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Intrinsic Energy Resolution and Light Yield Non-proportionality of BGO

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***Abstract*-- The intrinsic energy resolution and non-proportionality of the light yield as a function of gamma ray energies were studied for small BGO crystals at room and liquid nitrogen (LN_2) temperatures. The study showed that intrinsic resolution of BGO and the light yield non-proportionality as a function of energy does not depend on the crystal temperature. High light outputs of 14000±300 electron-hole pairs and energy resolution of 6.5±0.2% for 662 keV gamma rays were measured with the 9 mm diameter, 4 mm thick crystals, coupled to a Large Area Avalanche Photodiodes and cooled down to LN_2 temperature. Special attention was paid to analyze the energy resolution of the escape peaks, which were well separated from the full-energy photopeaks due to the good energy resolution of BGO at LN_2 temperatures and the energy of bismuth K X-rays. The intrinsic energy resolution of the BGO crystal for escape peaks does not show deviations from analysis of total energy absorption photopeaks. This is in spite of the fact that the contributions of X-rays and Auger electron cascade in creation of escape peaks are much smaller than in full-energy peaks. In the small volume crystal, mainly electrons produced in the photoelectric absorption create the escape peaks.**

I. Introduction

In the previous papers we have shown excellent properties of pure NaI and CsI scintillators operating at liquid nitrogen (LN_2) temperature [1-5] with Large Area Avalanche Photodiodes (LAAPD) readout. High light output above 100000 ph/MeV for CsI [4,5] and the energy resolution of 3.8 % for the 662 keV photopeak with NaI [2] have been recorded for the best crystals. Moreover, a good proportionality of the light yield versus energy of γ-quanta was obtained, in contrast to that measured at the room temperature. However, both the non-proportionality and intrinsic energy resolution of pure NaI and CsI were affected by a purity of studied crystals [2,3,5].

BGO is also known as the crystal showing a high light output at LN_2 temperature [4,5], allowing for good energy resolution [4]. Thus a similar study of non-proportionality and energy resolution versus energy of γ-quanta for the BGO crystals at LN_2 and room temperatures were of interest.

Measurements carried out with small BGO crystals at LN_2 temperatures showed escape peaks distinct from photopeaks due to a good energy resolution of BGO and the energy of bismuth KX-rays of about 76 keV. This allowed for analyzing their light output and energy resolution in comparison to that of the full energy peaks.

A full-energy peak after gamma energy absorption results from electrons produced in the photoelectric absorption followed by emission and subsequent absorption of a cascade of X-rays and Auger electrons, and electrons and photons generated by Compton scattering and terminated by photoelectric absorption. At the end the amount of scintillation light corresponding to the complete energy absorption consists of contributions of the numerous secondary electrons that have a variety of energies. For the low energy gammas detected in small volume crystals, photoelectric absorption dominates and the spread in the amount of light is due to different contributions from the X-ray and Auger electron cascade.

In contrast, mainly electrons produced in the photoelectric absorption create the escape peaks in small volume crystals. A contribution coming from a cascade of X-rays and Auger electrons is much reduced. Thus a determination of intrinsic resolution of escape peaks and comparison with that of the full energy peak would allow discussing a contribution introduced by the γ-rays stopping process.

To fulfill this task the scintillation response, light yield and energy resolution of small BGO crystals coupled to the Large Area Avalanche Photodiode and cooled down to LN_2 temperature have been measured. Furthermore, the same crystals were tested at room temperature using XP3212 photomultiplier. Obtained data on e-h pairs and photoelectrons yields versus energy of γ-rays and corresponding energy resolution allowed determination of the light yield non-proportionality and intrinsic resolution characteristics of BGO at both temperatures. At LN_2 temperature a special attention was paid to the escape peaks of single γ-rays in the 122 keV to 834.8 keV energy range.

II. Experimental details

Manufactured by Bicron three BGO crystals, 9 mm in diameter, marked 1 to 3 have been tested. Two of them, 4 mm thick, were tested at room and LN_2 temperatures, third one 9 mm thick was measured only at room temperature.

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In γ-ray measurements at room temperature, the crystals coated with Teflon tape were coupled by silicone grease to XP3212 photomultiplier having a high blue sensitivity of 11 μA/lm blue.

In the study at liquid nitrogen temperatures, the crystals were mechanically coupled (no silicone grease) to a 16 mm in diameter UV enhanced LAAPDs and cooled down to about 100 K temperature in a typical cryostat used for X-ray Si-detectors, see Fig. 1.

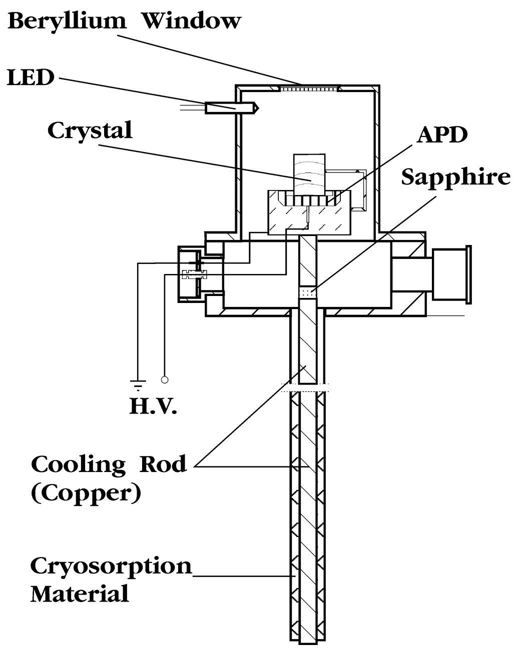

Fig. 1. Schematic drawing of the cryostat with the BGO crystal coupled to the LAAPD.

The 9 mm diameter BGO crystal placed in the center of the 16 mm diameter LAAPD left the remaining 3.5 mm wide ring open, enabling for X-ray direct illumination. This allowed use of the 5.9 keV X-ray photopeak from a ^{55}Fe source as the reference to measure a number of e-h pairs produced by BGO scintillation light.

In the experiments the signal from LAAPD was fed to a preamplifier (Ortec 142AH) and then to a spectroscopy amplifier (Tennelec TC244). The PC-based multichannel analyzer (Tukan) was used to record energy spectra. Gaussian functions were fitted to the full energy peaks, using procedures in the analyzer, to determine their position and energy resolution. The principle of procedures used to analyze escape peaks are presented in the next section.

III. Results

A. BGO Crystals at Room and LN_2 Temperatures

Fig. 2 shows the energy spectra of 662 keV γ-rays from a ^{137}Cs source, as measured with BGO crystal at room and LN_2 temperatures. A significant improvement of energy resolution at LN_2 temperature to 6.5% is observed, measured at 12 μs shaping time constant. This is owing to much higher light output of BGO at low temperature. It is, however, associated also with a much slower decay time of the light pulse [4,5]. Note the well separated escape peak of K X-rays, which allows analysis of its energy resolution.

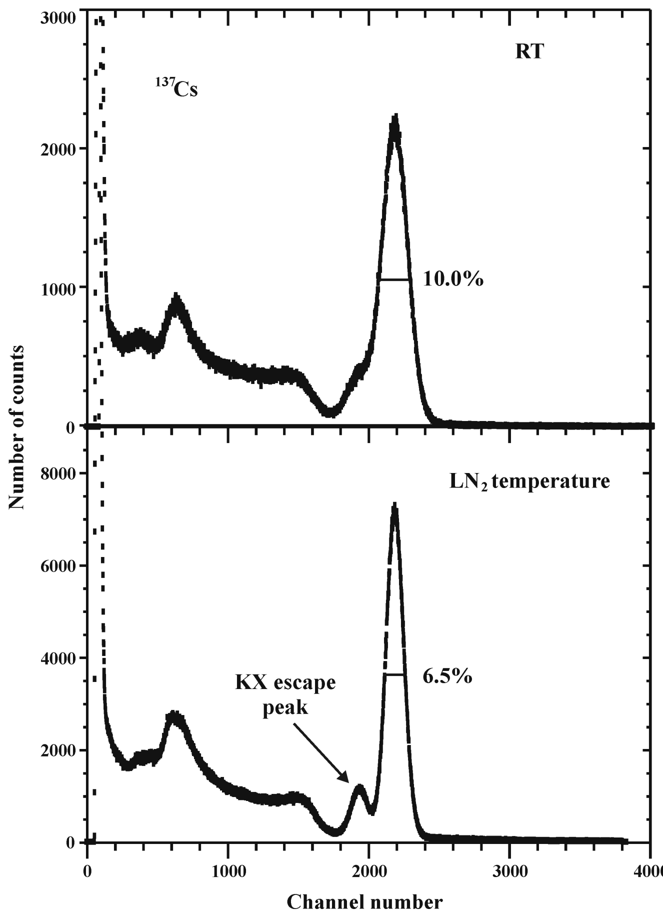

Fig. 2. Energy spectra of 662 keV γ-rays from a ^{137}Cs source, as measured with BGO crystal at room and LN_2 temperatures.

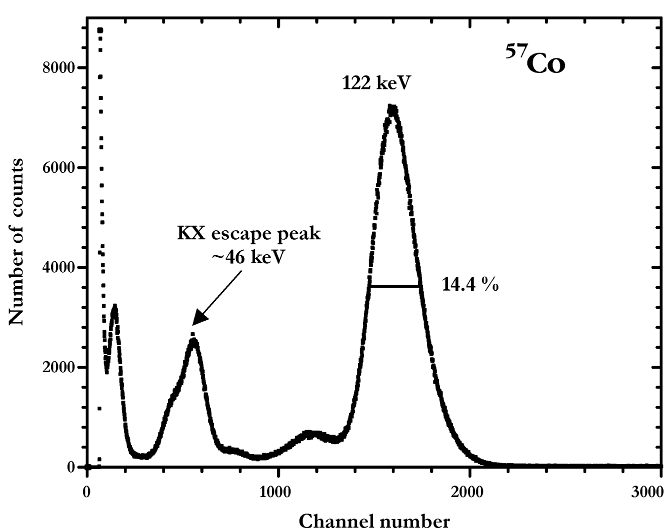

Fig. 3. The energy spectrum of γ-rays from a ^{57}Co source measured at LN_2 temperature

Fig. 3, in turn, presents the energy spectrum of γ-rays from a ^{57}Co source measured at LN_2 temperature. In this case the escape peak is shifted down to about 46 keV. The complex structure of the peak due to the escape of K_α and K_β X-rays can be recognized. It points out that a precise analysis of escape peaks is necessary to evaluate their energy resolution.

The escape peak corresponding to a given full-energy photopeak was treated in the analysis as a sum of five gaussian distributions related to an escape of $K_{\alpha1}$, $K_{\alpha2}$, $K_{\beta1}$, $K_{\beta2}$, and $K_{\beta3}$ components of the bismuth X-rays. In the fitting procedure the same FWHM value for all escape peaks, except of the 122-keV photopeak, was assumed and allowed to vary. Area and FWHM of the photopeak were set as free parameters. Areas of the escape peaks were fitted with one independent parameter ($K_{\alpha1}$ peak area) and the known relative intensity ratios between escape peaks.

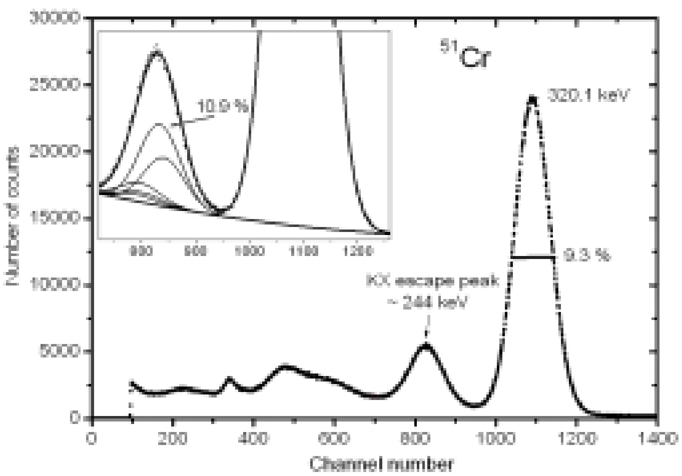

Fig. 4. The energy spectrum of 320 keV γ-rays from a ^{51}Cr source. In the insert the fitting result of the escape peak is shown.

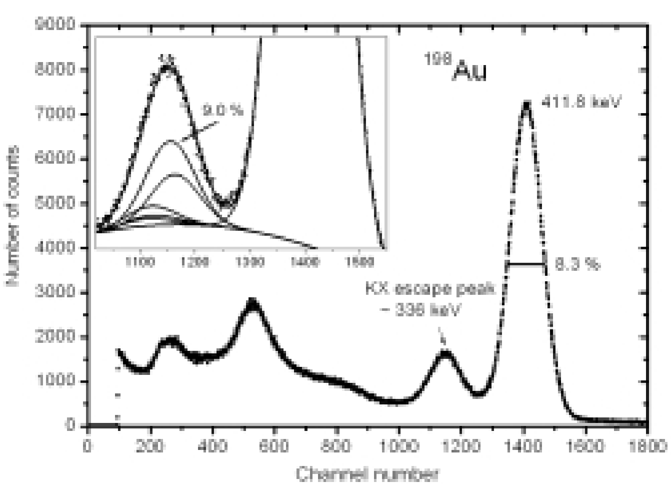

Fig. 5. The energy spectra of 411 keV γ-rays from a ^{198}Au source. In the inserts the fitting result of the escape peaks is shown.

The relative intensities of escape peak components were kept constant. The peak positions were treated as a set of constant input parameters, which were varying to achieve the smallest χ^2 values. The χ^2 minimization provided the best-fit parameters as well as the peak positions, thus the non-proportionality characteristics versus energy of escape components. The analysis of the escape peaks of single γ-rays in the 122.1 keV – 834.8 keV energy range shows that contributions from the KX-ray cascade in a crystal are negligible. Fig. 4 and 5 present the energy spectra and fitting result of the escape peaks of the 320.1 keV photopeak from the ^{51}Cr source and 411 keV photopeak from a ^{198}Au source.

Table I summarizes the obtained results. The yields of photoelectron and e-h pairs and the energy resolution recorded for the tested BGO crystals at both room and LN_2 temperatures are shown.

TABLE I

NUMBER OF PHOTOELECTRONS AND e-h PAIRS, AND ENERGY RESOLUTION MEASURED WITH BGO CRYSTALS

Crystal	Thickn. [mm]	Temp.	Time const. [μs]	$N_{e\text{-}h}$ or N_{phe} /MeV	Energy res. [%]	Light output [ph /MeV]
BGO 1	4	RT	2	1370 ±40	10.0 ±0.3	6900 ±140[a)]
BGO 3	9	RT	2	1200 ±40	10.6 ±0.3	6300 ±180[a)]
BGO 1	4	LN_2	12	14000 ±400	6.5±0.2	29000 ±2000
BGO 2	4	LN_2	12	14200 ±400	6.3±0.2	29500 ±2000

a) according to [6], measured for the same crystals by the XP2020Q PMTs with the calibrated quantum efficiency and Hamamatsu PIN photodiodes.

In the last column the light outputs of studied BGO crystals are collected. The light output at room temperature follows data from [6], measured for the same crystals by XP2020Q PMTs with a calibrated quantum efficiency and PIN photodiodes. At LN_2 temperature, the light output was determined based on the 83% integral quantum efficiency of the LAAPD. In this case, an additional correction factor of 0.58±0.03 was introduced, because of light collection looses associated with the mechanical contact of the crystal at LAAPD [1]. Note about 4 times higher light output of BGO at LN_2 temperature.

Fig. 6 presents the non-proportionality characteristics of BGO crystals determined at both room and LN_2 temperature. The non-proportionality is defined here as the ratio of e-h pair yield or photoelectron yield measured for photopeaks and escape peaks at specific γ-ray energy relative to the yield at 662 keV γ-peak.

The positions of the points corresponding to the escape peaks, represented by the $K_{\alpha1}$ components fit well within the curve. However, the precise analysis of the light output of escape peaks in relation to that of the full energy peak exhibited an excess of 1%. This deviation is not significant, considering accuracy of the above plot. The observed excess of the light yield is understandable, since a contribution of X-ray cascade, with lower light yield, in the escape peaks, is much reduced.

Note a good agreement of both the curves and an absence of temperature dependence. This implies that in contrary to halide crystals [2,3,7], the non-proportionality of BGO seems to be a fundamental characteristic of BGO material.

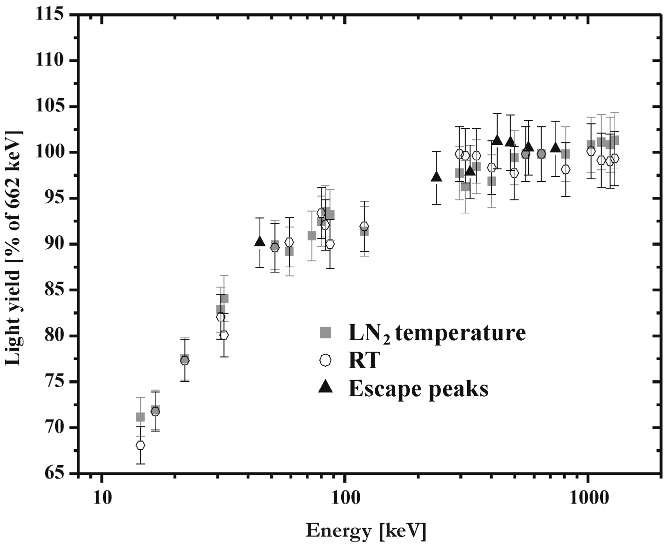

Fig. 6. The non-proportionality characteristics of BGO crystals determined at both room and LN_2 temperatures. The points corresponding to the escape peaks, represented by the triangles, fit well within the curve.

Fig. 7 presents the energy resolution versus energy of gamma rays measured with 4 mm thick BGO crystals at room and LN_2 temperatures.

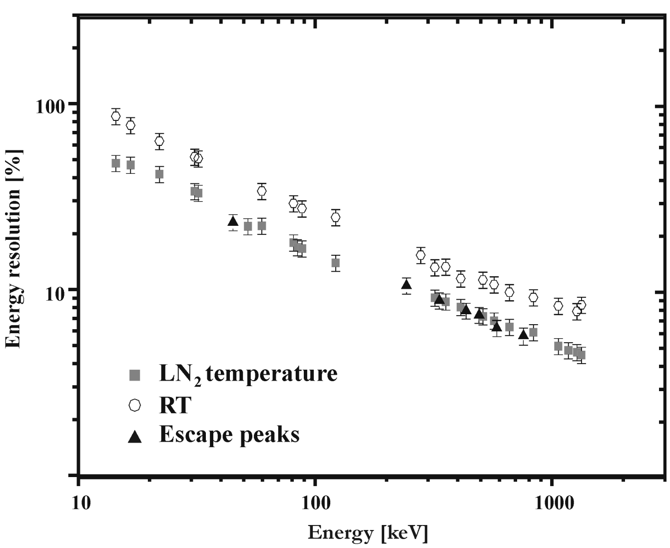

Fig. 7. Energy resolution measured with BGO at room and LN_2 temperature versus energy of gamma rays. The points corresponding to the escape peaks, represented by the triangles, fit well within the curve.

The parallel straight lines in the double logarithmic plot show that the energy resolution of BGO in the whole range of measured energies is about 40% better at LN_2 than room temperature. It is mainly associated with the higher light output reflected by more then 10 times higher number of e-h pairs in LAAPD, comparing to the number of photoelectrons in PMTs. Some of the points of the LN_2 temperature curve correspond to the escape peaks. They fit very well within the curve.

B. Analysis of energy resolution measured with BGO crystals at room and LN_2 Temperatures

The energy resolution, $\Delta E/E$, of the full energy peak measured with a scintillator coupled to a photodetector can be written as [8,9]:

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_{st})^2 + (\delta_n)^2 \tag{1}$$

Where δ_{sc} is the intrinsic resolution of the crystal, δ_{st} is the statistical contribution and δ_n is the dark noise contribution to the resolution. Eq. (1) omits a contribution of so-called transfer resolution associated with the variation of light and photoelectron collection [8,9]. This quantity is negligible in the modern PMTs and LAAPDs [8].

The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [8], [9] and many effects such as inhomogeneities in the scintillator causing local variations of the light output and non-uniform reflectivity of the crystal covering.

The statistical uncertainty of the signal from the PMT is described, as:

$$\delta_{st} = 2.35 \text{ x } 1/N^{1/2} \text{ x } (1 + \varepsilon)^{1/2} \tag{2}$$

Where N is the number of photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.12 for the XP3212.

The statistical accuracy of the signal from an APD is affected by the excess noise factor F, reflecting the statistical fluctuation of the APD gain. Assuming Poisson statistics of primary e-h pairs, contribution of the statistical processes to the energy resolution is given by:

$$\Delta N/N_{e\text{-}h} = 2.355\ (F/N_{e\text{-}h})^{1/2}, \tag{3}$$

The dark noise contribution in the case of the measurements with PMTs is negligible. The dark noise of 11 r.m.s. electrons was measured with LAAPD readout, at LN_2 temperatures and 12 µs shaping time.

Fig. 8 presents the calculated, according to eq. (1), intrinsic energy resolution of BGO, based on the measurements carried out at both room and LN_2 temperatures.

A common straight line in the double logarithmic plot does not show temperature dependence in the contrary to that observed with the halide crystals [2,3,7]. This observation agrees well with the common non-proportionality characteristics presented in Fig. 6. Also, it confirms further that the intrinsic resolution is strongly correlated with the non-proportionality of the scintillator response [3,7,8].

The similar dependence of the measured energy resolution and intrinsic resolution on gamma ray energy, as for BGO, was recently reported for the $LaCl_3$ crystal in [10] and [11] and earlier for LSO, GSO and YSO in [12]. In all the cases, the non-proportionality curves showed a monotonic decrease of the light yield below 50 – 100 keV energy.

In turn, the NaI(Tl) intrinsic energy resolution and non-proportionality dependence on energy reported in [8] differ from BGO data. The intrinsic resolution of BGO is approximately inversely proportional to the square root of energy, while a step-like dependence of NaI(Tl) resolution [8] was evidently correlated with the shape of the light non-proportionality curve.

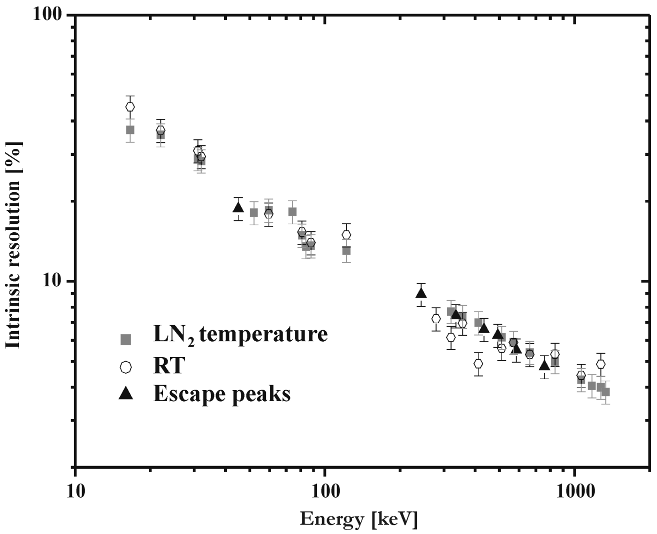

Fig. 8. Intrinsic resolution of BGO crystal at room and LN_2 temperatures. The points corresponding to the escape peaks, represented by the triangles, fit well within the curve.

It is important to note that the points corresponding to the intrinsic resolution of the escape peaks, represented by the $K_{\alpha 1}$ components, fit well to the measured dependency in spite of much reduced, compared to the full energy peaks, contribution coming from a cascade of X-rays and Auger electrons [13].

This supports the conclusion of ref. [8] that the scattering of secondary electrons in the crystal, so called δ-rays, mainly creates the intrinsic resolution. The importance of the contribution of δ-rays to the intrinsic resolution was discussed earlier by Iradele [14].

IV. Conclusions

The study of BGO crystals at LN_2 temperature showed a high light output of 31000±2000 ph/MeV and the energy resolution of 6.5±0.2% for 662 keV γ-rays from a ^{137}Cs source, both much better than the same parameters measured at room temperature.

The study of non-proportionality and intrinsic resolution as a function of gamma energy exhibited common curves independently of temperature. It suggests that, in contrary to halide crystals, both curves for BGO represent fundamental characteristics of the crystal material itself.

The analysis of the energy resolution of the escape peaks in the energy range of 122 keV to 835 keV showed the good agreement between the intrinsic resolutions evaluated from the escape peaks in relation to the full energy peaks. This infers that the X-ray cascade, generated in the stopping process of γ-rays in scintillator, weakly affects the measured intrinsic resolution. Only a small excess of the light yield of about 1% was observed for the escape peaks. This study confirmed that the scattering of electrons (δ-rays) is the most dominant component of the intrinsic resolution, as was concluded in a previous study [8].

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