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AN ANTICOINCIDENCE GAMMA RAY SCINTILLATION SPECTROMETER

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AN ANTICOINCIDENCE GAMMA RAY SCINTILLATION SPECTROMETER- ABSTRACT -

A new type of scintillation spectrometer is described. It is designed to suppress signals arising from incident photons that are not completely absorbed in the primary detecting NaI(Tl) crystal. When an undesired signal is detected by means of a cluster of NaI(Tl) crystals surrounding the primary crystal, it is removed through the discriminating action of an anticoincidence electronic gate. Under conditions of good discrimination, this type of arrangement offers the advantage of yielding the gamma ray spectrum directly without any interpretation of results which might become laborious, particularly if the incident gamma radiation has a complex spectrum. Results are presented that illustrate the discrimination action of the device by means of photographic representations of the scintillation gamma ray spectra of Cs¹³⁷ and Na²².

AN ANTICOINCIDENCE GAMMA RAY SCINTILLATION SPECTROMETER1. INTRODUCTION

The successful use of a photomultiplier-fluorescent material combination for detection of radioactivity has been reported by many investigators^{1-6/}. High efficiency large organic crystals (naphthalene), which are transparent to their own fluorescence, were first used by Kallman^{2/} as scintillators for beta and gamma radiation. Since then a large number of useful transparent scintillation crystals have been discovered, of which anthracene, stilbene, and sodium iodide (thallium activated) because of their relatively large light outputs are in popular use today. A careful study of the properties of sodium iodide for gamma ray detection has been carried out by Hofstadter^{7/}. Both he and Bell* compared the light output of sodium iodide with anthracene, and found that the pulse height from the former is about five times that of the latter when a slow amplifier is used.

There are further advantages in using NaI(Tl) for gamma ray detection, since the crystal has a relatively large density (3.7), and contains a medium atomic number element; iodine constitutes 85% of the crystal weight. Therefore, it has a good detection efficiency for gamma radiation in the low energy region characterized by Compton and photoelectric interactions with matter, as well as in the higher energy region where the dominating interaction is pair formation. All three effects have been observed in various spectra obtained using NaI(Tl) scintillation crystals^{8-14/}.

In the low energy region, the photoelectron spectrum is a useful key to identification of a complex gamma ray spectrum^{8-10/}. This is because all of the energy of the incident gamma photon is essentially transferred to a photoelectron and X-Ray or X-Rays which are then easily absorbed by the scintillator.

* Results presented at conference (July 1948) on high speed counters at the University of Rochester.

In this way, one obtains a line that is in unique correspondence to the incident gamma ray, rather than a continuous spectrum which is characteristic of the Compton process.

Likewise, in the higher energy region, the pair spectrum is a useful tool for identification of gamma rays^{12-14/}. However, here one does not always obtain a unique one to one correspondence between the measured and incident gamma ray spectrum since one or both of the annihilation photons may escape from the crystal without being degraded. This results in the possible appearance of a triad of lines in the spectrum for an initially monoenergetic gamma ray corresponding to absorption of both, absorption of one, or absorption of neither of the annihilation photons by the detecting crystal.

A number of methods have been advanced for eliminating the multiplicity of lines and continuum that appear in the scintillation spectrum and complicate the interpretation necessary to determine the spectrum of the incident gamma rays. It will be of interest to discuss them briefly, since all of the types operate on a coincidence or anticoincidence principle.

The method developed by Hofstadter and McIntyre^{15/} limits the measuring sensitivity of the instrument to a particular angular range of gamma scattering, and thus to a certain portion of the Compton spectrum. This is done by gating the pulses produced when collimated gamma radiation is detected by one crystal with coincident pulses occurring when the scattered radiation strikes a second crystal nearby. In order to get the best resolution-solid angle relationship, the second crystal is oriented so that it receives radiation that has been back-scattered from the first crystal, which corresponds to the peak of the Compton electron distribution. However, these authors observed that at high energies additional lines appear in the spectra due to pair formation. These are gated by detection of the annihilation photons in the gating crystal. This suggested to the authors a method of observing only the pair spectrum by gating pulses from the primary crystal with a coincident pulse occurring when two crystals flanking the primary crystal are simultaneously excited.

Following this suggestion, an instrument was constructed by Bair and Maenschein^{16,17/} which possessed pair peak resolutions of about ten to fifteen per cent full width at half maximum. The same investigators developed a method for observing the Compton spectrum only by gating the output of the primary crystal so that a pulse appears only when a coincidence in one and an anticoincidence in the other flanking crystal occurs. Results have also been reported for a similar pair spectrometer independently developed^{18/}.

2. PRINCIPLE OF OPERATION

The instrument to be described here differs from the types of unique response gamma ray scintillation spectrometers previously discussed. Its operating principle is to discriminate against signals that arise when the transfer of energy from the incident gamma ray photon to the primary detecting crystal is not complete. This is achieved by arranging a cluster of scintillation crystals to surround as completely as possible the primary crystal. By making the cluster volume large, most of the radiation that is not fully degraded by the primary crystal will be sensed by photomultipliers arranged to detect cluster scintillations only. If a signal occurs simultaneously in these photomultipliers and the photomultiplier that detects radiation from the primary crystal only, it is removed from the primary pulse height distribution by the action of an anticoincident gate.

Under conditions of good discrimination, this type of arrangement offers the advantage of yielding the gamma ray spectrum directly* without any interpretation of results which might become laborious, particularly if the incident radiation possesses a complex spectrum**. It differs from types

*We assume that the resolving time of the apparatus is much larger than the time required for complete degradation of the gamma ray.

**It will later be shown that only a modest amount of NaI(Tl activated) crystalline material is required to remove 90% of the undesired signal; and it does not appear impractical to increase this figure to 99%.

previously described in that its response is essentially independent of whether the energy of detection arises from Compton, photoelectric, or pair formation interactions. The following events contribute mainly to full degradation of the gamma ray by the primary detecting crystal:

Production of

- a. A photoelectron and X-rays;
- b. A high energy Compton electron, and a photoelectron and X-rays arising from absorption of the back-scattered photon; or several Compton electrons, a photoelectron and X-rays;
- c. A positron-electron pair, when energetically possible, and electrons and X-rays arising from absorption of the annihilation radiation and bremsstrahlung through processes (a) and (b) above. A much larger primary crystal is required for completing this event than for possibilities (a) and (b) above.

5. DESIGN OF INSTRUMENT

The main features of the instrument are shown in the illustrations of Figure 1a and 1b. It consists essentially of two concentric aluminum cylinders. The inner cylinder, whose thin walls are less than $1/32$ " in thickness, forms the inner chamber which contains the primary NaI(Tl) crystals (about $1/2$ " cube) surrounded by a hemispherical aluminum reflector that, in combination with the light pipe, directs the primary fluorescence to an EMI 5311 photomultiplier tube. The volume between the two cylinders forms the outer chamber, which is completely filled with random size chunks of NaI(Tl) immersed in heavy paraffin oil which serve to detect radiation scattered into them by the primary crystal. Gamma rays from the source, at the left, are axially collimated so that the gamma ray beam is intercepted by the primary crystal. The primary crystal is displaced on the axis toward the source in order that a longer path exist in the outer crystal cluster for radiation deflected in the forward hemisphere than for the less energetic back scattered

FIGURE 1a SCHEMATIC CROSS-SECTION OF ANTICOINCIDENCE SCINTILLATION GAMMA RAY SPECTROMETER

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Photograph of spectrometer chamber and photomultipliers with cutaway drawing to show view of spectrometer interior.

Figure 1b

radiation. Part of the outer cluster occupies the region behind the primary crystal in order to increase suppression of back scattered gamma rays, although at the expense of primary beam intensity. All crystals are NaI (Tl activated) grown by Harshaw Chemical Company.

Fluorescence from the primary crystal is propagated forward through a quartz or lucite pipe to an EMI 5311 photomultiplier which is magnetically shielded. Fluorescence from the surrounding crystals is detected by one or more of three 5819 photomultipliers attached to the outer aluminum cylinder by means of Wilson seals. The entire arrangement is placed in a light-tight box during operation. Power and signal leads go to electronic components mounted in an adjoining relay rack.

4. PROCEDURE FOR REDUCING LIGHT REFLECTION AND TRANSMISSION LOSSES

In order to reduce light losses several precautions are necessary. In the inner chamber losses may occur during reflection at the surface of the aluminum hemispherical shell housing the spectrum crystal and internal reflections occurring while propagating through the light pipe. Such losses are considerably reduced by highly polishing the reflecting surfaces. A light pipe of quartz is superior to one of lucite because of its greater hardness which enables it to acquire a high polish that it retains under handling over a long period of time. Light losses resulting from internal reflections at the crystal-lucite and lucite-photomultiplier contact surfaces are reduced in the customary way by replacing the intervening air surface layer with paraffin oil, canada balsam, or some other highly transparent substance having a refractive index closer to that of NaI and lucite.

Light transmission losses are reduced by cleaning and polishing all crystals. The crystals are polished using a fine grade of emery and then chemically cleaned with acetone.

Reflection losses in the outer chamber are reduced by means of a high polish on the inside surface of the outer cylinder and outside surface of the inner cylinder. Light losses resulting from internal reflection at the crystal surfaces are reduced by filling the outer chamber volume not occupied by NaI (Tl) crystals with heavy paraffin oil.

In order to test whether serious light losses occur in the outer chamber volume, the coincidence rate between two of the circumferential tubes was measured. Since the value obtained is 80% that of the single counting rate, further efforts to reduce light losses are not considered necessary or worth while.

5. GEOMETRICAL DESIGN CONSIDERATIONS

Several factors were considered in choosing the geometrical design, i.e., the shape and dimensions of the inner and outer chambers, position of primary crystal, and location of photomultipliers and other components.

The light pipe is situated so that light is transmitted in the direction away from the source to the photomultiplier. This direction was chosen so that anti-coincidences resulting from gamma rays that pass through the light pipe and thus do not create scintillations in the outer chamber transfer a very small energy during a single Compton scattering in the primary crystal. The small pulses thus resulting do not significantly affect the spectrum in the energy region which has so far been investigated (below 1.28 mev.).

The three outer photomultipliers are located in the rear half of the chamber so that the light detection efficiency of the system for back and laterally scattered gamma radiation is greater than for forward scattered radiation. This is desirable since the light output from the back scattered radiation is less than that of the more energetic forward scattered radiation.

6. GEOMETRICAL DESIGN SPECIFICATION

The geometrical design of the spectrometer was calculated on the premise that the most important contribution to the undesired signal arises from Compton radiation singly scattered in the detecting crystal. An instrument of this design will operate effectively, although not in an optimum manner, in the higher energy region where pair formation processes dominate. The theoretical curve for the spectrum of Compton scattered electrons produced by an incident one MeV gamma ray is shown in the upper curve of Figure 2. The criterion chosen as a basis for the design calculation is that the Compton electron spectrum be transformed to an energy independent value of one-tenth its former peak amplitude. The response of the spectrometer to Compton electrons is then that of the lower curve of Figure 2, for the particular case when the incident gamma ray energy is one MeV.

The experimentally measured spectrum will differ somewhat from that obtained theoretically, because of neglect of photomultiplier response and multiple scattering in the crystal. The former depends on statistical variation in the number of electrons emitted at the photocathode and variations in the multiplication factor of the succeeding dynodes^{19/}. However, it is felt that neglect of these effects will not introduce a serious error in determination of the proper design geometry.

7. GEOMETRICAL DESIGN CALCULATION

Consider the primary detecting crystal to be a point source of gamma rays scattered out of an incident collimated beam and radiating into a medium whose gamma absorption coefficient $\mu(E_0, \theta)$ is a function of the incident gamma ray energy and the angle θ through which it is deflected. The angular dependence of $\mu(E_0, \theta)$ arises from the Compton relation between energy and scattering angle.

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Cross sectional area per electron presented to an incident one mev. photon for formation of a compton electron of energy E in the element of energy dE.

Figure 2.

The polar plot of $r(\theta)$ which is determined according to the criterion previously mentioned represents the cross-sectional area and shape of the desired cylindrically symmetrical spectrometer volume; the origin of the plot corresponds to the location of the primary crystal.

The polar graph of the mean free path ($1/\mu$) in the outer chamber scattering volume as a function of the angle through which gammas are deflected by the primary crystal is plotted in Figure 3. It is obtained using semi-empirical gamma ray mass absorption curves²⁰, and considering only the 85% iodine constituent contribution to the scattering. The absorption coefficients include Compton scattering which should be effective since it is only necessary that the gamma ray give up enough of its energy to the outer chamber medium to produce a detectable photomultiplier pulse.

The expression for the Compton electron energy distribution as a function of the angle through which a collimated beam of gamma rays is deflected by a scattering volume, which, of course, is directly related to the electron energy, is

$$N_e(\theta, E_0) = \phi_\gamma \frac{d\sigma(\theta, E_0)}{d\Omega} \frac{d\Omega}{dE} \rho_e V$$

where ϕ_γ is the incident gamma ray flux, $\frac{d\sigma}{d\Omega}$ is the Klein-Nishina differential scattering cross-section per electron, E_0 is the incident gamma ray energy, θ is the angle of deflection, ρ_e is the electron density, V is the scattering volume, and $d\Omega$ is the solid angle element.

Plot of mean free path in $\text{NaI}(\text{TI})$ for a incident photon of energy E_0 scattered through an angle θ by means of Compton effect. (includes only the 85% iodine contribution to the scattering.)

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Figure 3.

We wish to find the function $r(\theta)$ that will transform the Compton electron spectrum to a constant one-tenth that of the peak amplitude. Therefore, $r(\theta)$ must satisfy the following relation derived in the appendix, where $l(\theta)$ is the mean free path of the outer chamber scattering volume:

$$0.1 [N_e(\theta, E_0)]_{\max} = 0.1 \phi_\gamma \int_{\theta=\pi}^{\theta=0} \frac{d\sigma(\theta, E_0)}{d\Omega} \frac{d\Omega}{dE} \rho_e V$$

$$= \phi_\gamma \frac{d\sigma(\theta, E_0)}{d\Omega} \frac{d\Omega}{dE} \exp\left[-\frac{r(\theta)}{l(\theta)}\right] \rho_e V$$

since the peak amplitude in the electron spectrum corresponds to a Compton scattering through the angle $\theta = \pi$.

The values of $r(\theta)$ obtained from this relation are plotted for various initial gamma ray energies in Figure 4. As previously stated, the shape and dimensions of the desired spectrometer volume are obtained when these figures are rotated about the zero degree axis.

The manner in which the shape of these curves varies with the incident gamma ray energy is of some interest. It depends, of course, on the criterion chosen which requires that more of the distribution be removed at the energy spectrum peak than elsewhere. However, contributions to the peak arise from a constantly increasing backward angular scattering range as the initial gamma ray energy increases. Thus, it will be noted that the curves of Figure 4 change shape as the initial gamma ray energy increases in a manner as to increase the backward scattering detection efficiency at the expense of that for scattering in the forward hemisphere.

Polar plot of $r(\theta)$ versus θ for various incident gamma ray energies, E_0 . These curves determine the shape and dimensions of the outer spectrometer chamber and location of primary crystal required to satisfy a 90% figure of merit. The outside border represents the outline of the outer chamber cylinder that was finally chosen after a fifty percent crystal packing factor correction is applied to the volume.

Figure 4.

The border of the polar plot shown in Figure 4 is the outline of the spectrometer after compressing it radially so the inner cylinder coincides with the zero degree axis, and the primary crystal coincides with the origin of the polar plot and after the spectrometer volume has been corrected for an estimated fifty per cent loss in packing of the crystal cluster. Comparison of these curves indicates that the present spectrometer design is easily capable of satisfying the ninety per cent figure of merit arbitrarily chosen for these calculations.

8. RESULTS

Results obtained thus far with the instrument are shown in the photographs of Figures 5 and 6 which are time exposures of triggered pulse height distributions appearing on an oscilloscope screen. Each photograph represents the superposition of a large number of signals originating in the primary crystal which have been amplified by a photomultiplier and liner amplifier, the oscilloscope sweep being initiated by the forward edge of each pulse signal. Therefore, a good indication of the spectrum is obtained by a study of the variations in pattern intensity along the y-axis (energy axis) of each photograph. The spread of the pulse along the x-axis (time axis), which is of no significance in this particular interpretation, is due to the finite resolving time of the photomultiplier tube and amplifying circuits.

Each figure contains a set of photographs of equal time exposure. The top member of each set is a conventional representation obtained when all of the primary signal pulses are triggered. The bottom representation shows the omission of certain of the distributions caused by the unique response of the spectrometer when triggering of the sweep is performed by anticoincidence signals only.

In Figure 5 are shown the two representations of the gamma ray spectrum of Cs^{137} which emits a monoenergetic 663 kev. gamma ray. At the top of

Two representations of the gamma ray spectrum of Cs^{137} . The top oscilloscope photograph shows the conventional scintillation gamma ray pulse height distribution obtained by triggering the oscilloscope sweep so that all primary crystal pulses are displayed. The bottom oscilloscope photograph shows the distribution obtained by triggering the oscilloscope sweep so that only anticoincidence primary pulses are displayed.

Figure 5.

Two representations of the gamma ray spectrum of Na^{22} . The top oscilloscope photograph shows the conventional scintillation gamma ray pulse height distribution obtained by triggering the oscilloscope sweep so that all primary crystal pulses are displayed. The bottom oscilloscope photograph shows the distribution obtained by triggering the oscilloscope so that only anticoincidence primary pulses are displayed.

Figure 6.

both representations there appears a bright line due to complete absorption of the 663 kev. gamma ray in the primary crystal. The continuous distribution arising from single compton scattering in the primary crystal appears just below this in the top representation only. In the bottom representation the compton distribution is removed, however; the presence of another line of about 180 kev. seems indicated. Since the energy of this line is very close to that expected for 180° back-scattering of the 663 kev. gamma ray, it appears likely that it is of spurious origin. Such spurious lines have previously been observed in scintillation gamma ray spectra by Hofstadter^{21/} and attributed to the effect of gamma ray back-scattering from surroundings and crystal walls.

In Figure 6 are shown the two representations of the gamma ray spectrum Na^{22} which emits positrons and 1.28 mev. gamma rays. The line appearing at the top of both representations corresponds to total absorption of the 1.28 mev. gamma ray in the primary crystal. Just below this, in the top representation only, appears the compton distribution associated with the 1.28 mev. gamma ray. Below this in both representations there appears a bright band due to total absorption of positron annihilation radiation by the primary crystal; the associated compton distribution is omitted in the bottom representation. A bright band caused by back-scattered radiation appears near the bottom of both representations.

The resolution of the apparatus is actually somewhat better than indicated by the appearance of the annihilation radiation line width in this photograph. This is because it was necessary to overexpose the most intense part of the photograph in order to clearly view the less intense 1.28 mev. gamma ray shown at the top. However, it should be possible to effect a considerable gain in the intensity of this line at the expense of its associated compton distribution by increasing the size of the primary crystal.

- APPENDIX -

Derivation of Spectrometer Design Specification

We define the following symbols:

- E_0 - Energy of incident photon.
- E_θ - Energy of photon after deflection through the angle θ .
- E - Energy of electron corresponding to deflection of photon through the angle θ .
- $\phi_Y(E_0)$ - Incident photon flux striking the scatterer.
- $N_Y(E_\theta)$ - Number of photons of energy E_θ scattered through the angle θ by unit area of scatterer in unit time interval.
- $N_e(E, E_0)$ - Number of compton electrons with energy between E and $E + dE$ resulting from collisions occurring in a volume of scattering in a unit time interval for incident photon energy E_0 .
- $\bar{N}_e(E, E_0)$ - Value of $N_e(E)$ at the maximum electron energy for incident photon energy E_0 .
- I_0 - Intensity of incident gamma radiation intercepted by unit area of the crystal, energy per unit area per second.
- I_θ - Intensity of gamma radiation scattered by one electron of the crystal through an angle θ , energy per unit solid angle per second.
- $\frac{d\sigma(\theta, E_0)}{d\Omega}$ - Differential scattering cross-section per electron for an incident gamma ray of energy E_0 scattered through an angle θ .
- $l(\theta)$ - Mean free path in the absorbing medium for a scattered gamma ray of energy E_θ .
- ρ_e - Number of electrons per unit scattering volume.
- mc^2 - Rest energy of electron (0.5108 mev.).
- V - Scattering volume.

It will first be of interest to derive the spectrum of secondary electrons resulting from Compton scattering of gamma rays possessing an initial fixed energy E_0 . We are primarily interested in the shape of the spectrum; the magnitude of it is determined by the incident gamma flux and volume of NaI crystal scatterer which are variable quantities. The electron spectrum is determined by the relation

$$(1) \quad N_e(\theta, E_0) = \left[\phi_\gamma(E_0) \right] \frac{d\sigma(\theta, E_0)}{d\Omega} \frac{d\Omega}{dE} \rho_e V$$

where θ is a function of E . The values of $\frac{d\sigma(\theta, E_0)}{d\Omega}$ as a function of θ were obtained from tabulated values^{22/} of the Klein-Nishina^{23/} cross-section by using the relation, $\frac{d\sigma(\theta, E_0)}{d\Omega} = \frac{I_\theta}{I_0} \frac{E_0}{E_\theta}$. The values of $\frac{d\Omega}{dE}$ as a function of θ are obtained by means of the Compton relation

$$(2) \quad 1 - \cos \theta = mc^2 \left(\frac{1}{E_\theta} - \frac{1}{E_0} \right).$$

From the relation $E_0 = E_\theta + E$, we get

$$dE_\theta = -dE.$$

Therefore,

$$\frac{d\Omega}{dE} = -\frac{d\Omega}{dE_\theta} = + \frac{2\pi d(\cos \theta)}{dE_0}$$

$$(3) \quad \left| \frac{d\Omega}{dE} \right| = \frac{2\pi mc^2}{E_\theta^2} = \frac{2\pi mc^2}{E_0^2} \left[1 + \frac{E_0}{mc^2} (1 - \cos \theta) \right]^2$$

Knowing N_e as a function of θ , we can obtain it as a function of E by referring to (2) again. The shape of the electron spectrum, corresponding to incident one mev. gamma radiation, shown in Figure 2 was obtained from the product

$$\left[\frac{d\sigma(E, E_0)}{d\Omega} \frac{d\Omega}{dE} \right] \text{ and is expressed in units of } \left(\frac{e^2}{mc^2} \right)^2.$$

From the shape of the spectrum of Figure 2, it is seen that the peak occurs at the maximum value of electron energy: this value corresponds to 180° back-scattered gamma radiation, and is found by substituting $\theta = \pi$ in relation (1). The criterion of design which we have arbitrarily chosen is that the size and shape of the volume of the outer chamber reduce the spectrum to a constant background amounting to ten per cent of the peak amplitude.

The probability that a pulse produced in the spectrum crystal, as a result of a photon deflected by an angle θ , be preserved is proportional to the escape

probability for the particular photon $e^{-\frac{r(\theta, E_0)}{l(\theta, E_0)}}$ in the outer crystal cluster.

A plot of the mean free path, l , as a function of θ with E_0 as parameter is shown in Figure 3. Our criterion of design is then satisfied by solving for $r(\theta)$ in the following relation:

$$\begin{aligned} (4) \quad 0.1 \bar{N}_e(E) &= 0.1 e_Y \left[\frac{d\sigma(\theta, E_0)}{d\Omega} \frac{d\Omega}{dE} \right]_{\theta=\pi} \\ &= e_Y \frac{d\sigma(\theta, E_0)}{d\Omega} \frac{d\Omega}{dE} e^{-\frac{r(\theta)}{l(\theta)}} \end{aligned}$$

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