CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0351156 3

ORNL-2801
Physics and Mathematics

OBSERVATION OF NONPROPORTIONALITY OF RESPONSE FOR A NaI(Tl) SCINTILLATION CRYSTAL

R. W. Peelle
T. A. Love

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

Printed in USA. Price 50 cents. Available from the

Office of Technical Services
Department of Commerce
Washington 25, D.C.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

CRL.
Cy 4

ORNL-2801 ERRATA
(Internal Distribution Only)

Page iii, line 8: Substitute "light" for "pulse."
Substitute "crystal" for "phototube."

Page 1, lines 5-8: Delete sentences in parentheses.

Page 4, lines 9-10: Lines should be revised to read as follows:

.....absorbed, and if the first dynode of the photomultiplier receives a number of photoelectrons proportional to the.....

Page 6, Table 2, column 6, 2nd line: Replace "2454" with "2754."

ORNL-2801

Copy_____

Contract No. W-7405-eng-26

Neutron Physics Division

OBSERVATION OF NONPROPORTIONALITY OF RESPONSE FOR A NaI(Tl) SCINTILLATION CRYSTAL

R. W. Peelle and T. A. Love

Date Issued

OCT 20 1959

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES

3 4456 0351156 3

ABSTRACT

By observing "sum" pulse-height distributions which correspond to the simultaneous detection of cascade gamma-rays, the nonproportionality of response of a 3-in.-dia by 3-in. NaI(Tl) crystal was briefly studied in a manner which did not depend critically on the performance of the pulse-height discriminator system. The sum peaks occurred at pulse heights higher than would have been expected on the hypothesis that the integrated ~~pulse~~ * light output of the ~~phototube~~ * crystal was proportional to the absorbed gamma-ray energy. This difference corresponded to an average of (30 ± 6) kev for the experiments performed using the cascade gamma-ray transitions of Y^{88} and Bi^{207}.

* See errata

INTRODUCTION

Uncertainty exists for NaI(Tl) scintillators concerning the detailed linearity or proportionality* of light output to absorbed gamma-ray energy. This uncertainty prompted a study of proportionality in one NaI(Tl) crystal, which was carried out in conjunction with gamma-ray energy measurements reported elsewhere.[1] ~~(It is assumed here that photomultiplier tubes are an integral part of the scintillator and that they yield charge-output strictly proportional to light production in the phosphor.)~~ It is commonly observed that over reasonably large energy ranges the light output response appears linear, or at least that, within the precision of measurement possible, nonlinearity is not observed in the region above a few hundred kilovolts. The linearity of response in the low-energy region, and consequently the proportionality at higher energies, is in considerably more doubt. Some authors have reported that the response of a NaI(Tl) crystal to gamma rays is proportional,[2] while others have found a fairly linear response but a negative

---

*There is some confusion apparent in the literature concerning the meanings of "linearity" and "proportionality." In this paper, the following definitions are used: Scintillator response is linear in a given energy region if the integrated pulse output from the phototube (C) is related to the gamma-ray energy (E) by a linear equation $C = kE + b$, where k and b are constants. Response is proportional if $b = 0$.

1. R. W. Peelle and T. A. Love, Scintillation Spectroscopy Measurements of Gamma-Ray Energies from Sources of Y^{88}, Mn^{54}, and Zn^{65}, ORNL-2790 (1959).

2. W. E. Mott and R. B. Sutton, Handbuch der Physik (Encyclopedia of Physics), edited by S. Flügge, Vol 45, p. 86-173, see esp. pp. 99-102, Springer-Verlag, Berlin, 1958. C. J. Taylor et al., Phys. Rev. 84, 1034 (1951).

* See errata

extrapolated intercept between 15 and 30 kev (Ref. 3). In all cases the results have depended heavily on the behavior of the pulse analysis equipment, and errors are seldom quoted. The existence of these uncertainties is natural, because linearity measurements are quite difficult to perform with precision over a large dynamic range, and statements concerning proportionality depend upon identification of the recorded pulse height which corresponds to zero absorbed energy in the crystal. Both the "zero" and the linearity measurements depend upon results obtained using precisely known pulses from an artifical source, but the experimenter has difficulty in showing that the analysis equipment reacts to these test pulses in the same way that it does to the "natural" pulses from the NaI(Tl)-phototube combination.

The work presented here consisted in the measurement of "sum" peaks corresponding to the simultaneous detection of cascade gamma rays in a NaI(Tl) crystal. Y^{88} and Bi^{207} sources were positioned very close to the crystal, and the apparent energy corresponding to each sum peak was compared to the known sum of the energies of the individual members of the cascade. This method allowed a study of proportionality independent of the zero adjustment of the pulse-height analyzer, and not sensitively dependent upon the linearity of the analysis system if a calibration energy is available close to the sum energy of the cascade.

METHOD

The study of sum peaks described here was performed at the same time

3. D. Engelkemeir, Rev. Sci. Instr. 27, 589 (1956); see also Mott and Sutton, op. cit.

as the measurements of gamma-ray energies from sources of Y^{88}, Mn^{54}, and Zn^{65} described elsewhere.[1] Therein is described in detail the standard pulse-handling equipment used, the precautions taken, and the general methods of data analysis. The chief precautions were designed to reduce any effects of equipment drift, pulse-height analyzer nonlinearity, interfering effects between pulse-height distributions from neighboring gamma rays, etc. The energies of unknown photopeaks were determined by linear interpolation (or extrapolation) using two nearby known energies of standard sources employed in the same experiment. The energy standards used are listed in Table 1.

Table 1. Energies of Calibration Sources

Source	Energy (kev)	References
Bi^{207}	569.6 ± 0.1	Way *et al.*[a]
	1063.7 ± 0.3	Way *et al.*
Na^{24}	1368.0 ± 1.0	Hedgran and Lind[b]
	2753.5 ± 1.0	Hedgran and Lind
La^{140}	1596 ± 2	Hedgran and Lind[b], Cork *et al.*[c]
Y^{88}	898.8 ± 1.2	Peelle and Love[d]
	1840.0 ± 2.0	Peelle and Love[d]

a. K. Way *et al.*, *Nuclear Data Sheets*, NCR 58-2-66, National Academy of Sciences, National Research Council.

b. A. Hedgran and D. Lind, *Arkiv Fysik* 5, 177 (1952).

c. J. M. Cork, *et al.*, *Phys. Rev.* 83, 856 (1951).

d. R. Peelle and T. A. Love, *Scintillation Spectroscopy Measurements of Gamma-Ray Energies for Sources of Y^{88}, Mn^{54}, and Zn^{65}*, ORNL-2790 (1959).

Studies of nonproportionality were performed in the following manner. A weak source of a radioactive substance which emits two gamma rays in cascade (Bi^{207} or Y^{88}) was placed close to one end of a 3-in.-dia by 3-in. NaI crystal.* Both gamma rays from a reasonable share of the cascades simultaneously lost all their energy in the crystal, giving a "sum" photopeak which has an apparent energy (E_a) determined by a calibration using gamma-ray photopeaks arising from individual interactions in the crystal. If the light-production in NaI(Tl) is proportional to the energy absorbed, and if the first dynode of the photomultiplier ~~yields pulses~~ receives a number of photoelectrons proportional to the amount of light produced, then the apparent energy of the sum peak should be, within error, the sum of the energies of the two gamma rays which constitute the cascade. If not, then the excess is presumably a function of the individual energy components of the sum** and perhaps of properties of the scintillation crystal which are associated with any nonproportionality. If the light production phenomena associated with each gamma ray can be considered independent, then the observed excess represents the nonproportionality of that crystal in usual operation. This excess can be defined as follows, omitting explicit dependence upon unidentified variables:

$$D(E_1,E_2) \equiv E_a - E_1 - E_2, \text{ with error } \sigma(D),$$

---

*Supplied in a standard mounting by the Harshaw Chemical Company of Cleveland, Ohio.

**If the response is perfectly linear throughout the energy region which includes the possible components of the cascade, then the excess is independent of the energies of the components.

* See Erata

where

D = "nonproportionality excess energy" (kev),

$E_a \pm \sigma(E_a)$ = apparent energy of sum peak determined on the basis of the two closest calibration lines (refer to Eqs. 1 and 2 in Ref. 1),

E_1, E_2 = known energies of the two cascade gamma rays simultaneously detected to produce each sum pulse.

The errors in the above quantities were determined by a straightforward combination of experimental errors in the manner described in Ref. 1, assuming that all errors are random and representable by normal error distributions.

RESULTS

Table 2 shows the results obtained in three experiments* to determine the apparent energy of sum peaks. The third case, illustrated in Fig. 1, involved the well-known cascade in the decay of Bi^{207}. The results strongly suggest a lack of proportionality which is in qualitative agreement with that observed by the other authors who have noted a nonproportionality. If the response is considered linear in the region of calibration (energies > 0.5 Mev), this excess is just equal to the negative of the energy-axis intercept of the linearly extrapolated pulse-height vs energy curve which would have been observed using a pulse-height analyzer having output strictly proportional to the pulse output of the photomultiplier.

---

*These experiments are numbered 1, 2, and 10, respectively, in Ref. 1.

Table 2. Summary of Measurements on Nonproportionality Excess

Emitter of Cascade	Energies of Cascade Gamma Rays[a] (kev)		E_a, Apparent Sum Energy (kev)	D, Nonproportionality Excess (kev)	Nearest Calibration Energy (kev)
	E_1	E_2			
Y^{88}	1840	899	2765.6 ± 5.5	26.8 ± 6.2	2754 (Na^{24})
Y^{88}	1840	899	2762.5 ± 3.7	23.7 ± 4.2	2754 (Na^{24})
Bi^{207}	1063.7	569.6	1677.0 ± 6.0	43.7 ± 6.0	1596 (La^{140})
				$\bar{D}$ = 30 ± 6[b]	

a. See Table 1.
b. If D is assumed independent of E_1 and E_2 (light output truly linear with energy in the range of measurements), a weighted average of the three values obtained from this 3-in.-dia by 3-in. crystal is (30 ± 3) kev. A chi-square test indicates a probability of 2% that a larger value of chi-square would have been observed if the three results had been consistent. The quoted error of 6 kev is based on the scatter between the values. There is, however, little reason to assume that precise linearity of light output exists over this range of energies, so D may well be a function of E_1 and E_2.

CONCLUSIONS

At least one 3-in.-dia by 3-in. NaI(Tl) crystal showed a nonproportionality of response demonstrated in an experiment which was not influenced by the zero of the pulse-height analysis system. Sum peaks from the simultaneous detection of cascade gamma rays from Y^{88} and Bi^{207} sources were observed to fall at pulse heights greater than would have been expected on the basis of proportionality of response by an average amount corresponding to (30 ± 6) kev. It is felt that this method avoids pitfalls present in other methods used to study the pulse-height response of NaI(Tl) to gamma rays and that it should be used to measure nonproportionality for a range of gamma-ray energies and for scintillators with a variety of properties.

* See errata

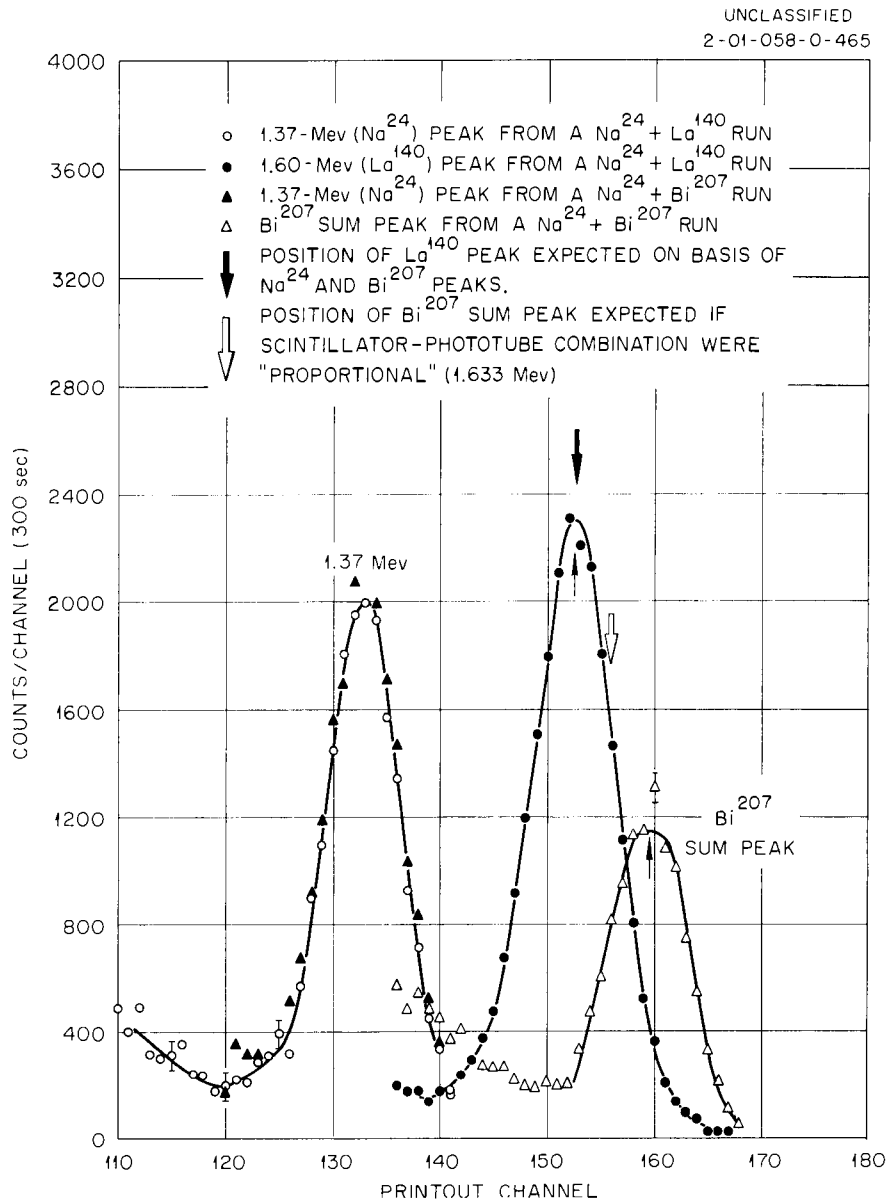

Fig. 1. Illustration of Nonproportionality of Response to the Gamma-Ray Cascade in the Decay of Bi^{207} for a 3 × 3-in. NaI(Tl) Crystal. The circled points were obtained from a run in which a combined source of La^{140} and Na^{24} wos used, while the triangular points are from a run in which Bi^{207} and Na^{24} were combined as the source. In all cases appropriate subtractions were performed to remove distortions caused by interfering pulse-height distributions. The figure shows that the dota from the two runs for the Na^{24} 1.368-Mev line overlap almost perfectly, indicating that drift between the runs was unimportant. The 1.6-Mev peak of La^{140} appears in the expected position within a small fraction of a channel, as indicated by vertical arrows. The "sum" peak from the cascade gammo rays in Bi^{207}, however, falls almost four channels from the position it would have if the crystal-phototube combination had proportional response. This discrepancy is noted with vertical arrows. The weak 1.77-Mev gamma ray of Bi^{207} interfered with the sum peak which has an apparent energy of 1.68 Mev. The plot shown was produced by subtracting two normalized Bi^{207} spectra, one with the source adjacent to the crystol and one with a source a few centimeters away. The difference between the two allowed exhibition and analysis of the sum peak.

ORNL-2801
Physics and Mathematics
TID-4500 (15th ed.)
August 1, 1959

INTERNAL DISTRIBUTION

1. C. E. Center
2. Biology Library
3. Health Physics Library
4-5. Central Research Library
6. Reactor Experimental Engineering Library
7-26. Laboratory Records Dept.
27. Laboratory Records, ORNL R.C.
28. A. M. Weinberg
29. L. B. Emlet (K-25)
30. J. P. Murray (Y-12)
31. J. A. Swartout
32. E. H. Taylor
33. E. D. Shipley
34. A. H. Snell
35. E. P. Blizard
36. M. L. Nelson
37. W. H. Jordan
38. R. A. Charpie
39. S. C. Lind
40. F. L. Culler
41. A. Hollaender
42. J. H. Frye, Jr.
43. M. T. Kelley
44. J. L. Fowler
45. R. S. Livingston
46. K. Z. Morgan
47. T. A. Lincoln
48. A. S. Householder
49. C. P. Keim
50. C. S. Harrill
51. C. E. Winters
52. D. S. Billington
53. H. E. Seagren
54. D. Phillips
55. A. J. Miller
56. M. J. Skinner
57. R. R. Dickison
58-67. R. W. Peelle
68-72. T. A. Love
73. F. C. Maienschein
74. L. B. Holland
75. F. L. Keller
76. A. D. Callihan
77. F. J. Muckenthaler
78. C. D. Zerby
79. N. H. Etheridge
80. R. R. Coveyou
81. L. Jung
82. R. B. Murray
83. E. G. Silver
84. A. Simon
85. D. K. Trubey
86. W. Zobel
87. G. T. Chapman
88. G. deSaussure
89. E. B. Johnson
90. K. M. Henry
91. G. Estabrook
92. C. A. Preskitt
93. F. L. Friedman (consultant)
94. H. Goldstein (consultant)
95. H. Hurwitz, Jr. (consultant)
96. L. W. Nordheim, Jr. (consultant)
97. R. F. Taschek (consultant)
98. ORNL-Y-12 Technical Library, Document Reference Section

EXTERNAL DISTRIBUTION

99-708. Given distribution as shown in TID-4500 (15th ed.) under Physics and Mathematics Category (75 copies - OTS)
709. Division of Research and Development, AEC, ORO