Nonproportionality in the scintillation light yield of bismuth germanate

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Abstract

We present measurements of nonproportionality in the scintillation light yield of bismuth germanate (BGO) for gamma-rays with energies between 6 keV and 662 keV. The scintillation light was read out by avalanche photodiodes (APDs) with both the BGO crystals and APDs operated at a temperature of ≈ 90 K. Data were obtained using radioisotope sources to illuminate both a single BGO crystal in a small test cryostat and a 12-element detector in a neutron radiative beta-decay experiment. In addition one datum was obtained in a 4.6 T magnetic field based on the bismuth K x-ray escape peak produced by a continuum of background gamma rays in this apparatus. These measurements and comparison to prior results were motivated by an experiment to study the radiative decay mode of the free neutron. The combination of data taken under different conditions yields a reasonably consistent picture for BGO nonproportionality that should be useful for researchers employing BGO detectors at low gamma ray energies.

Keywords: avalanche photodiode, bismuth germanate, nonproportionality, photon detection, radiative neutron decay

29 **1. Introduction**

30 For an ideal scintillator, the number of optical photons produced by a gamma
 31 ray is linearly proportional to the energy deposited in the scintillator by the
 32 gamma ray. However, many scintillators exhibit nonproportionality in their
 33 response, especially for gamma-ray energies below 100 keV [1]. Nonproportion-
 34 ality is typically defined by the ratio of the scintillation yield for a particular
 35 energy deposit to the scintillation yield due to complete energy deposit for a
 36 661.7 keV gamma ray from a ^{137}Cs source. Measurements of nonproportionality
 37 in the response of bismuth germanate (BGO) in the following energy ranges
 38 have been reported: 4.5 keV to 662 keV [2], 6 keV to 1332 keV [3], 14 keV to
 39 1330 keV [4], 60 keV to 662 keV [5], and 11 keV to 100 keV [1]. We report new
 40 measurements in the energy range from 6 keV to 662 keV.

41 Our measurements and comparison to prior results were motivated by an
 42 experiment to study the radiative decay mode of the free neutron. We reported
 43 the first observation of this process in a pilot experiment in which gamma-rays
 44 were detected by a single BGO crystal [6, 7]. To accurately test theoretical pre-
 45 dictions for the photon energy spectrum, a higher precision experiment has been
 46 performed with a 12-element BGO detector [8]. Analysis of this experiment re-
 47 quires both Monte Carlo modeling of the energy deposit spectrum in the BGO
 48 crystals and understanding of the light yield from BGO. For our minimum de-
 49 tectable gamma ray energy deposit of ≈ 5 keV, the nonproportionality for BGO
 50 is an important source of uncertainty. Hence we performed measurements to
 51 determine the nonproportionality observed for our crystals, temperature, elec-
 52 tronics, and other operating conditions. Our results were determined by both
 53 tests on a single BGO crystal with radioactive sources in a small test cryostat
 54 and with radioactive sources illuminating a 12-element detector in our apparatus

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for neutron beta-decay. In addition, the multi-element detector allowed for an additional measurement approach in which for any given crystal, a continuum of background gamma rays incident on the other 11 crystals produced escaping bismuth K x-rays that are detected by that given crystal. This particular determination of nonproportionality at the Bi K x-ray energy (mean of 78.7 keV) was performed in a magnetic field of 4.6 T. As in prior published studies, we determined the nonproportionality by comparing the center of the peak in the pulse height spectrum for a given gamma-ray energy deposit E , C_E , to that expected based on linear scaling from the center for the 661.7 keV gamma rays from a ^{137}Cs source, $C_{661.7}$. This definition applies for sufficiently high photoelectron yields so that a symmetric peak is obtained, which applies to all of our results except the ^{55}Fe source results presented at the end of the paper. For this definition, the measured nonproportionality N_E at energy E expressed in percent is given by

$$N_E = 100 \frac{C_E}{(E/661.7) C_{661.7}} \% \quad (1)$$

Hence for an ideal scintillator, N_E is unity for all energies. In Sec. 2, we describe the apparatus we employed for our measurements of nonproportionality. In addition to presenting our results in Sec. 3, we compare to previous reports. The combination of data taken under different conditions yields a consistent picture for BGO nonproportionality that should be useful for researchers employing BGO at low gamma ray energies. In Sec. 4 we present results for data obtained in the test cryostat with 6 keV x-rays from an ^{55}Fe source and in Sec. 5 we summarize the paper.

2. Apparatus and measurements

2.1. Test cryostat measurements

The test cryostat apparatus has been previously described in its use for studies of APD response [9]. For these nonproportionality studies, a 12 mm by 12 mm by 200 mm BGO crystal [10] coupled to an APD with an active area of 13.5

82 mm by 13.5 mm [11] was thermally sunk to the bottom of the inner reservoir of
 83 a cryostat (see Fig. 1). As discussed in Ref. [8], the opposite end of the BGO
 84 crystal was painted with reflective paint to improve the light collection efficiency.
 85 The long sides of the BGO crystal were covered by two layers of aluminized My-
 86 lar (total of 12.4 μm Mylar and 0.065 μm aluminum). The long-side regions near
 87 the end faces (≈ 12 mm) had additional layers of polytetrafluoroethylene (PTFE)
 88 tape and aluminized Mylar for mechanical protection. (≈ 300 μm PTFE and 6
 89 μm Mylar with 0.033 μm aluminum). The cryostat's vacuum jacket included a
 90 3.5 cm diameter tube with a 7 cm diameter metal-sealed flange through which
 91 we illuminated the crystal about midway along its 200 mm length (see right side
 92 of Fig. 1). The light collection efficiency for gamma rays incident at different
 93 locations along this crystal can vary by ≈ 8 % from the center, with greater
 94 light collection efficiency obtained at either end of the crystal. This geometric
 95 effect is independent of the incident energy [8]. For these nonproportionality
 96 tests, the tubing for the vacuum port and the cryostat itself provided sufficient
 97 collimation to ensure that only the central region of the crystal was illuminated.
 98 For the highest energy (thus most penetrating) source, this was confirmed by
 99 a check with additional collimation that consisted of 1.2 cm thick, 2.8 cm tall
 100 copper pieces clamped in place above and below the horizontal 3.5 cm tube.

101 An oil-free turbomolecular pumping station was used to reduce the pressure
 102 in the chamber to a typical base pressure of on the order of 10^{-6} mbar. Upon
 103 filling the cryostat with liquid nitrogen, the typical pressure was on the order
 104 of 10^{-7} mbar. The APD signal was sent to a charge-sensitive preamplifier[12],
 105 followed by an amplifier set for a gain of 100 and a 6 μs shaping time constant.
 106 The output of the shaping amplifier was registered by a multichannel analyzer
 107 (MCA)[13]. The lower level discriminator of the MCA was typically set slightly
 108 below the detector noise level at ≈ 10 channels, for a 1000-channel full range of
 109 ≈ 1000 keV. The APD was operated at a typical bias of 1380 V (≈ 40 V below
 110 breakdown). Using studies with a precision pulser, we found that our electronics
 111 for the test cryostat apparatus exhibited a slight nonlinearity that was consistent
 112 with an offset of one channel out of 1000 in the MCA. To determine the BGO

nonproportionality, we corrected for this nonlinearity, which yielded a shift of 4.3 % for our lowest energy datum at 23.3 keV, decreasing to 1 % at 60 keV.

A thin polyimide window in the side port allowed the sources to be outside the cryostat vacuum space so they could be easily manipulated. The gamma-ray energies and sources employed in the test cryostat apparatus are listed in Table 1. A ^{137}Cs source was used to provide the 662 keV reference. The mean energy deposit listed was determined for multiple-line emissions from the following components in keV, using the weighting percentages [14] in parentheses: ^{109}Cd - 22.1, 25.1 (83.7*0.026, 17.7*0.078); ^{133}Ba - 30.6, 31.0, 35.1, 35.9 (54, 100, 29, 7); ^{153}Gd - 97.4, 103.2 (29, 21); and ^{57}Co - 122.1, 136.5 (85.5, 10.7). For the low energy lines of the ^{109}Cd source, the weighting included the factors shown to account for energy-dependent transmission through a 0.25 mm thick stainless steel cover on this particular source. To evaluate the nonproportionality, the pulse height spectrum for a given emission was fit with a single Gaussian, except for the multiple-line emissions noted above for which multiple-line Gaussians were employed. In the case of multiple-line emission, a single nonproportionality value was used for all the Gaussians and is listed for the mean energy deposit.

Fig. 2 shows the background-subtracted pulse height distributions for three of the seven sources. The background was determined by counting without the source. However, we found that the apparent threshold was higher with a source than without a source and it varied between sources. The response observed near the 10 keV threshold is due primarily to this source-related response, with a smaller contribution from imperfect background subtraction.

2.2. Measurements in neutron beta-decay apparatus

The apparatus for neutron beta-decay has been described elsewhere [6–8]. In brief, a neutron beam passes through the cylindrical bore of a superconducting magnet and gamma rays emitted during neutron beta decay are detected by an annular array of twelve 12 mm by 12 mm by 200 mm BGO crystals. As in the test dewar, each crystal is coupled to an APD with an active area of 13.5

143 mm by 13.5 mm. However, the electronic processing and spectrum analysis were
 144 different than that performed for the test dewar studies. The preamplifier output
 145 signals were directly fed into two 8-channel GaGe cards [15], which were set to
 146 digitize the signals with 40 ns per channel resolution in 80 μ s long traces. The
 147 decay time constant of the preamplifiers is 50 μ s. The amplitude of the digitized
 148 signals (see Ref. [8] for an example) were determined by fitting the signals to a
 149 template waveform. The template was a model waveform constructed from the
 150 average of preamplifier signals for gamma rays with a range of energies.

151 To test the detector response with radioactive sources, we employed a reen-
 152 trant tube that allowed the sources to be outside the magnet's vacuum chamber
 153 while illuminating the liquid nitrogen-cooled detector in a similar way as the
 154 photons from neutron decay. These tests were performed with the apparatus
 155 off the neutron beam line and with the magnetic field off. Pulse height spectra
 156 with ^{137}Cs , ^{133}Ba , ^{57}Co , and ^{241}Am sources are shown in Ref. [8], along with
 157 the results of modelling the detector response based, in part, on MCNP5 [18]
 158 predictions for the energy deposit spectrum in each BGO crystal. Nonpro-
 159 portionality was not included in the modeling. For this paper, we determined
 160 nonproportionality by comparing the location of emission lines from sources to
 161 those determined from Geant4 [16] and MCNP5 modelling with the assumption
 162 that the nonproportionality was 100 % for all energy deposits. A ^{137}Cs source
 163 was used to provide the 662 keV reference line for lines from a ^{133}Ba (nominally
 164 31 keV, 81 keV, and 356 keV) and a ^{57}Co source (122 keV and an X-ray escape
 165 peak at 43 keV).

166 An additional datum was obtained using x-ray escape from continuum back-
 167 ground gamma-rays detected during operation on the neutron beam line. Al-
 168 though nonproportionality may depend weakly on the energy deposition process[17],
 169 in Ref. [4] the values obtained with escape peaks were the same within uncer-
 170 tainties as those obtained from direct sources. For a given BGO crystal, escape
 171 of Bi K x-rays from all the other crystals yields a peak at a mean energy of
 172 78.7 keV. If the pulse height spectra from all the crystals are summed, this peak
 173 disappears because detection of the x-ray only occurs when the x-ray escaped

174 from a different crystal. To improve the extraction of the peak location, we
 175 combined all the data from the 12 crystals but also subtracted off the summed
 176 spectrum. In this case, the magnetic field was 4.6 T. The gamma-ray energies
 177 and sources employed in the neutron radiative decay (RDK) apparatus are listed
 178 in Table 2.

179 3. Results

180 Information on our results and those from past literature are summarized in
 181 Table 3. Whereas most results were obtained with a variety of radioisotopes, the
 182 work in Ref. [1] employed monochromatic gamma-ray beams from a synchrotron
 183 with resolutions varying from 2 eV at 9 keV to 20 eV at 100 keV. In Ref. [4] a
 184 photomultiplier tube (PMT) was used for measurements at room temperature
 185 and an APD [19] was used for measurements near liquid nitrogen temperature,
 186 with similar results being obtained for the nonproportionality. In addition to
 187 direct gamma ray lines, nonproportionality was determined using escape peaks,
 188 again with similar results.

189 Fig. 3 shows data for nonproportionality in the response of bismuth ger-
 190 manate (BGO). Our results from the test cryostat and neutron beta-decay ap-
 191 paratus are listed in Tables 1 and 2, respectively. The combination of our data
 192 and prior published results include a variety of crystal sizes and sources, op-
 193 tical readout methods, temperatures, radiation sources, and analysis methods.
 194 Nevertheless, all of the results generally agree within a few percent. The results
 195 from our test cryostat data are closest to the results of Ref. [1], whereas the
 196 results from Ref. [4] and our RDK apparatus are typically a few percent higher,
 197 in particular just below the Bi K-edge at ≈ 90 keV. The single datum obtained
 198 at the bismuth K x-ray energy at a 4.6 T magnetic field is consistent within un-
 199 certainties with the nonproportionality values obtained at near zero magnetic
 200 field.

201 4. Nonproportionality at 6 keV

202 Ref. [2] reported a monotonic increase in N_E with decreasing energy deposits
 203 in the range below the Bi L edge at ≈ 13 keV. Whereas the expected increase
 204 in the vicinity of the L-edge was reported in Refs. [1] and [3], N_E was found to
 205 continue decreasing further below the edge region. This discrepancy motivated
 206 us to extend our nonproportionality measurements as low as possible in photon
 207 energy, hence we illuminated a 1 cm³ BGO crystal with 6 keV x-rays from an
 208 ⁵⁵Fe source with an activity of ≈ 9 mCi. We obtained the best signal to noise
 209 ratio for observing these low amplitude pulses by operating the APD at between
 210 5 V and 15 V below breakdown. For these data, the amplifier gain was 116, the
 211 shaping time constant was 6 μ s, and the lower level discriminator of the MCA
 212 was 13 channels, for a 1000-channel full range of ≈ 100 keV. Under these condi-
 213 tions, we obtained the background-subtracted pulse-height spectrum shown in
 214 Fig. 4. To avoid extremely low channels on the MCA, the nonproportionality
 215 was determined with respect to the 59.5 keV peak from the ²⁴¹Am source and a
 216 correction was applied based on our nonproportionality value for this line. Be-
 217 cause of the peak asymmetry, we fit the data to a gamma distribution[20] (see
 218 Fig. 4). Evaluation of the mean value yielded $N_{6.0} = (69 \pm 3) \%$, which corre-
 219 sponds to 1.4 times greater than that would be obtained based on the observed
 220 peak location.

221 An asymmetric spectrum would be expected from Poisson statistics for a
 222 yield of only a few photoelectrons or less. In this case APD noise might mask the
 223 appearance of resolved peaks for specific photoelectron numbers. We modelled
 224 the pulse height spectrum using Poisson statistics and obtained the best results
 225 for a photoelectron yield of 2.6. However, an estimate of the photoelectron yield
 226 is not consistent with such a low value. For our large crystals we had estimated
 227 the photoelectron yield to be ≈ 1000 for 662 keV gamma rays[8]. For 6 keV and
 228 $N_{6.0} = 100 \%$ this estimate corresponds to 9 photoelectrons, but the observed
 229 nonproportionality reduces the estimated yield to 6 photoelectrons. In addition,
 230 the small crystal employed for the ⁵⁵Fe studies provided a higher yield. Hence

the substantial asymmetry observed might indicate a pulse height distribution with a variance greater than its mean, due to APD noise. The impact of the value of the yield is on the probability of zero photoelectrons, which yields a small correction to the observed nonproportionality. Based on our expected yield the correction is negligible. However, we cannot discount the possibility that the yield is ≈ 2.6 photoelectrons, which would result in a probability of zero photoelectrons of $\exp(-2.6)=0.074$. From these considerations we assign an asymmetric one-sigma uncertainty interval, hence our final value for the nonproportionality at 6.0 keV is $N_{6.0} = (69^{+3}_{-8})$ %. Our result falls midway between the values reported in Refs. [1] and [3]. In Ref. [3] it was suggested that the difference between their results and those of Ref. [2] could be related to the surface state of the investigated crystals.

5. Conclusions

Motivated by an experiment to study neutron radiative beta-decay, we performed new measurements of the nonproportionality in the light yield of BGO. For the energy range from 23 keV to 662 keV, these measurements were obtained in two different apparatus by illuminating long crystals at a temperature of ≈ 90 K with gamma rays from radioisotope sources and reading out the scintillation light with APDs. In addition one datum was obtained in a 4.6 T magnetic field based on the bismuth K x-ray escape peak produced by a continuum of background gamma rays. The combination of data taken under different conditions yields a reasonably consistent picture for BGO nonproportionality. An additional measurement using 6 keV X-rays from an ^{55}Fe source was obtained with a small crystal. An asymmetric peak was observed, hence a value for the nonproportionality was obtained by extracting the mean value for a fit to a gamma distribution.

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E [keV]	source	N_E [%]
23.3	^{109}Cd	73.4 (3.0)
31.7	^{133}Ba	81.2 (2.5)
59.5	^{241}Am	88.8 (1.0)
81.0	^{133}Ba	91.6 (1.5)
88.0	^{109}Cd	90.7 (1.0)
99.8	^{153}Gd	86.5 (1.5)
123.7	^{57}Co	89.2 (1.5)
185.7	^{235}U	93.3 (2.0)
661.7	^{137}Cs	100.0

Table 1: Mean gamma-ray energy deposits and nonproportionality N_E for the sources employed with the test cryostat apparatus. For sources with multiple, unresolved emission lines, the first column shows the mean energy E deposit (see text). The numbers in parentheses show the one-sigma uncertainty in the last digits, which were estimated based primarily on reproduceability and uncertainty in the determination of peak locations. The nonproportionality is defined to be 100 % for the photopeak produced by the 661.7 keV gamma ray from ^{137}Cs .

E [keV]	source	N_E [%]
32.3	^{133}Ba	81.0 (2.0)
42.7	^{57}Co (escape)	87.0 (2.0)
78.7	Bi escape	94.0 (2.0)
79.7	^{133}Ba	93.0 (1.0)
121.9	^{57}Co	92.0 (1.0)
356.0	^{133}Ba	98.6 (0.2)
661.7	^{137}Cs	100.0

Table 2: Mean gamma-ray energy deposits and nonproportionality N_E for the sources employed with the neutron beta-decay apparatus. The first column shows the mean energy E deposit as determined from our Monte Carlo modeling of the apparatus, without including nonproportionality. (Note that these values are not necessarily exactly the same as those in Table 1 because of a different experimental configuration and different methods for their determinations.) The nonproportionality is defined to be 100 % for the 661.7 keV gamma ray from ^{137}Cs . The numbers in parentheses show the one-sigma uncertainty in the last digits, which were determined from uncertainty in the determination of peak locations and the variance in the expected peak location from the Monte Carlo for the 11 BGO crystals.

ID	source	T [K]	opdet	crystal [mm]
	[2], R	295	PMT	not stated
	[3], R	295	PMT	10-20, 1-5
1	[4], R	295	PMT	9, 4 or 9
2	[4], R	100	APD	9, 4
3	[4], ER	100	APD	9, 4
	[5], R	3, 10, 30	PMT	20x10x5
4	[1], S	295	PMT	5-10, 1
5	test, R	90	APD	12x12x200
6	RDK, R	90	APD	12x12x200
7	RDK, E	90	APD	12x12x200

Table 3: Identification (ID) for numerical references in Fig. 3 and information on experimental conditions from the literature. The information for our data obtained using the test cryostat (test) and the neutron beta-decay apparatus (RDK) are reported. We list the source of the gamma rays, the approximate temperature T of the BGO crystal, the method for readout of the optical photons (heading “opdet”), and the crystal dimensions in millimeters (heading “crystal”) . The source are synchrotron radiation (S), radioisotopes (R), escape peaks from radioisotopes (ER), and Bi escape X-rays produced from a continuum of background gamma rays in our neutron beta-decay apparatus (E). For cylindrical crystals the dimensions listed are in the form: diameter, length.

FIGURE CAPTIONS

Fig. 1. Photographs of test cryostat apparatus. The left photograph shows the crystal and APD with the cryostat vacuum jacket and radiation shield removed. At the top of the photograph the inner reservoir of the dewar is visible. In the right photo (taken with cryostat rotated by 90 degrees) the cryostat is closed up and the vacuum port for illumination of the center of the crystal with sources located outside the cryostat vacuum is shown.

Fig. 2. Pulse height distributions obtained in the test cryostat apparatus for three of the seven sources employed (^{109}Cd , ^{133}Ba , and ^{137}Cs). The resolution is primarily determined by photoelectron statistics.

Fig. 3. Data for nonproportionality in the response of BGO in the gamma-ray energy range from (a) 0 keV to 800 keV and (b) 0 keV to 110 keV. See Table 3 for identification and information on each data set. For clarity, the nearly continuous and precise (0.5 % uncertainties) synchrotron data from Ref. [1] are shown as a line with small points. The error bars for our test dewar and RDK apparatus data correspond to the uncertainties listed in Table 1 and Table 2, respectively.

Fig. 4. Pulse height distribution obtained in the test cryostat apparatus for 1 cm³ BGO crystal illuminated by an ^{55}Fe source. The solid line is a fit to a gamma distribution.

Figure 1: Color online only

Figure 2: Color online only

Figure 3: Color online only

Figure 4: Color online only