# ENERGY RESOLUTION OF $\gamma$ -RADIATION UP TO 18 MeV. BY SODIUM IODIDE SCINTILLATION COUNTERS

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#### Summary

The pulse-height distributions to be expected from scintillation counters using thallium-activated sodium iodide crystals have been investigated for  $\gamma$ -radiation of energies 6, 12, and 18 MeV. with two sizes of cylindrical crystal (2.5 cm. long by 2.5 cm. diameter and 7.5 cm. long by 4 cm. diameter). A Monte Carlo calculation of the partial shower process by which photons are absorbed in the crystals has given the probability distributions of ionization energy from homogeneous incident photons. The spreading of the distributions by statistical effects in the photomultiplier has been measured using light flashes of constant integrated intensity from a rotating mirror. The curves predicted by combining these two factors are compared with experimental pulse-height distributions for  $\gamma$ -radiation from proton bombardment of fluorine, boron, and lithium.

It is concluded that energy resolution above 10 MeV. is severely limited by broadening of peaks introduced by an appreciable and variable fraction of the energy of each incident photon escaping from the crystal as bremsstrahlung. This could be minimized by use of crystals with dimensions large compared with 8 cm., the maximum mean free path of bremsstrahlung photons in sodium iodide.

## I. INTRODUCTION

Hofstadter and McIntyre (1950a, 1950b) have shown that for energies of the order of 1 MeV. the pulse-height distributions obtained from a scintillation counter using a thallium-activated sodium iodide crystal can be interpreted in terms of the energy distributions of secondary electrons from the  $\gamma$ -capture processes of photoelectric effect, Compton scattering, and pair production. Maeder and Wintersteiger (1952a, 1952b) have shown the further effect of capture of degraded Compton-scattered quanta in the crystal.

When such a counter is used with  $\gamma$ -radiation above 6 MeV. it is found that the peaks in the pulse-height distributions are considerably broader than can be accounted for by the above-mentioned processes alone. The work described herein was undertaken to ascertain whether the additional broadening could be explained by a more complete investigation of the partial shower process which takes place when each high energy quantum is absorbed.

The use of a scintillation counter as a  $\gamma$ -ray spectrometer depends on the approximate proportionality which exists between the output pulse height and the energy of the incident photons. By breaking down the operation of the

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counter into consecutive stages it is possible to see where significant departures from strict proportionality can arise. Considering only homogeneous  $\gamma$ -quanta of energy E, possible sources of variation in output pulse height are:

- (i) The energy I given up by ionization in the crystal. We will let f(I,E)dI denote the probability that a quantum loses energy between I and I+dI.
- (ii) The number of light photons emitted per unit energy of ionization in the crystal. In sodium iodide this number is independent of I for electrons above 1 keV. (Taylor *et al.* 1951), and any variation in fluorescence efficiency through the crystal will here be neglected. Since, furthermore, the number is large, purely statistical variation is small, and so it will be assumed that the number of photons emitted is proportional to I.
- (iii) The fraction of the light photons which reach the photosensitive surface of the photomultiplier. The main variation in this arises from the dependence of the efficiency of light collection upon the position in the crystal from which the light emanates. This can be due to imperfections in the optical properties of the crystal (Garlick and Wright 1952) or to differences in the effective solid angle subtended by the photosurface. For the purposes of the present paper, this source of variation will also be neglected, and the number of photons reaching the photosensitive surface will be taken to be aI, where a is constant.
- (iv) The fraction of photons reaching the photosensitive surface which eject electrons, and
- (v) The multiplication of electrons in the multiplier.

The last two factors have been considered in detail by various workers (Seitz and Mueller 1950; Garlick and Wright 1952). We will let g(p,aI)dp denote the probability that aI photons reaching the photosensitive surface will give rise to an output pulse between p and p+dp in height.

The probability that a  $\gamma$ -quantum of energy E absorbed in the crystal gives rise to an output pulse from the photomultiplier between p and p+dp is now h(p,E)dp, where

In Section II of the present paper a method is described for finding the form of f(I,E) as a function of I. This was carried out for three values of E (6, 12, and 18 MeV.), and for two particular cylindrical sodium iodide crystals, one  $2\cdot 5$  cm. long by  $2\cdot 5$  cm. diameter, and the other  $7\cdot 5$  cm. long by 4 cm. diameter, referred to as the 1 in. crystal and the 3 in. crystal respectively.

The function g(p,aI), which includes the effect of statistical variation in the number of electrons ejected from the photosurface by equal flashes of light and in the multiplication of the tube, was assumed, for any value of aI, to be a Poisson distribution function of p with mean value proportional to aI. Section III

describes a measurement of the ratio of the standard deviation to the mean of the distribution as a function of the mean value.

In Section IV experimental pulse-height distributions are compared with the function h(p,E) derived from (1).

# II. CALCULATION OF f(I,E)

The function f(I,E) could in principle be calculated analytically from the theory of  $\gamma$ -ray absorption and of energy loss of electrons. However, the multiplicity of the processes involved renders this approach impracticable. Instead, it was decided to use the "Monte Carlo" technique, which Wilson (1952) has applied to the similar problem of shower production in lead.

The principle of the method consists of following through a large number of individual events, the course of each being determined by a statistical process according to predetermined probabilities. The crystal is divided into equal intervals of length, and an incoming  $\gamma$ -quantum is followed through each of these sections in turn. Sets of curves are drawn, representing the probabilities of the processes of  $\gamma$ -ray absorption, and also of bremsstrahlung from electrons, in each interval. These are shown in Figures 1 (a)-(d), the intervals being 0.5 cm. in sodium iodide. For a particular value of energy, the difference in ordinate between adjacent curves represents the probability of the particular process occurring per interval and giving products of the stated energy. For this purpose the energy is here "quantized" into increments of 0.5 MeV. The total ordinate of the figure represents unit probability per interval.

An incident  $\gamma$ -quantum of a particular energy E is followed using Figure 1 (a), important areas of which are shown magnified in Figures 1 (b) and 1 (c). Its energy fixes a value of the abscissa. A four-figure number is taken from a published list of random numbers (Tippett 1927; Kendall and Smith 1939). If the ordinate corresponding to that number falls within a set of curves, then the appropriate absorption process is said to have taken place in the first interval, giving products whose energies can be read from the curves. On the pair production curves, the numbers give the energy  $E_n$  of the negative electron, so that the positron has energy

$$E_{b} = E - E_{n} - 1$$
 MeV.

The Compton-scattering curves are marked with the energy of the electron  $E_n$ , so that the scattered photon has energy

$$E' = E - E_n$$

If the ordinate does not fall within a set of curves, then the quantum is said to have passed through the interval without absorption, and a further random number is taken to decide its fate in the next interval. The quantum is followed through in this manner until it either is absorbed or escapes from the crystal.

The positive and negative secondary electrons are treated using Figure 1 (d), where the curves represent differential radiation probability per 0.5 cm. in sodium iodide. An electron is followed through successive intervals, in each of which its energy is reduced by ionization, and further energy may be radiated as bremsstrahlung, until either its energy is reduced to zero or it escapes from the crystal.

The bremsstrahlung photons are then followed using Figure 1 (a), as also are positron annihilation quanta, until the shower is complete. The total energy I lost by electrons in ionization is then noted. This process is repeated for a large number of incident quanta of the same energy E, and the resultant histogram of the number of events giving ionization energy I against I is then an estimate of the form of f(I,E) for the particular value of E in that crystal.



Fig. 1.-Monte Carlo probability curves.

- (a) Probability curves for photon absorption.
- (b) Enlargement of Compton section in (a).
- (c) Enlargement of pair production section in (a).
- (d) Probability curves for radiation from electrons.

Wilson carried out the random selection of ordinate by spinning a cylinder, around which the curves were wrapped. The ordinate was determined by a fixed pointer when the cylinder came to rest. Since such a cylinder would need to be spun tens of thousands of times during this investigation, a long time would be spent waiting for the cylinder to stop. Also, precautions would be necessary against bias in the cylinder. It was decided that the use of random number tables to give the ordinate directly would be more rapid and more reliable.

Figures 1 (a)-(d) were derived mainly from data given by Rossi and Greisen (1941) and Davisson and Evans (1952).

The curve for the photoelectric absorption probability in Figure 1 (a) was derived from Figures 13–17 in the paper by Davisson and Evans.

The Compton-scattering curves in Figure 1 (b) were derived from the Klein-Nishina formula (Heitler 1936). For incident photons with energy E, the differential cross section per electron for scattered quanta with energy E' is

$$\frac{\mathrm{d}\sigma}{\mathrm{d}E'} = -\frac{2\pi\mu g}{E^2} \Big\{ \frac{\mu^2}{E'^2} + \frac{1}{E'} \Big( E - 2\mu - \frac{2\mu^2}{E} \Big) + \frac{\mu}{E} \Big( 2 + \frac{\mu}{E} \Big) + \frac{E'}{E} \Big\}$$

over the range

$$E\left(1+\frac{2E}{\mu}
ight)^{-1}\leqslant E'\leqslant E,$$

where  $\mu$  is the rest energy of an electron and g is a constant equal to  $3 \cdot 92 \times 10^{-26}$  cm.<sup>2</sup>. When this was plotted as a function of E' for various values of E, the areas in each 0.5 MeV. range gave the spacing of the curves.

The curves of Figure 24 in the paper by Davisson and Evans were replotted as a function of negative electron energy  $E_n$ , and the areas under them measured in each 0.5 MeV. interval. These areas gave the spacing of the pair production curves in Figure 1 (c).

The spacing of the curves in Figure 1 (d) was derived from Figure 7 in the paper by Rossi and Greisen. This gives the differential radiation probability for fast electrons per radiation length in lead, and, since this quantity varies little between the elements, it was taken to apply to sodium iodide directly. A radiation length in sodium iodide is almost exactly 2.5 cm. and so 0.5 cm., the interval of length used, is one-fifth of a radiation length.

In the present paper it is assumed that the  $\gamma$ -rays enter the crystal parallel to the axis and that all primary processes take place in the forward direction, i.e. all the products proceed forward in the same line. For pair production by 6 MeV. photons, the solid angle into which the positron and electron are emitted is of the order of 0.1 sterad.; as the energy increases, the solid angle decreases. For the photoelectric effect, which is of importance only at energies of 0.5 and 1 MeV., the angle which the photoelectrons make with the direction of the  $\gamma$ -rays is of the order of 10°. For Compton scattering, one extreme case is that which occurs when the electron receives the maximum energy possible; it then moves off in the forward direction, while the scattered photon, whose energy is approximately 0.25 MeV., moves backward. At the other extreme, the photon moves on practically undeflected, while a low energy electron is scattered at 90°. Thus, when products of Compton encounters move in directions far removed from that of the incident photon, their energies are small and they are soon captured. In all these cases there is little error introduced if it is assumed that all the products move off in the forward direction.

The electrons do not continue to move in a straight line but are multiply scattered until eventually their motion becomes completely random. Since it would be very difficult to treat this accurately, it was decided to adopt the approximation found satisfactory by Wilson (1951, 1952). He assumed that an electron continues to move in a straight line until its energy falls to some critical

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energy, after which it remains at the same point in the crystal executing a "tight random motion" until its energy is reduced to zero. Using the data given by Wilson, a curve was drawn giving the relation between initial energy and critical energy for sodium iodide.

Since a bremsstrahlung photon emitted by an electron moves in nearly the same direction as the electron, the radiation from electrons above their critical energy travels longitudinally through the crystal, whilst that emitted by randomly moving electrons is isotropic. The probability that this isotropic radiation is captured in the crystal was calculated for the case when it originates from the centre. The values obtained for various photon energies were assumed to hold even when the radiation does not come from the centre of the crystal, thereby underestimating somewhat the amount of bremsstrahlung which escapes. The further assumption was made, that, once the isotropic radiation is captured, its energy is completely dissipated as ionization within the crystal, thus further underestimating the energy which escapes.

A disadvantage of the technique just described is that any detail is lost which is as fine as or finer than the increments into which energy is quantized. Using increments of 0.5 MeV., this is particularly serious when the incident energy is only 6 MeV., and yet it would not be practicable to carry out the whole calculation with smaller increments. The difficulty can be overcome by breaking down the situation into its component processes, ascertaining in which of these the fine detail is introduced and using smaller increments in those sections of the calculation alone. In the present problem, primary photoelectric effect and Compton scattering do not introduce any fine detail, and neither does primary pair production up to the annihilation of the positron. It is the two 0.5 MeV. annihilation quanta, either of which may or may not be absorbed, which distribute into three sharp peaks 0.5 MeV. apart the pulses from those primary pairs which otherwise give all their energy to the crystal. For this reason, it was decided to quantize energy in 0.1 MeV. increments when absorption of annihilation quanta was considered.

Therefore, in the procedure actually adopted for finding the form of f(I,E), the Monte Carlo technique was not used for the primary processes. Instead, the primary processes giving products in each energy range were assumed to occur in each section of the crystal with frequencies proportional to the calculated probabilities. The life histories of the products for a number of examples in each case were then followed through by the Monte Carlo method, and the results combined appropriately.

The length of the larger crystal was divided into 20 sections, the first 10 each 0.25 cm., and the second 10 each 0.5 cm. long. For each of the three energies, 6, 12, and 18 MeV., 200 photons were considered, each of which produces a pair in the first section of the crystal. From Figure 1 (c) the number of pairs was found in which the electron has zero energy, the number in which the electron has 0.5 MeV., etc. The development of each shower was recorded in the manner shown by the typical diagrams in Figure 2. Taking each pair in turn, the motion first of the electron and then of the positron was followed through the crystal. From the data of Heitler (1936) and of Halpern and Hall

(1948), the ionization loss was found to be close to 2 MeV./cm. in sodium iodide and to be practically independent of electron energy above 1 MeV. Therefore 1 MeV. was subtracted from the electron energy on crossing each 0.5 cm. interval. Whether or not an electron also loses energy by radiation in an interval is determined by a random number from the tables. With the electron energy as the abscissa and the random number as the ordinate, a point is determined on the figure. If it lies above the topmost curve, no energy is lost by radiation. Otherwise, the electron loses the energy associated with the region in which the point lies, and a photon of the same energy appears in that section of the crystal.



Fig. 2.—Typical 18 MeV. showers commencing in the first interval of the 1 in. crystal.

This process is continued until the energy of the electron falls below the critical energy. Then the processes occurring in each interval of length are still determined as above until the electron energy falls to zero, the only difference being that the electron moves no further forward in the crystal. The motion of the positron is treated similarly.

There exist now on the diagram two types of photons : those radiated by an electron moving in a straight line and those radiated during its random motion. The former are followed through each remaining interval in turn. In each case a new random number determines whether or not the photon is absorbed, and if so, in what way. The photon may continue unabsorbed and eventually escape from the crystal, or it may form a pair, be Compton scattered, or eject a photoelectron, in which cases all the products are followed until they are completely absorbed or escape. Those photons radiated during the random motion of the electron require only one random number to determine whether they are absorbed or not, as mentioned above. If a positron-electron pair is formed as a secondary or tertiary process, the quanta resulting from the annihilation of the positron are treated in a similar way to isotropically emitted bremsstrahlung. We now have a set of diagrams representing the consequences. of a number of photons producing pairs in the first section of the crystal.

Considering just the first 10 intervals, i.e. the 1 in. crystal, it can be seen that by removing the last section and everything that occurs in it, we have the consequences of a number of photons producing pairs in the second interval of the crystal, and by removing the last two sections, of those forming pairs in the third interval, and so on. Thus, to obtain a number against energy distribution for the number of photons forming a pair anywhere within the crystal against the energy I lost by them in ionization in the crystal, we first obtain separate number-energy distributions for the energy lost by ionization for initial pair formation in each section. These relations are then combined by adding them



Fig. 3.—Distributions of ionization energy from primary pair production. (a) The 1 in. crystal. (b) The 3 in. crystal.

in the ratio of the number of photons absorbed by pair production in each section, as determined by values of the absorption coefficient. Except for some modification arising from the two sizes of interval, the same treatment was applied for the 3 in. crystal. Thus we obtain the distributions of energy lost by ionization in the crystal by quanta initially absorbed by pair production, except for the annihilation radiation from the positrons which has yet to be considered. These are shown as the dashed curves in Figure 3. The full curves exclude the contribution of those showers resulting in the escape of the positron.

The probability of annihilation of a positron before coming to rest is small (Heitler 1936), and it is here assumed that the two annihilation quanta are emitted in opposite directions along a line randomly oriented in space. A numerical calculation was undertaken to determine the energy loss to the crystal of such pairs of quanta with points of origin spread uniformly through the volume of the crystal. The annihilation quanta were assumed to be of 0.5 MeV., and energy was considered in discrete steps of 0.1 MeV. The energy loss

distributions for single quanta obtained at one stage of the calculation were consistent with those found by Maeder and Wintersteiger (1952a, 1952b). The histograms obtained for the energy loss probability distributions for pairs of annihilation quanta are shown in Figure 4.

The distributions in Figure 3 were then corrected for additional absorption of annihilation energy by spreading out the full curves numerically according to Figure 4 and then replacing the area between the curves, thus completing the study of the energy loss of those incident quanta which are absorbed by the pair production process.



Fig. 4.—Distributions of ionization energy from annihilation quanta. (a) The 1 in. crystal. (b) The 3 in. crystal.

Since at the energies considered primary photoelectric absorption is rare (with absorption coefficient about 1 per cent. of that of either of the other processes at 6 MeV.), it is here neglected, and it remains now only to consider those quanta initially absorbed by the Compton process.

As in the case of pair production, 200 incident quanta were considered to undergo Compton scattering, the relative numbers giving products of each energy being proportional to the calculated probabilities. The products were then followed by the Monte Carlo method until each shower was complete, in the manner previously outlined. The resultant probability distributions for ionization loss from initial Compton scattering are shown in Figure 5.

The complete distributions were then obtained by adding the pair and Compton distributions in the ratio of their relative probabilities. The final curves giving the form of f(I, E) for all the cases considered are shown in Figure 6.

In some of the distributions there appears a spurious peak in the low energy region. This is a result of the particular assumption made as to multiple scattering. Consider a number of pairs with the same total energy starting from the same point in the crystal. These will all move forward approximately



Fig. 5.—Distributions of ionization energy from primary Compton scattering. (a) The 1 in. crystal. (b) The 3 in. crystal.



Fig. 6.—Distributions of ionization energy from all processes. (Final forms of f(I,E).) (a) The 1 in. crystal. (b) The 3 in. crystal.

the same distance losing a certain amount of energy by ionization, whereafter they virtually come to rest and the remainder of their energy is dissipated in the same part of the crystal. Thus, if only the contribution of electrons which finally escape from the crystal is considered, it will be seen from the above that,

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because of the particular assumption made, the number as a function of energy lost will be almost constant up to a certain energy, whence it will drop quickly to zero. Now in reality this would not be so, because the distribution from this cause begins to fall gradually from the low energies and extends further on the high energy side. In constructing the curves in Figure 10 these spurious peaks were smoothed over in accordance with this explanation.

### III. DETERMINATION OF g(p,aI)

The EMI type 5311 photomultiplier tube used in this experiment has a photocathode sensitivity of 40  $\mu$ A./lumen and a gain of 10<sup>7</sup> at 160 V. per stage (manufacturer's figures). In order to measure directly the broadening of the pulse-height distribution which is introduced by statistical effects in the tube, it was necessary to expose it to flashes of light of constant integrated intensity and to measure the spread in height of the resulting output pulses.

The statistical spread is a function not only of the integrated light intensity but also of the wavelength of the light and the manner in which the intensity varies with time. Thus, ideally, it would be necessary to measure the spread for light flashes of colour and shape identical with those from the phosphor with which the photomultiplier is to be used. However, by inserting at the collecting electrode an integrating time constant long compared with both the test pulses and the scintillation pulses (and ensuring that the multiplier is never overloaded), the effect of differences in pulse characteristics can be minimized so that only the integrated light intensity remains important. In the present work an integrating time constant of 4  $\mu$ sec. was used, which is long compared with the light flashes from thallium-activated sodium iodide, which have a decay time of  $0.25 \mu$ sec.

The method used to produce short flashes of light was to allow a beam of light reflected from a rotating mirror to pass across the photocathode of the photomultiplier. If the mirror rotates at f rev./sec., the angular velocity of the reflected beam is  $4\pi f$  rad./sec., and with the photocathode l cm. from the mirror the reflected beam passes across it at a linear speed of  $4\pi f l$  cm./sec.

The experimental arrangement, shown in Figure 7, was set up in a service tunnel beneath one of the University buildings. A 35 mm. slide projector P, with an object lens of 16 in. focal length, was used to provide the optical system, the slit being cut in a piece of fogged film. It was found necessary to feed a direct current to the projector lamp to avoid A.C. ripple in the light intensity. The rotating mirror, RM, was mounted on a spindle which was inserted in the chuck of a high speed drill, D. The back of the mirror, as well as the mounting, was painted black, leaving a reflecting area 2 cm. by 1.5 cm. The reflected beam could pass through a hole in a black curtain, C, into the otherwise dark section of the tunnel, T. At the other end of the tunnel was a fixed plane mirror, FM, to return the beam to the photomultiplier, PM, which was situated near the black curtain, so that the tunnel length was used twice. The path length traversed by the reflected beam from the rotating mirror to the photocathode was 107 m.

The rotational speed of the mirror was found with a "Stroboflash" unit to be 11,100 r.p.m. after warming up, with short-term variations of less than 0.2 per cent., so that the linear speed of the slit image over the photocathode was 24.2 cm./µsec. Two slits were used, giving images 5 cm. and 10 cm. wide at the photocathode, which is 2.5 cm. in diameter. Thus the flashes of light gave pulses of total length 0.3 and 0.5 µsec. Both these times are short compared with the integrating time constant.

The output pulses from the photomultiplier were fed through a preamplifier with a gain of -1 to the  $A_1$  amplifier of a Cossor model 1035 oscilloscope and thence to a single channel pulse-height analyser built to the design of Farley (1953) and to a type 1009A discriminator unit used as monitor. Pulse heights at the analyser were between 15 V. and 45 V., and the channel width was about 0.8 V. No correction was made for the broadening introduced by this channel width.



Fig. 7.-Experimental arrangement for measurement of statistical spread.

A number of runs back and forth over the pulse-height distribution was made for each of several lamp voltages with both slits, using total multiplier voltages of 930, 1040, and 1150 V. The gain of the amplifier was set in each case to give output pulse heights within the range of the analyser. The two runs shown by the points in Figure 8 illustrate the spread obtained in the two extreme cases, for the least and for the greatest integrated light intensity per flash. It can be seen that these distributions are consistent in shape with the Poisson distribution (curves), as were all others.

The mean value and standard deviation of the pulse-height distribution were calculated in each case, and the value of (standard deviation/mean) was plotted against  $\sqrt{(\text{multiplier gain/pulse height at collector)}}$ , the result being shown in Figure 9. The multiplier gain was measured relative to that at 930 V., and the pulse height at the collector was found by dividing the mean of the pulse-height distribution in volts by the measured amplifier gain. The fact that the points are well fitted by a straight line through the origin indicates that the fractional spread of the distributions is inversely proportional to the square root of the number of electrons entering the multiplier, a characteristic feature of the Poisson distribution, and therefore that sources of spread in pulse heights from uniform light flashes other than statistical variations are unimportant. Furthermore, there is no great dependence of the spread on multiplier voltage in the range used.



Fig. 8.—Pulse-height distributions from light flashes of constant integrated intensity.



Fig. 9.-Results of determinations of statistical spread.

The slope of the line drawn through the points in Figure 9 was determined by the least mean square method. Taking the case of a 1 V. pulse at the collector (which in our apparatus had a measured capacity to earth of  $20 \cdot 0 \ \mu\mu$ F.) with 1150 V. on the multiplier, this value of the slope gives the ratio of the standard deviation to the mean to be 0.047, corresponding to an effective number of electrons subject to statistical variation of 453.

#### IV. COMPARISON WITH EXPERIMENT

The thallium-activated sodium iodide crystals used in this experiment were obtained from the Harshaw Chemical Company, Cleveland, Ohio. They were polished with successively finer grades of abrasive paper, and finally a cloth, while being kept wet with "Nujol".

For mounting on the photomultiplier tube, the axis of which was vertical, the smaller crystal (2.5 cm. long by 2.5 cm. diameter), with aluminium foil wrapped round its curved and upper flat surfaces, was immersed in "Nujol" in a small glass beaker which was stood directly on the photocathode, with a few drops of "Nujol" added to couple optically the two glass surfaces. Thus the small crystal was mounted almost immediately on the photocathode.

The larger crystal (7.5 cm. long by 3.8 cm. diameter) was a composite one consisting of three separate crystals, each 2.5 cm. long by 3.8 cm. diameter, placed together on the one axis. As it was too large to mount directly on the photocathode, it was sealed into a "Perspex" block, the narrow space between crystal and "Perspex" being filled completely with "Nujol". The block was tapered down to a flat circular face 1 in. in diameter, which was placed against the photocathode with a layer of glucose for optical coupling, so that the centre of the crystal lay on the axis of the photomultiplier about 4 in. from the photocathode, and the crystal axis made a right angle with the photomultiplier axis. The external surface of the "Perspex" had all sharp corners bevelled and, except for the area facing the photocathode, was covered by a layer of magnesium oxide about 1 mm. thick deposited by holding it in the smoke of burning magnesium ribbon. Mechanical support was achieved by a light frame of  $\frac{1}{2}$  in. brass rod, into which the "Perspex" block was inserted before the deposition of the magnesium oxide; the frame was then bolted to the steel plate on which the photomultiplier was mounted.

Stray light was excluded from the system by a box of thin brass sheet, which enclosed crystal, photomultiplier, high tension bleeder, and preamplifier and was screwed down to the steel plate and sealed with a rubber gasket. The vacuum tubes of the preamplifier were wrapped in black insulating tape to block light from their heaters.

The type 5311 photomultiplier, preamplifier, amplifier, and pulse-height analyser were those described in Section III. The range of the analyser was increased to 150 V. by the addition of further variable bias to the threshold amplifier, and the channel width used was 2 V.

The  $\gamma$ -radiation was obtained by bombarding thick fluorine, boron, and lithium targets with protons from a 700 kV. electrostatic generator. The targets were deposited on copper backings by evaporating lead fluoride, by pressing amorphous boron, and by evaporating lithium hydroxide, respectively. The bombarding energy used on all three targets was 510 keV. The  $\gamma$ -radiation entered the 1 in. crystal 9 in. from the target, and the 3 in. crystal 5 in. from the target.

The experimental pulse-height distributions so obtained are shown as histograms in Figure 10. Because of the greater distance of the 3 in. crystal from the photocathode and the intervening thickness of "Perspex", the light collection efficiency from it was much less than from the 1 in. crystal. Using the same amplifier gain throughout, the pulses from both crystals were brought within the range of the analyser by using a multiplier voltage of 766 V. with the 1 in. crystal and 1040 V. with the 3 in. one.

The distributions for the complex spectra resulting from the three nuclear reactions were obtained from Figure 6 as follows :

 $F^{19}(p,\alpha\gamma)$ . The predominant  $\gamma$ -ray energy is 6.1 MeV., with 4 per cent. of 7.1 MeV. (Hornyak *et al.* 1950). The curve for 7.1 MeV. was assumed to



Fig. 10.—Results for nuclear spectra.

Histograms—experimental pulse-height distributions; smooth curves—predicted forms of h(p,E). (a) The 1 in. crystal. (b) The 3 in. crystal.

have the same shape as the 6 MeV., and was drawn so that the area under it was 4 per cent. of the area under the distribution for  $6 \cdot 1$  MeV., and the two were then added to give the distribution for this reaction.

 $\operatorname{Li}^7(p,\gamma)$ . The  $\gamma$ -ray energies are  $17 \cdot 6$  and  $14 \cdot 8$  MeV. The distribution for  $14 \cdot 8$  MeV. was found by interpolation between those for 12 and 18 MeV. According to Walker and McDaniel (1948) the intensities of the two components are in the ratio 2:1. The ratio in which the components are absorbed differs slightly from this because of the difference in absorption coefficients for the two components in sodium iodide. No allowance was made for the natural spread in energy of the  $14 \cdot 8$  MeV. component.

B<sup>11</sup> $(p,\gamma)$ . The relative intensities of the three components at 4.4, 11.8, and 16.3 MeV. are 4:4:1 (Walker 1950). The 4.4 MeV. distribution was

taken to be that for 6 MeV. displaced along the energy axis, and the  $16 \cdot 3$  MeV. estimated by interpolation between 12 and 18 MeV. The components were then added in the ratios given above, slightly modified to allow for the different absorption coefficients.

From the positions of the peak in the fluorine distribution it was found from the results of Section III that the effective number of electrons subject to statistical variation for each MeV. of ionization in the crystal was 335 with the 1 in. and 48 with the 3 in. crystal. Since some of the variation is introduced in the multiplier stages, these correspond to somewhat higher numbers of electrons from the photocathode entering the first stage. From these data, the integration necessary to obtain the form of h(p,E) was carried out graphically.

The predicted distributions, shown as continuous curves in Figure 10, were scaled horizontally to fit the peak in the experimental fluorine distribution for each crystal. The resulting factors are  $5 \cdot 70 \text{ V}$ ./MeV. for the smaller and  $7 \cdot 26 \text{ V}$ ./MeV. for the larger crystal. The vertical scales were chosen in each case to fit the experimental histogram.

## V. DISCUSSION OF RESULTS

It can now be seen that the broadening of the peaks in pulse-height distributions for homogeneous  $\gamma$ -radiation above 6 MeV. is explained as the effect of bremsstrahlung escaping from the crystal and taking with it an appreciable and variable fraction of the incident photon energy.

The histograms of f(I,E) in Figure 6, neglecting statistical fluctuations and the spurious peaks due to the multiple scattering assumption, correspond to the pulse-height distributions obtainable if the spread due to electron statistics in the photomultiplier were very small. These show that at 12 and 18 MeV. the increase in crystal size from 1 in. to 3 in. considerably reduces the tail at the low end but leaves the peak still quite broad.

The increase in crystal size increases the reabsorption probability not only of bremsstrahlung but also of positron annihilation radiation. At 6 MeV. this results in a change from one prominent peak (events in which neither annihilation quantum is captured) to three peaks of comparable height which would be difficult to resolve experimentally. Thus for many purposes the smaller crystal would be preferable at this energy, unless it were possible to use a crystal so much larger again that only the peak due to capture of both quanta is prominent.

Another feature seen in these histograms is that, at energies where bremsstrahlung loss is important, the peak of each histogram corresponds to some energy less than the incident  $\gamma$ -energy, and the nearer to it the larger is the crystal. The cut-off at the high end corresponds in each case to the incident energy.

The forms of h(p,E) are compared with the experimental pulse-height distributions in Figure 10. The important features in the comparison are the abscissa values at which peaks and cut-offs occur in the B<sup>11</sup> $(p,\gamma)$  and Li<sup>7</sup> $(p,\gamma)$  spectra, the position of the F<sup>19</sup> $(p,\alpha\gamma)$  peak having been used for calibration.

These are such as to confirm the results of the Monte Carlo calculation of the effect of bremsstrahlung escaping from the crystal. As regards the actual shapes of these peaks, it appears that in each case the experimental peak is spread out even more than predicted. This can be explained by the fact that, wherever approximations were necessary in the Monte Carlo calculation, they were made in such a direction as to underestimate the amount of bremsstrahlung which escapes, so that the predicted curves exhibit energy resolution better than attainable.

The width of the peak for the  $F^{19}(p,\alpha\gamma)$  radiation is also greater than predicted. This could be due to the channel width of the analyser, non-uniform sensitivity over the photocathode surface, and inequality of light collection efficiency between different parts of the crystals. In the case of the 3 in. composite crystal, any variation in fluorescence efficiency between the individual crystals would add to the spread.

The lower ends of the pulse-height distributions diverge upward from the predicted curves. This can be explained as the effect of radiation scattered into the crystal from matter outside it. No collimation was used, and no particular care was taken to minimize the amount of scattering material in the neighbourhood of the target and crystal. In particular, the  $\gamma$ -radiation entered the 3 in. crystal through a depth of about  $\frac{3}{8}$  in. of "Perspex".

Following a suggestion of Hofstadter and McIntyre (1950), some workers have reported improved energy resolution with scintillation counters above 1 MeV. by using three crystals in triple coincidence to isolate those pulses due to events in which a positron is annihilated in the main crystal and both annihilation quanta escape from it (Johansson 1950; Bair and Maienschein 1951). At the energies considered in the present paper, the form of f(I,E) for a detector of this type would be practically that given by the full lines in Figure 3, since the creation of a positron by other than the incident photon does not often For 6 MeV. radiation, the low energy tail is small, but it increases both occur. with increasing energy and with decreasing crystal size. Comparing Figure 3 with Figure 6, it is seen that the spreading due to bremsstrahlung is less than in the single crystal case, because the average energy of the secondary electrons is less. At 18 MeV., however, the spreading has become so great that there is little to be gained by using the triple coincidence technique and, at higher energies still, all advantage disappears because pair production becomes virtually the only primary process.

Therefore with  $\gamma$ -radiation above 10 MeV. it is a fundamental limitation to the energy resolution of scintillation counters that a fraction of the incident energy is lost to the crystal by bremsstrahlung in each detecting event and that this fraction differs widely between events. The only way to overcome this appears to be to increase the size of the crystal so that the escaping fraction is kept small. Thus, while at lower energies the minimum crystal size is determined by the necessity to ensure that electron ranges are small compared with crystal dimensions, above 10 MeV. the crystal dimensions would need to be large compared with the mean free path of the bremsstrahlung photons. In sodium iodide, this is about 4 cm. for 1 MeV. photons, increasing to 8 cm. for 5 MeV. and then decreasing very slowly at higher energies.

Nevertheless, sodium iodide crystals of the sizes we have used are still useful up to 18 MeV., having the properties that

- (i) the efficiency of detection, as shown in Table 1 for photons entering parallel to the crystal axis, is good;
- (ii) the energy of a component of the radiation can be estimated by the position not of the peak but of the end-point of the distribution;
- (iii) pulses from the highest energy component can be isolated by a simple discriminator set above the end-point of the distribution due to the component next below.

Photon	l in. Crystal	2 in. Crystal
(MeV.)	(%)	(%)
ß		69
12	30	66
12	33	70

## TABLE 1

PERCENTAGE DETECTION EFFICIENCY OF SODIUM IODIDE CRYSTALS

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