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ENERGY AND ANGULAR DISTRIBUTION OF GAMMA RADIATION FROM A Co^{60} SOURCE AFTER DIFFUSION THROUGH MANY MEAN FREE PATHS OF WATER

R. W. Peelle
F. C. Maienschein
T. A. Love

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R. W. Peelle F. C. Maienschein

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PREFACE

This report discusses the results of an experiment which should be considered as preliminary in nature. It is presented now because it is felt that the apparent discrepancy between the experiment and "moments method" calculations should be examined further. It will be some time before suitable equipment becomes available at the Oak Ridge National Laboratory for a more definite experiment.

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The authors wish to acknowledge the interest of E. P. Blizard, who suggested the importance of the problem and helped in the planning and in the early experimental work. We are also indebted to A. T. Futterer and G. Estabrook for their considerable calculational help in the analysis of the results.

ABSTRACT

Measurements have been made of the energy and angle spectrum of the photon energy flux resulting from 1.17- and 1.33-Mev gamma rays from a point source after diffusion in an infinite water medium. Energy flux spectra were measured for penetration distances corresponding to approximately six and nine mean free paths for the uncollided source photons and for angles of gamma-ray emission varying from 0 to 90 deg. Spectral measurements were made with a Compton spectrometer having a peak-to-total ratio of 0.7 and a resolution of 14% (full width at half maximum) for 1.12-Mev photons. The resulting energy and angle spectra of the energy flux are compared, after integration over angle, with calculated results which have been obtained by the "moments method." The results are also compared with the available calculated differential results.

INTRODUCTION

Considerable efforts have been expended[1] to calculate the details of the intensity distribution of gamma rays which penetrate or diffuse through media whose linear dimensions are large compared with an interaction mean free path for the photons involved. Difficulty arises in the calculations because of the complexity of the scattering process. The calculational problem tends to be more severe for absorbers consisting of elements of low atomic number, where Compton scattering is the most important effect, and for penetrations which correspond to many mean free paths of the primary radiation.

The most trusted calculations available at this time are "moments method" calculations resulting from a combined National Bureau of Standards and Nuclear Development Associates computing program.[1-4] These calculations, supposed to be valid for an infinite homogeneous medium, have provided the energy spectrum of the energy flux, integrated over all solid angles for an isotropic detector. Further integration has been performed to obtain various significant buildup factors. The dose buildup factors obtained from the moments method calculations have been compared favorably with dose measurements obtained in water[1,5] as

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1. L. V. Spencer and U. Fano, Phys. Rev. 81, 464 (1951); see also L. V. Spencer and U. Fano, J. Research Nat. Bur. Standards 46, 446 (1951).

2. L. V. Spencer and F. Stinson, Phys. Rev. 85, 662 (1952).

3. U. Fano, J. Research Nat. Bur. Standards 51, 95 (1953).

4. H. Goldstein and J. E. Wilkins, Jr., "Calculations of the Penetration of Gamma Rays," NDA-15C-41 or NYO-3075 (June 30, 1954).

5. Gladys R. White, Phys. Rev. 80, 154 (1950).

a function of detector separation distance from a Co^{60} point source. Dose rate checks have also been made in media of iron[6,7] and lead,[7] confirming the results of these calculations. In addition, Hayward[8] measured the electron spectrum observed in an anthracene crystal when it was placed in a water medium at various distances from a Co^{60} point source. This experiment agreed with a special moments method calculation designed for testing the experimental results. These integral experiments have been interpreted as substantiating the general validity of the results of the moments method calculational program.

In spite of the apparent agreement between these integral measurements and the calculations, it was considered important to check the differential spectra predicted by the calculations to provide a more stringent test. Therefore, an experiment designed to give these differential spectra was performed at ORNL. In this experiment the energy and angle spectra of the photon energy flux rerulting from the diffusion in an infinite water medium of 1.17- and 1.33-Mev gamma rays from a point (Co^{60}) source were determined. The gamma-ray spectra were measured at several angles between 0 and 90 deg for penetration distances corresponding to 6 to 9 mean free paths for the uncollided source photons. It has been possible to provide an absolute normalization for the ORNL experiment so that the integrated experimental results could be compared with the calculated values.

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6. L. A. Beach et al., Phys. Rev. 92, 355 (1953).
7. C. Garrett and G. N. Whyte, Phys. Rev. 95, 889 (1954).
8. E. Hayward, Phys. Rev. 86, 493 (1952).

Since the ORNL experiment was performed, a similar experiment was carried out by G. N. Whyte[9] for a concrete medium which terminated at the point of detection for angles up to 60 deg. Whyte's results integrated over solid angle appear to be in agreement with the calculated shapes. No absolute normalization was attempted in Whyte's experiment, however.

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9. G. N. Whyte, Can. J. Phys. 33, 96 (1955).

I. EXPERIMENTAL PROCEDURE

A double-crystal Compton scintillation spectrometer[10-12] was used in this experiment because of its reasonably unique response to incident monoenergetic gamma rays. Figure I shows the geometry of the spectrometer and the source as suspended in the pool of water at the Bulk Shielding Facility (BSF). Although the term "detector angle" implies that the spectrometer was repositioned throughout the experiment, in practice the source was actually moved by means of a swinging arm pivoted just above the end of the spectrometer collimator, as dedetermined by a plumb. This source could then be set so that the detector angle θ had any value between 0 and 90 deg, while maintaining a constant source-detector separation. The spectrometer was positioned about 10 ft from the sides and bottom of the BSF pool and about 15 ft from the water surface.

Two Co^{60} sources were used during the course of this experiment, one having a strength of approximately 100 curies and the other approximately 195 mc. The 195-mc source was calibrated against a small (1-mc) source whose strength was known to within about 3% (as the result of measurements with a calibrated high-pressure ionization chamber[13]). The ratio between the 195- and the 1-mc sources was obtained by counting and dosimetry techniques. The 195-mc source was also

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10. F. C. Maienschein, "Multiple Crystal Gamma-Ray Spectrometer,"ORNL-1142 (April 14, 1952).

11. F. C. Maienschein and T. A. Love, Nucleonics 12, No. 5 (1954).

12. T. A. Love, R. W. Peelle, and F. C. Maienschein, "Electronic Instrumentation for a Multiple-Crystal Gamma-Ray Scintillation Spectrometer," ORNL-1929 (Oct. 3, 1955).

13. This high-pressure chamber was used through the courtesy of the Analytical Chemistry Division.

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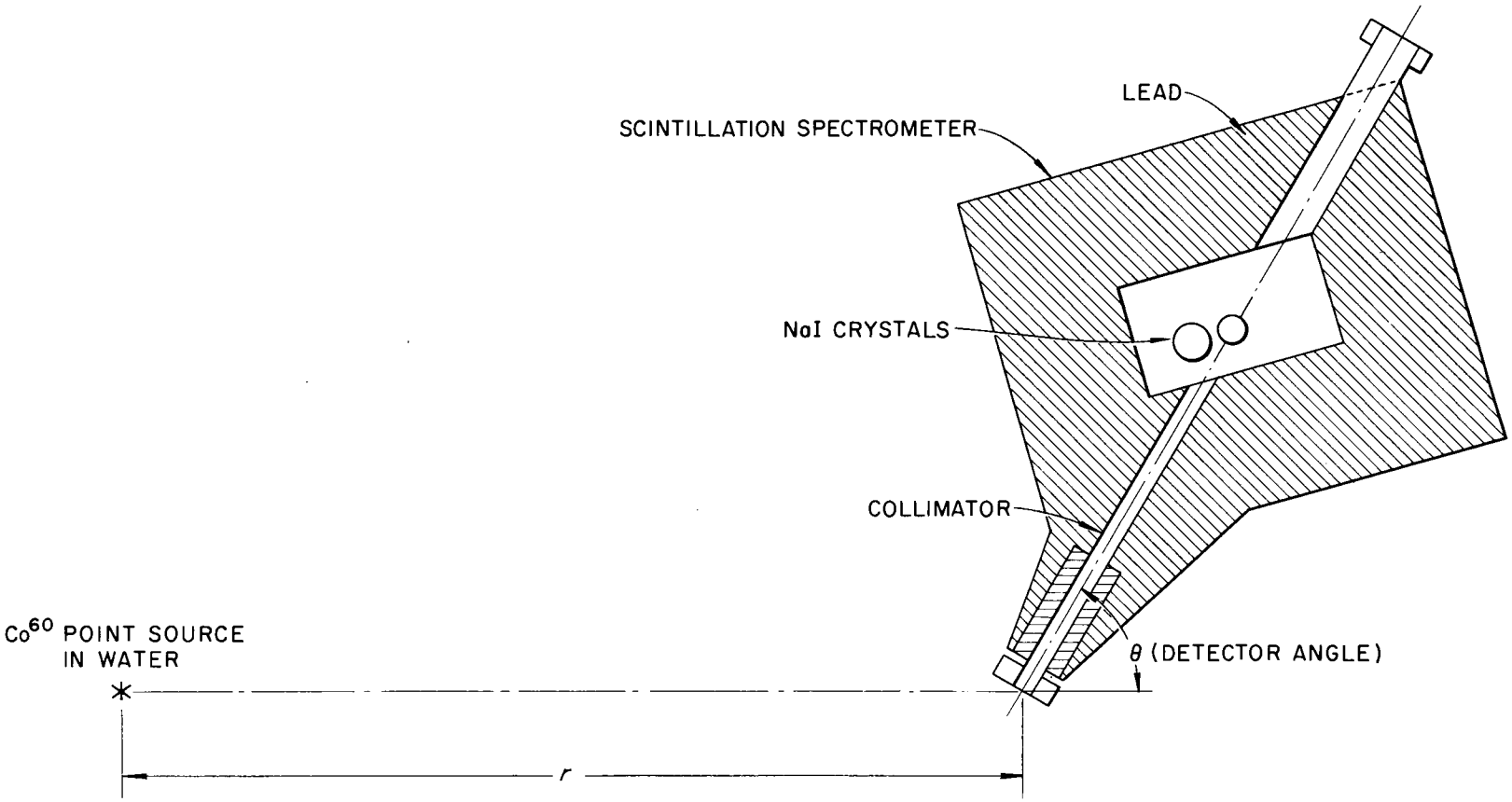

Fig. 1. Geometry for the Gamma-Ray Spectrometer and Co^{60} Source.

sent to the National Bureau of Standards for a dose calibration; the results were corrected approximately for degradation. The 100-curie source was calibrated against the 195-mc source. Table I shows the strengths of the sources used, the methods of calibration, and the precision to which they are believed to be known. For the calibration by detector responses through 90- and 140-cm thicknesses (r) of water, pure exponential attenuation was assumed with little error because of the smallness of the solid angle accepted by the spectrometer ($\sim 2.2 \times 10^{-3}$ steradians).

Figure II shows typical pulse-height response data of the double-crystal spectrometer to several sources. The uniqueness of response is not as good as that reported by Whyte,[9] presumably because of the fact that Whyte used a superior detector geometry. The spectral sensitivity of the spectrometer, divided by the gamma-ray energy, is shown in Fig. III. Only the approximately gaussian peak of the experimental response functions is counted in determining this curve. The basic efficiency curve was taken from an earlier report[10] except for a change of absolute normalization based upon the instrument response to the 1-mc Co^{60} source.

Energy spectra were obtained for a number of detector angles (θ) between 0 and 90 deg at each of two penetration distances (90 and 140 cm). Background spectra, corresponding to the detection of photons which penetrated the walls of the lead spectrometer shield, were obtained by repeating each run with a 6-in.-long lead plug filling the nose of the collimator. Table II shows a summary of all the sets of experimental parameters studied. It should be noted that measurements obtained for scattering angles between ± 3 deg are strongly affected by the angular aperture of the collimator.

Table I. Strengths of the Two Co^{60} Sources

Source	Method of Calibration	Source Strengths (disintegrations/sec) Calibrated	Used in Data Analysis
195 mc	Against 1-mc source	$0.72 \times 10^{10} \pm 5\%$	$0.72 \times 10^{10} \pm 4\%$
	NBS dose calibration	$0.73 \times 10^{10} \pm 4\%$	
	Response of Compton spectrometer, r = 90 cm, $\theta = 0$	$0.71 \times 10^{10} \pm 8\%$	
100 curies	Response of Compton spectrometer, r = 140 cm, $\theta = 0$	$4.2 \times 10^{10} \pm 8\%$	$3.8 \times 10^{12} \pm 8\%$
	Response of Compton spectrometer, r = 90 cm, θ = 10 deg as compared to response to 195-mc source	$3.8 \times 10^{10} \pm 7\%$	

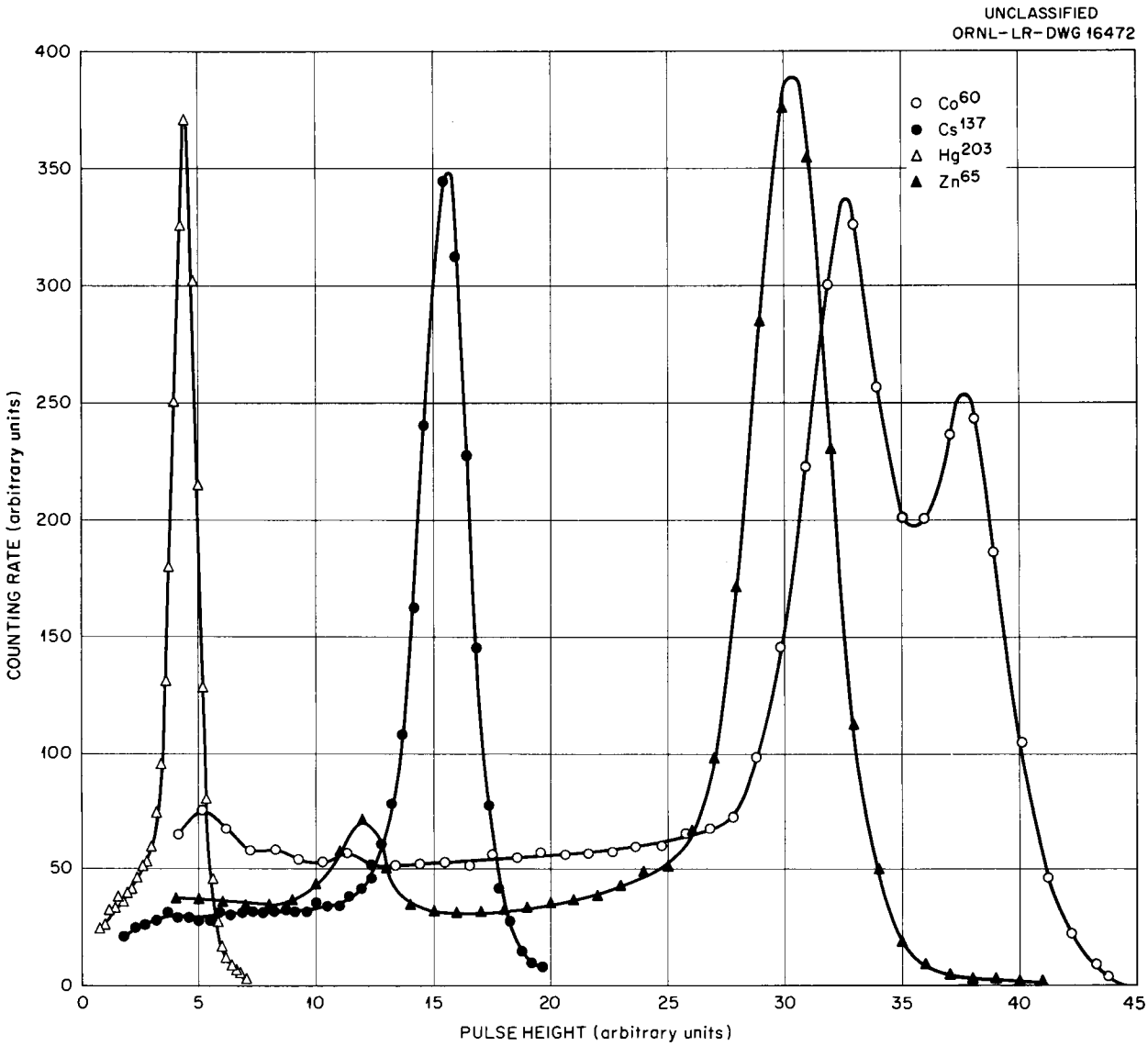

Fig. II. Response of the Compton Spectrometer to Monoenergetic Gamma Rays. Arbitrarily normalized pulse-height spectra are shown on the same relative energy scale for radioisotope sources placed at the nose of the spectrometer collimator. The sources used were Hg^{203} (280 kev), Cs^{137} (662 kev), Zn^{65} (1.12 Mev), and Co^{60} (1.17 and 1.33 Mev).

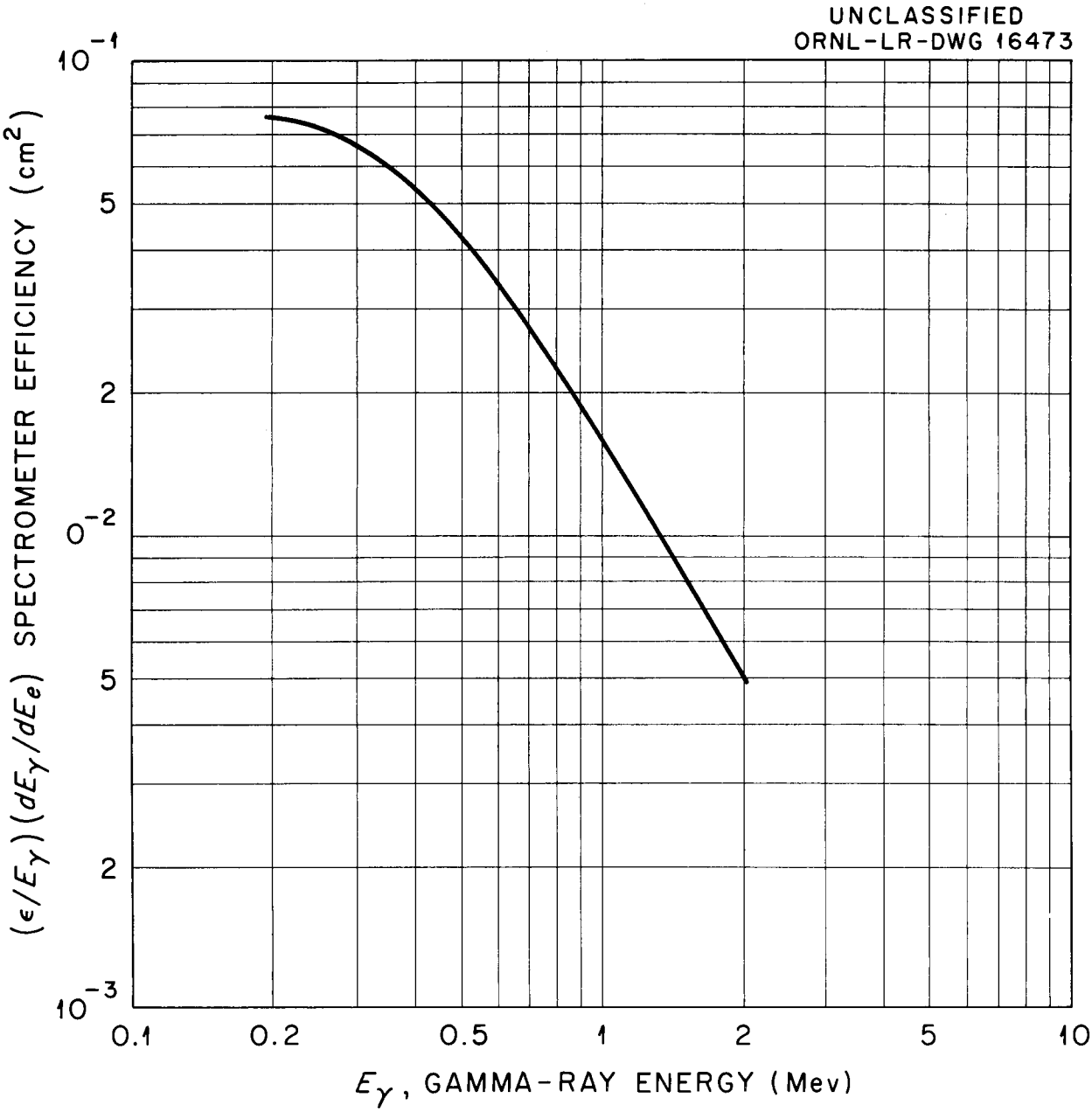

Fig. III. Spectral Efficiency of the Compton Spectrometer, $\frac{\epsilon}{E_\gamma}\frac{dE_\gamma}{dE_e}\left(\frac{\text{cm}^2}{\text{Mev}}\right)$. This function, when multiplied by the differential photon energy flux, I, at the end of the collimator and by the collimator solid angle, yields the counting rate observed per unit of absorbed electron energy E_e in the central crystal of the Compton spectrometer. This curve is based on the efficiency curve previously reported[10] and upon the spectrometer geometry.

Table II. Summary of Parameter Sets Studied in Experimental Attenuation Measurements

Source Strengths (disintegrations/sec)	r, Separation Distance (cm)	θ, Scattering Angle (deg)
0.72×10^{10}	90	0, ± 2, 4, 10
3.8×10^{12}	90	15, 30, 50, 70, 90
	140	0, ± 2, 4, 10, 30, 60, 90

II. METHOD OF ANALYSIS OF THE DATA

Calculation of the Corrected Yields

The raw-pulse-height distributions, observed experimentally in terms of counting rate per pulse-height channel, must be subjected to the following corrections in order to obtain the experimental photon number spectrum (photons Mev^{-1} sec^{-1} cm^{-2}):

(1) The pulse-height scale must be converted to a photon energy scale.

(2) The observed yield per pulse-height interval must be converted to pulses per photon energy interval.

(3) Correction must be made for the detection efficiency of the spectrometer as a function of energy.

(4) Correction should be made for the nonuniqueness of the response of the spectrometer.

These corrections are all straightforward except the last one. The need for this correction arises from the fact that a beam of monoenergetic gamma rays incident upon the spectrometer are represented at the spectrometer output by a distribution of pulses of the different size, as shown in Fig. II. The experimental results presented here have been corrected only for the "tail" corresponding to gamma rays near the original energies of 1.17 and 1.33 Mev.

Figure IV shows the manner in which the experimental response function for $\theta = 0$ deg and $r = 140$ cm was separated into two components supposed to correspond to the discrete source energies of 1.17 and 1.33 Mev. The spectral shapes of Fig. IV were then applied to data for larger angles in order to obtain the desired photon number spectrum.

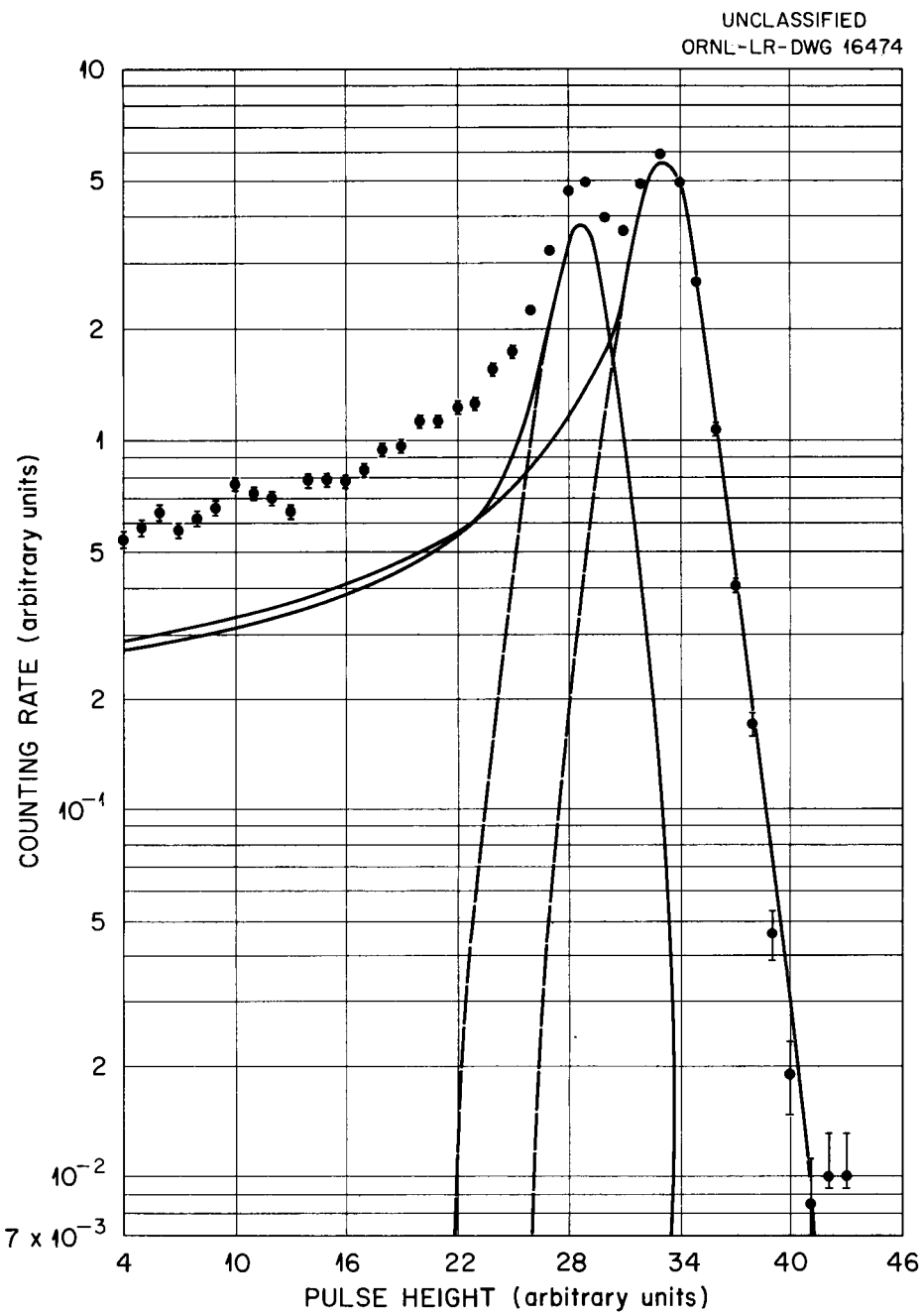

Fig. IV. Decomposition of the Observed Co^{60} Spectrum into Peaks Corresponding to the Two Source Energies (θ, 0 deg, r = 140 cm). The points indicate the pulse-height spectrum observed experimentally. The solid lines indicate the responses which would have been expected if individual sources of 1.17 and 1.33 Mev had been used. The dashed lines indicate the extension of gaussian resolution functions. The combined spectrum, corrected for spectrometer nonuniqueness, would consist of the sum of the two gaussian resolution functions shown.

Figure V shows the nonuniqueness subtraction made for a measurement at $\theta = 4$ deg and $r = 140$ cm. The normalized response functions are shown fitted to the humps arising at photon energies of 1.17 and 1.33 Mev. The experimental points at lower energies are shown before and after subtraction of the "tails" corresponding to these humps. There were three aspects of the nonuniqueness problem which are not considered in the method described above:

(1) An estimate of the errors arising from the continuous nature of the high-energy section of the scattered photon spectrum was made by observing the effect upon the estimated nonuniqueness correction of splitting the region above 1.1 Mev into more than two groups corresponding to the original source gamma rays. Separate "tail" subtractions were made for each of these energy groups, using interpolated monoenergetic response functions. In the cases where this alternate technique was used, the change of the resulting photon spectrum at any point was less than the known statistical uncertainties in the original pulse-height spectrum.

(2) Reasonably good instrument response curves were available for three gamma-ray energies but corrections were made only for the primary energies of the Co^{60} source, since the calculations were made by hand. The error induced in the photon number spectrum by the failure to subtract a tail corresponding to portions of the measured spectrum below 1 Mev has been estimated crudely for the points at the lowest measured energy (0.25 Mev). At these points, where the relative error from this source should be a

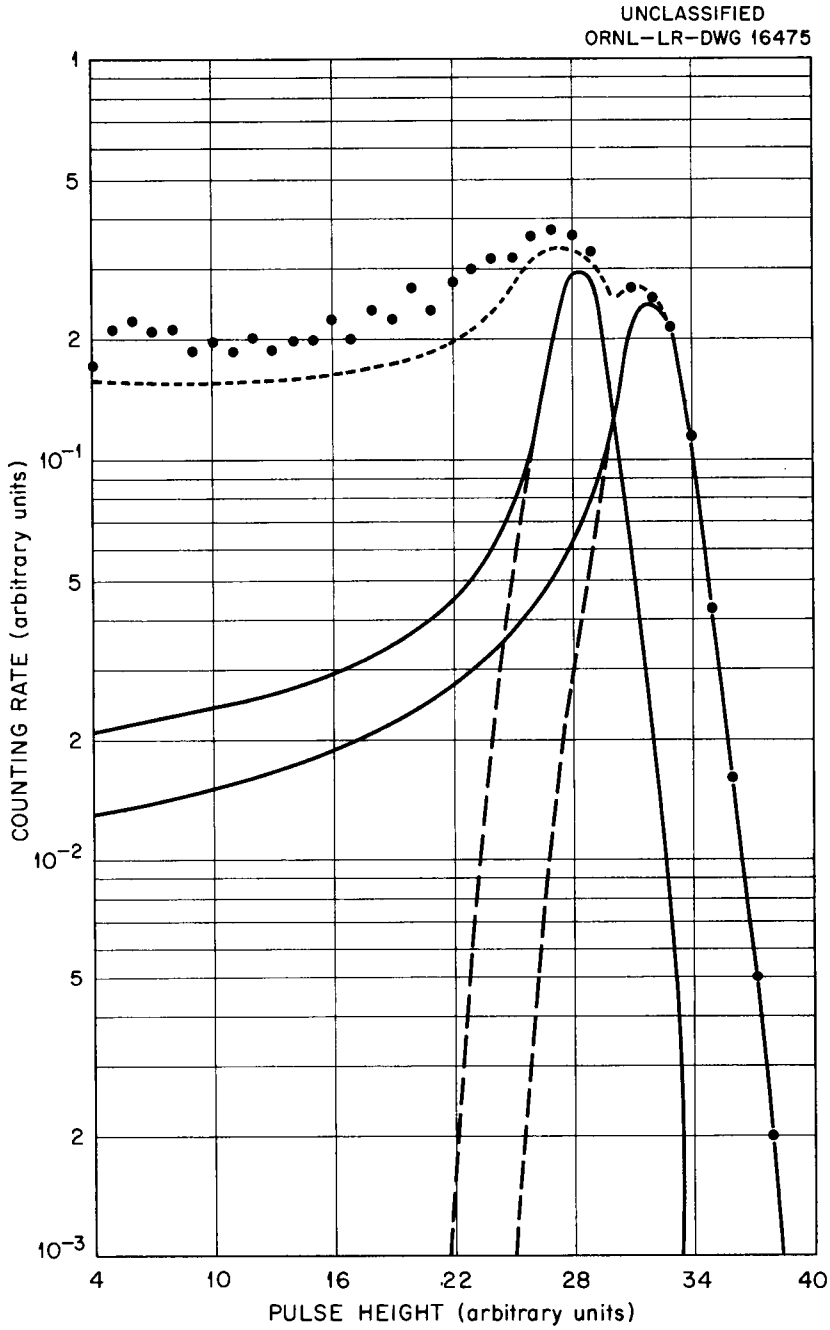

Fig. V. Nonuniqueness Correction of the Pulse-Height Spectrum Observed at $\theta = 4$ deg, r = 140 cm. The points indicate the pulse-height spectrum observed experimentally. The solid curves are the same shape as those in Fig. IV , but here they are normalized to fit the high-energy portion of the experimental spectrum shown. The dashed lines represent extensions of the normal resolution functions. Nonuniqueness corrections were made by subtracting from the experimental spectrum the area between the solid and dashed curves for both the 1.17- and 1.33-Mev peaks. The dotted line is final corrected curve. It was this curve that was used to derive the photon number spectrum observed by the spectrometer.

maximum, estimated errors range between 5 and 15% (the error leads to the acceptance of too large a flux value), increasing as the pulse-height distribution becomes more flat. These estimates were made by assuming an analytical form for the true spectrum and for the energy dependence of the "peak-to-total" ratio. The peak-to-total ratio consists of the total number of counts in the peak of the distribution divided by all the observed counts down to zero pulse height.

(3) The width of the resolution function at the peak of the spectrometer response was completely ignored in the analysis presented here. This is felt to have no appreciable effect upon the results.

Determination of the Energy Flux

The energy spectrum of the photon energy flux, I, is given by the relation:

$$I(r,E_o,E,\theta) = \frac{EN(r,E_o,E,\theta)}{S\Omega} = (cm^{-2}\ sterad^{-1})$$

where

r = source-detector separation distance (cm); the detector is consistently considered to be located at the nose of the spectrometer collimator (see Fig. I),

E_o = source photon energy, equal to 1.17 and 1.33 Mev for each source disintegration,

E = energy of the observed scattered photon, or more precisely the photon energy corresponding to the observed pulse height (Mev),

θ = detector angle (see Fig. I),

$N(r,E_o,E,\theta)$ = experimental number of photons per second per unit energy interval per unit collimator area for specified r and θ values,

S = absolute source strength (disintegrations/sec),

Ω = magnitude of the sensitive solid angle as determined by the spectrometer collimator (steradians).

III. RESULTS AND DISCUSSIONS

Presentations of data for gamma-ray penetration are normally presented in terms of the dimensionless function:

$$4\pi r^2 \; I(r,E_o,E,\theta)$$

where μ is the total absorption coefficient (cm^{-1}) at energy E_o. This quantity is convenient since it is dimensionless and the magnitude is near unity. The results presented here are in terms of this function, where μ has been taken as 0.0612 cm^{-1} for water, and the energy and angle spectrum of the energy flux are normalized to one disintegration yielding a 1.33- and a 1.17-Mev photon.

Figure VI shows the angle and energy spectral results obtained for r = 90 cm, and Fig. VII gives those for r = 140 cm. All data shown except those for θ = 0 and 2 deg are directly significant from the point of view of gamma-ray diffusion. The θ = 0 deg data is representative only of the attenuation in water, geometrical factors, and the resolution of the spectrometer.

Discussion of Errors

The representative errors shown on the curves of Figs. V and VI are only expressive of the statistical errors of counting. Other random errors do not influence the shape of the energy flux spectra shown. Such random errors, so called because their algebraic orientation is unknown, are summarized in Table III. These errors, if combined according to sums of squares, amount to a total random error in the normalization of the curves of about 12%.

In addition to these normalization errors, there exist difficulties which influence the shape of the resulting spectra. A 5 to 15% nonuniqueness error at the lower energies has already been described. An uncertainty of about

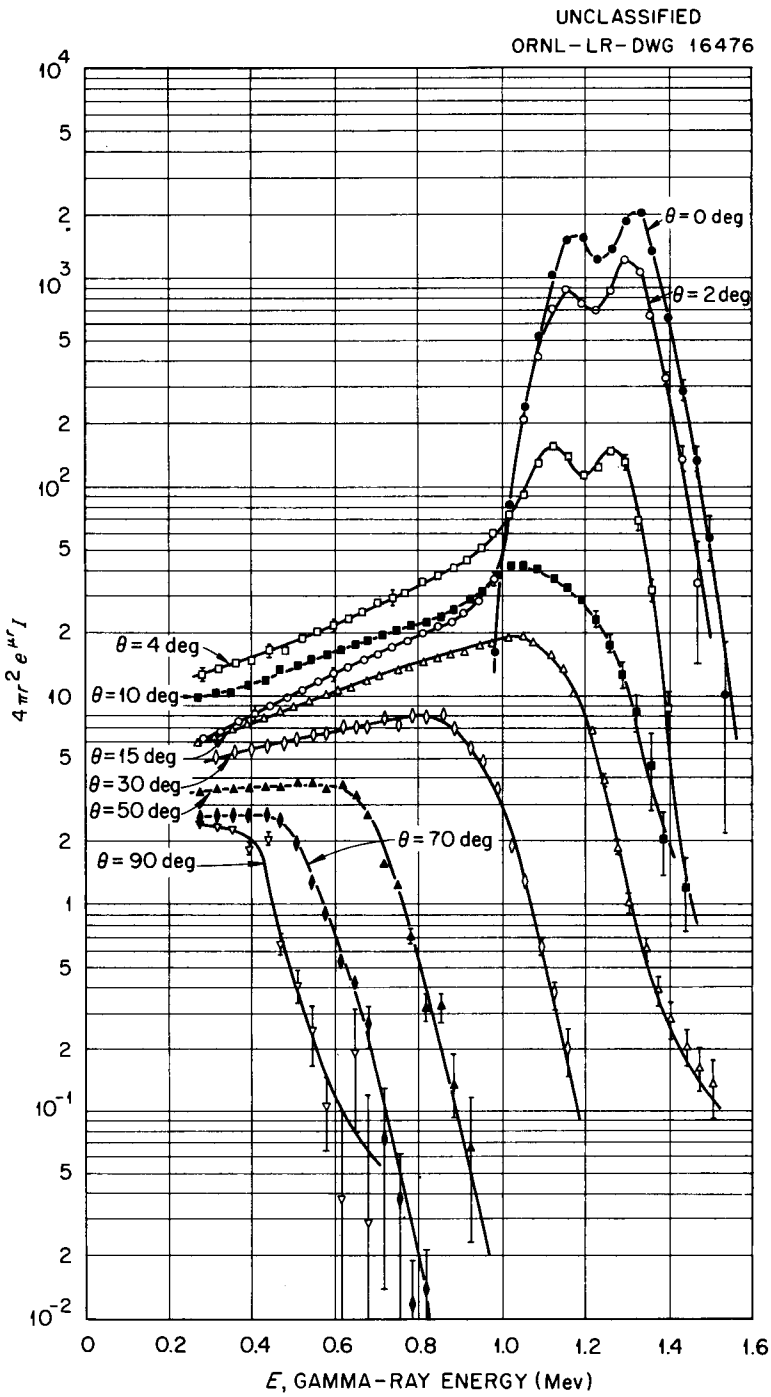

Fig. VI. Energy and Angle Spectrum of Energy Flux for a Co^{60} Source; r = 90 cm. The energy spectrum of the energy flux is shown for nine different detector angles as labeled. The energy flux is presented after multiplication by $4\pi r^2 e^{\mu r}$ in order to present a dimensionless quantity. The data for angles less than 30 deg has been smoothed out in the region below the peak by the process of non-uniqueness correction illustrated in Fig. V. The errors shown represent counting statistics.

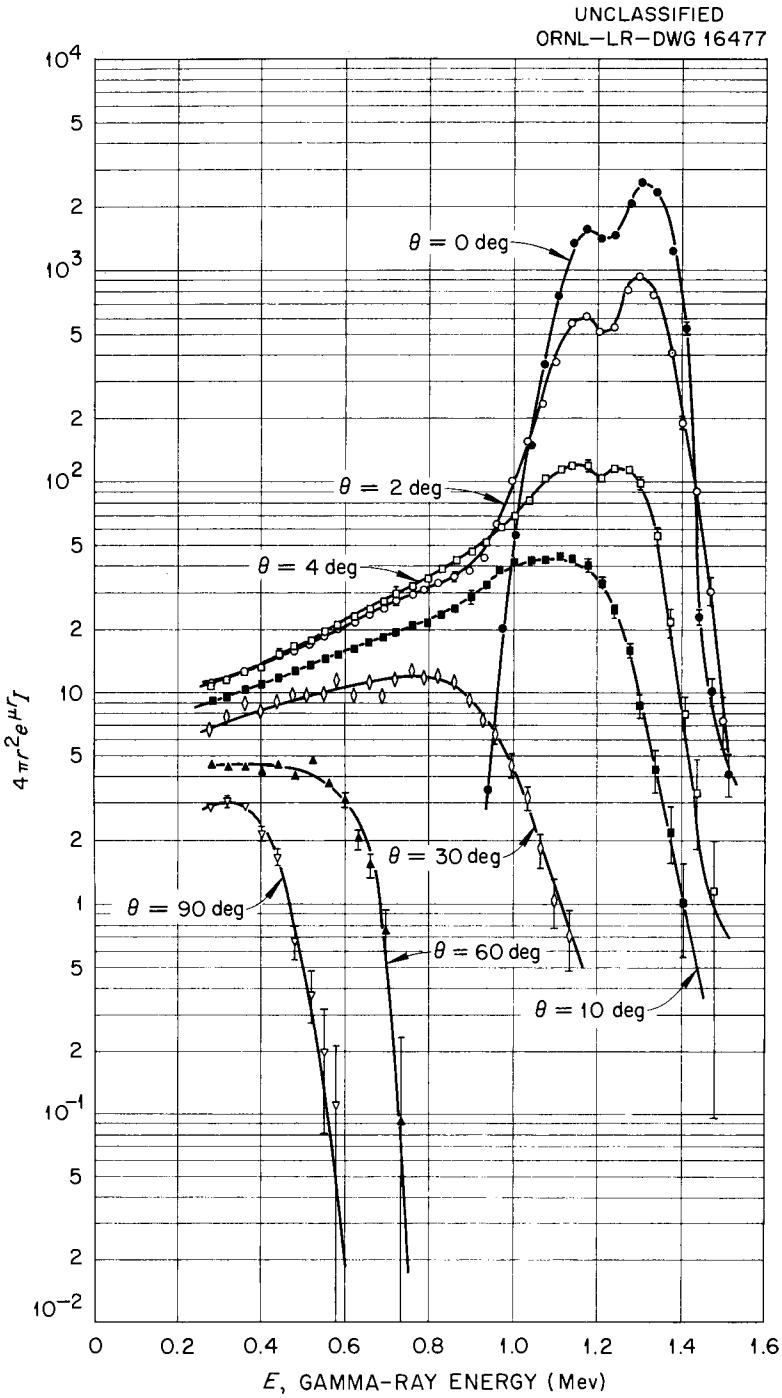

Fig. VII. Energy and Angle Spectrum of Energy Flux for a Co^{60} Source; r = 140 cm. The energy spectrum of the flux is shown for seven detector angles. For further discussion, see the caption of Fig. VI.

Table III. Estimated Standard Deviations for the Errors Induced in the Final Results

Origin of Error	Estimated Standard Deviation in Final Result (%)
Measurement of r	7
Detected photon energy	1
Source strength	8
$\bar{\epsilon}_{avg}\Omega$, average efficiency times spectrometer solid angle	7

15% is estimated to result from lack of exact knowledge of the efficiency of this spectrometer as a function of energy. This uncertainty is largely the result of the assumption of a relative spectral response unchanged from earlier experiments. The efficiency as a function of energy was not tested during the experiment described here.

The energy scale of the quoted results, both relative and absolute, has an estimated standard deviation of about 2%.

Comparison of Experimental Results for Water and Concrete Media

Figure VIII shows that the results obtained in this experiment for 5.5 mfp of water agrees favorably with those for 5.6 mfp of concrete.[9] The concrete experiment extended only to $\theta = 60$ deg. Only the shapes of the data curves in Fig. VIII may be compared, since Whyte's results were not absolutely normalized.

Comparison of Experimental and Calculated Results for a Water Medium

No calculated results are presently available for energy spectra for given detector angles. The paragraphs below discuss two comparisons which can be made by manipulation of the data of Figs. VI and VII.

Angle spectra are available for three scattered photon energies from Co^{60} in water in the early calculation of Spencer and Stinson,[2] who used hand calculations of only a few moments. Figure IX compares cross-plots of the data of Figs. VI and VII with the results of Spencer and Stinson. There is wide disagreement between the results, especially in the magnitude. If the experimental data were reduced by a factor of two, the shapes would be in good agreement for 0.256 and 0.365 Mev.

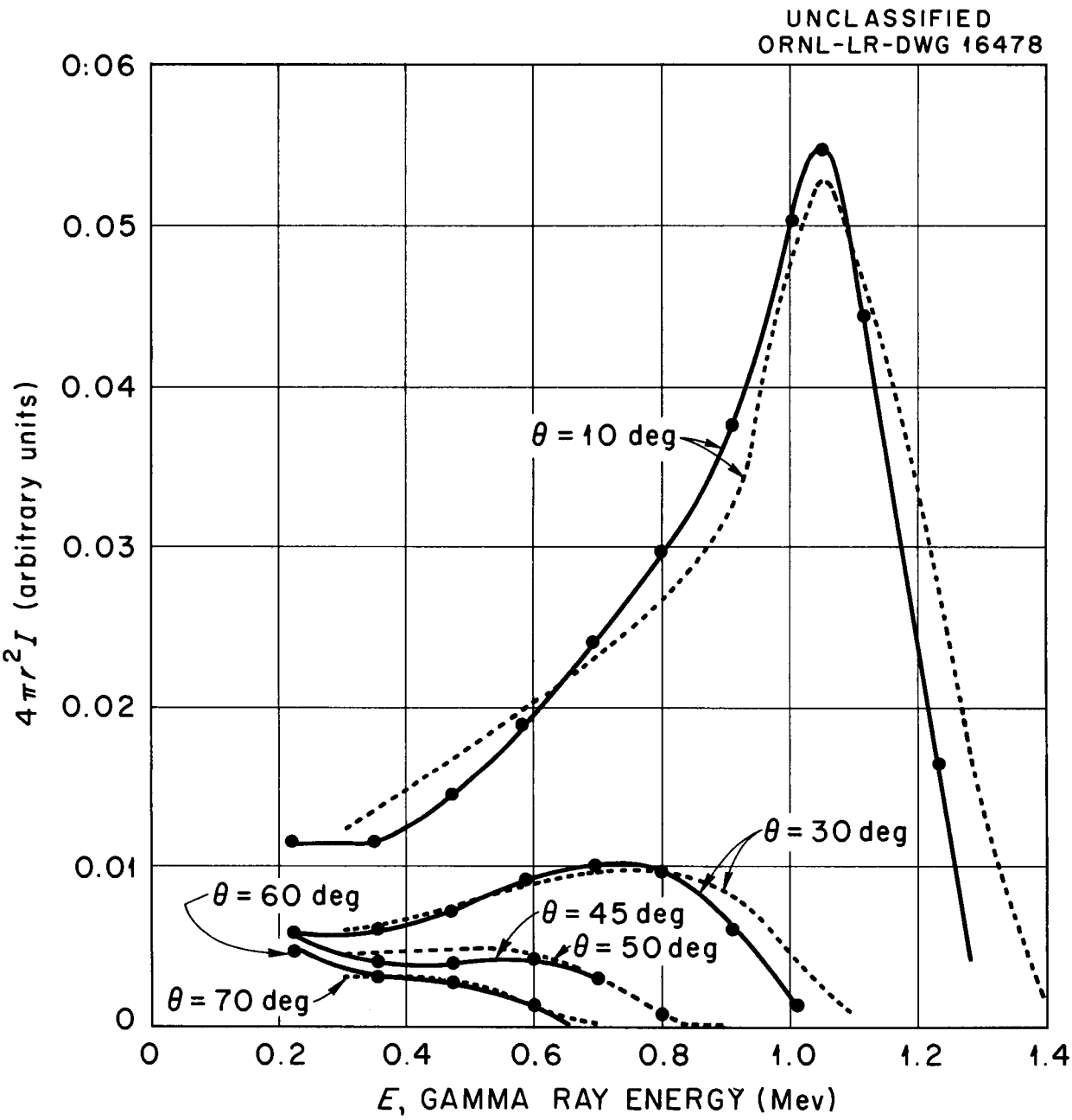

Fig. VIII. Comparison of Experimental Data (Dotted Line) for 5.5 mfp (90 cm) of Water with Data for 5.6 mfp of Concrete. These two sets of data were normalized at 30 deg since Whyte's data were not absolute.

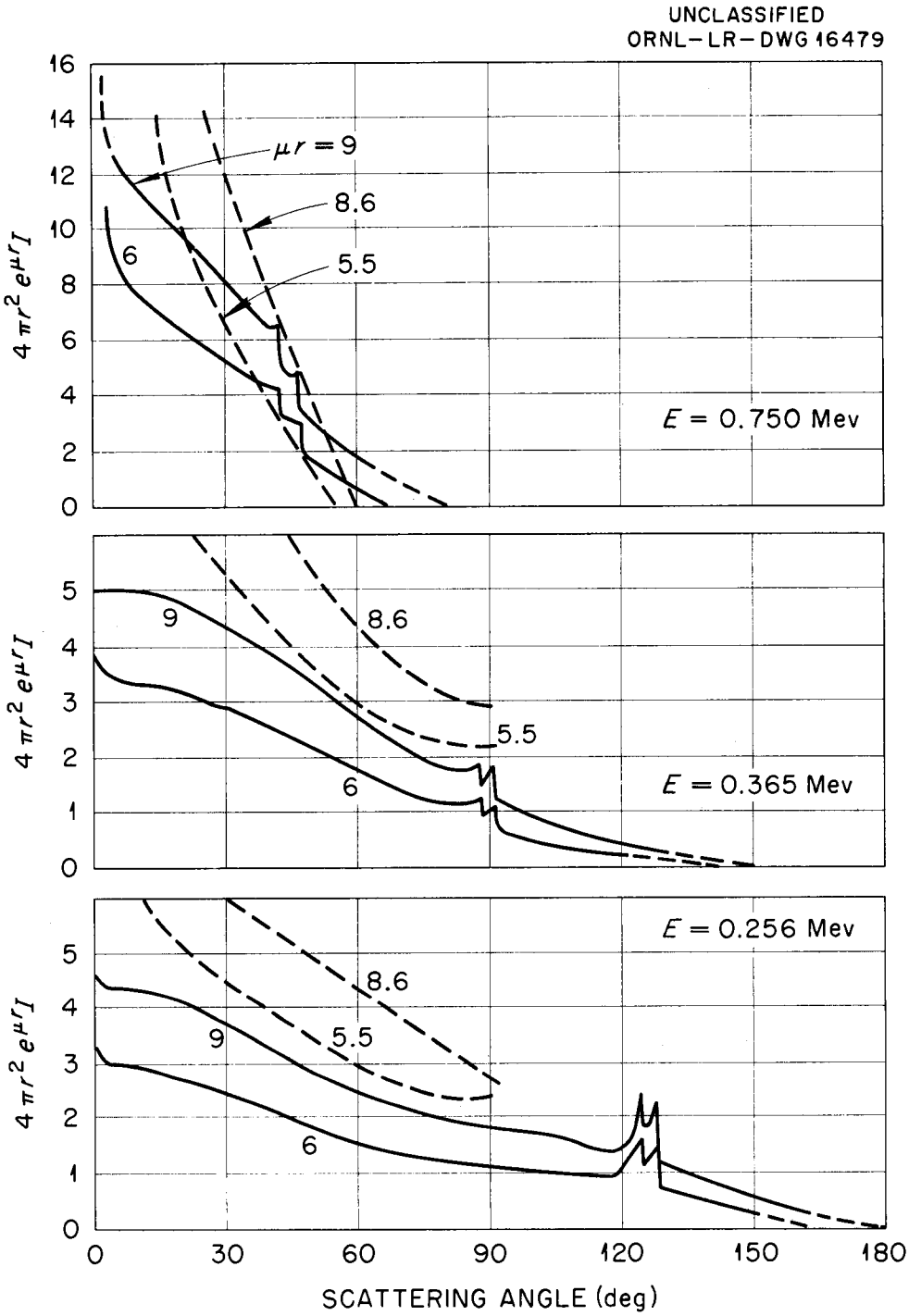

Fig. IX. Comparison of Experimental Data for Water with Calculated Data for Water. Three separate graphs are shown for each of three values of the scattered gamma-ray energy, E. The values shown on each curve refer to the number of mean free paths for 1.33-Mev radiation in water ($\mu = 0.0612$ cm^{-1}). The experimental data (dashed lines) disagree with the calculations (solid lines) by an almost constant factor of two for 0.256 and 0.365 Mev. The disagreement is not so uniform for the 0.75-Mev data.

Goldstein and Wilkins[4] present the energy spectra of the energy flux for Co^{60} gamma radiation in water as integrations over all solid angle of the differential spectra. To compare these calculated results with experiment it was necessary to perform an integration of the experimental results over the detector angle. Since only a few scattering angles were measured, interpolations were made for angles between those measured. The errors in the angle integrals induced by the interpolations can be $\sim 20\%$. Experimental observations were not possible for detector angles larger than 90 deg, although at scattered energies of less than 0.5 Mev larger angles are important to the integration. Extrapolations were made for large angles in all cases, but uncertainties of about 30% can exist for the lowest energies plotted. In addition, at energies above 1 Mev it was necessary to avoid, by extrapolation, effects of detector energy and angle resolution. This last factor is capable of introducing additional uncertainties of 20% in the integration at the highest energies.

Figure X compares the results of such integrations with the calculations of Goldstein and Wilkins.[4] It is seen that there again exists a discrepancy of about a factor of two in integrated intensity along with a very considerable difference in shape. The latter may be strongly influenced by the problems of the hand integration, but it is not likely that this error will account for the disagreement between the experiment and theory.

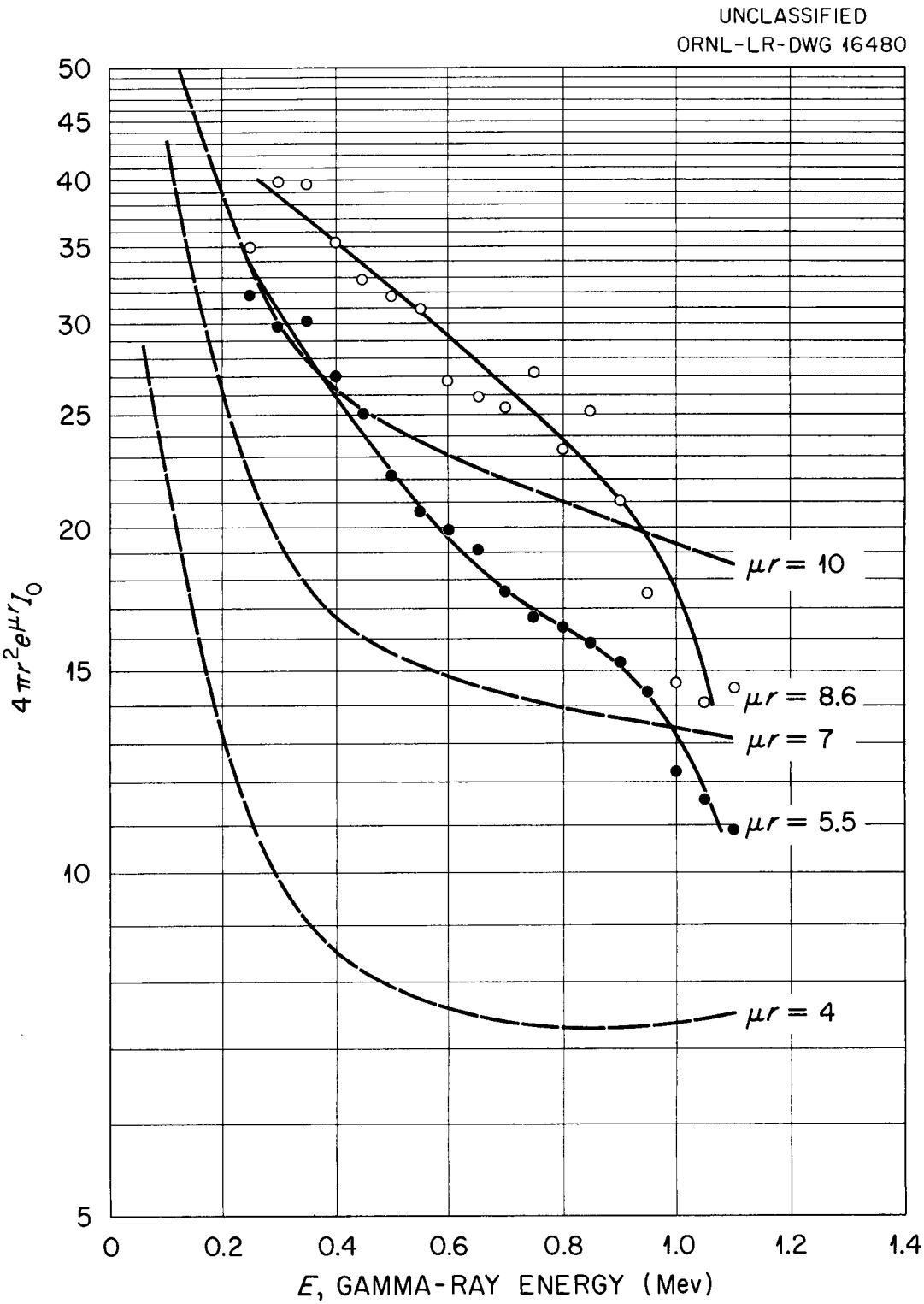

Fig. X. Comparison of the Experimental Differential Energy Flux after Integration over Angle (Solid Lines) with Calculated Values (Dashed Lines). The values shown for the various curves refer to the number of mean free paths for 1.33-Mev radiation in water ($\mu = 0.0612$ cm^{-1}).

IV. CONCLUSIONS

A considerable discrepancy in absolute magnitude seems to exist between the results of this experiment and those of calculational programs which have previously appeared to yield good results when compared to dose measurements. This discrepancy cannot be explained on the basis of known experimental uncertainties. However, when the experiment was performed it was not expected that absolute intensity results would be presented.

The shapes of the experimental spectra, after rough integration over detector angle, do not appear to agree with the calculated results of the moments method. However, this comparison of spectral shape cannot be made fairly until calculations have produced energy spectra directly comparable to experiment.

As mentioned previously, the experimental results presented agreed with those for concrete.[9] It is to be noted that Whyte's integration over angle of his normalized experimental data agreed in shape with the calculations of Goldstein and Wilkins. The only obvious explanation for this is the fact that Whyte's data extended to only 60 deg, while this experiment extended to 90 deg. Further, in the integration presented in Fig. X significant contributions for angles < 90 deg were present for energies > 0.5 Mev. These contributions were estimated on the basis of crude extrapolations.

It is planned that when additional suitable equipment becomes available, a more careful and complete experiment of this type will be performed. Both spectral and dose measurements will be made under similar conditions to improve the usefulness of the data.