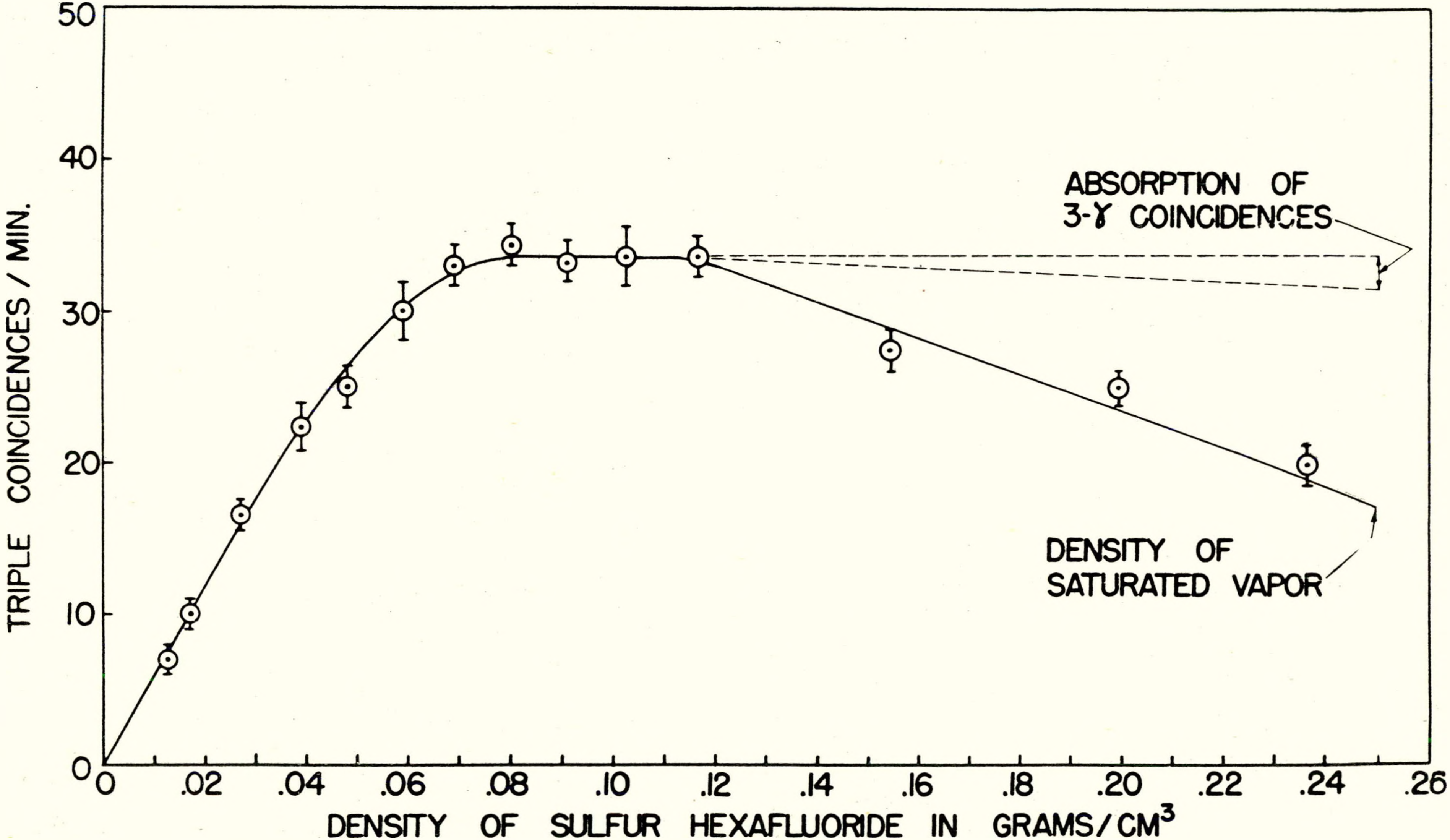


FIGURE 14: FORMATION OF POSITRONIUM VS. DENSITY IN SF_6 AT 26°C



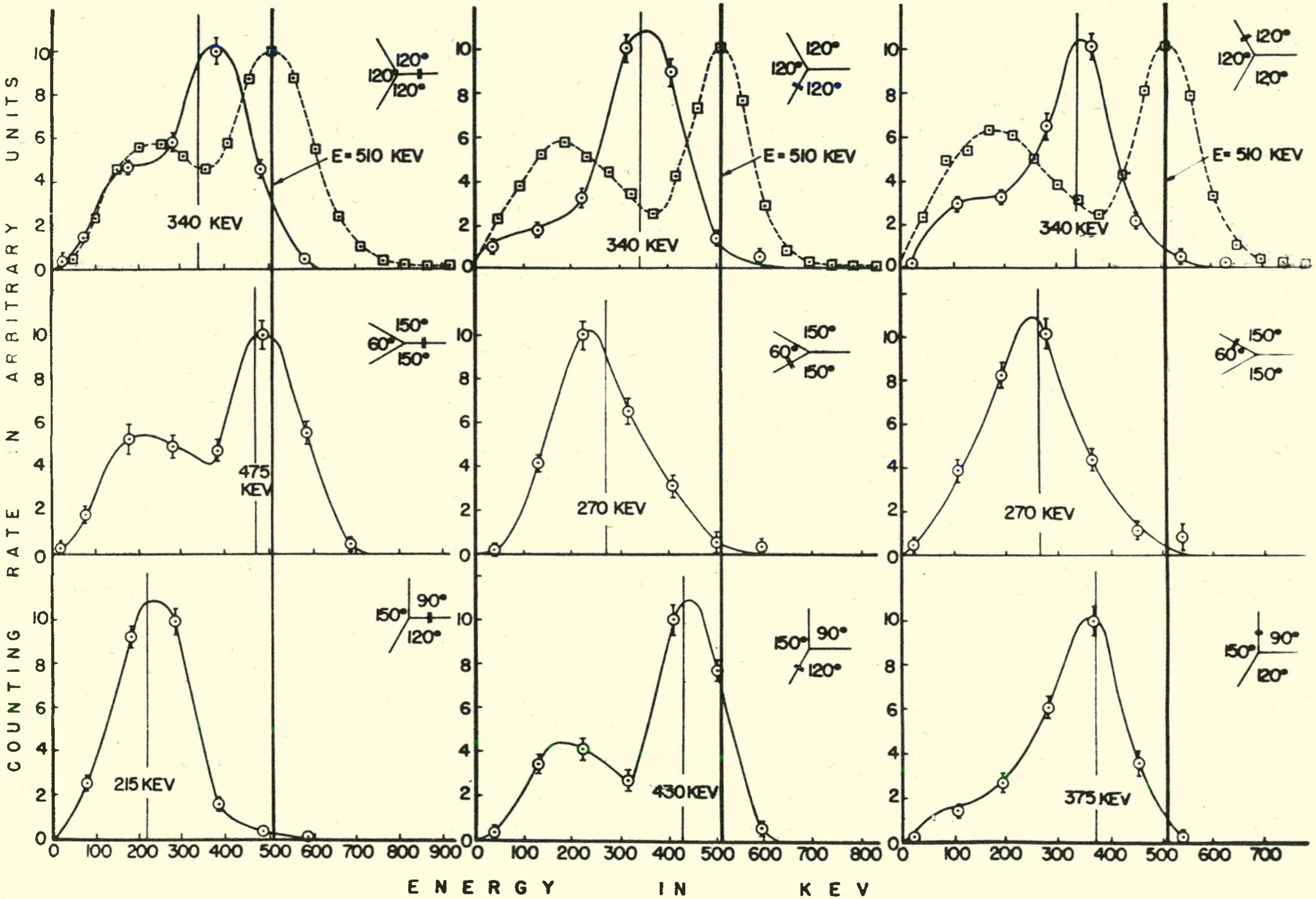


FIGURE 15: SPECTRA OF 3-γ COINCIDENCES FOR VARIOUS ARRANGEMENTS OF COUNTERS.