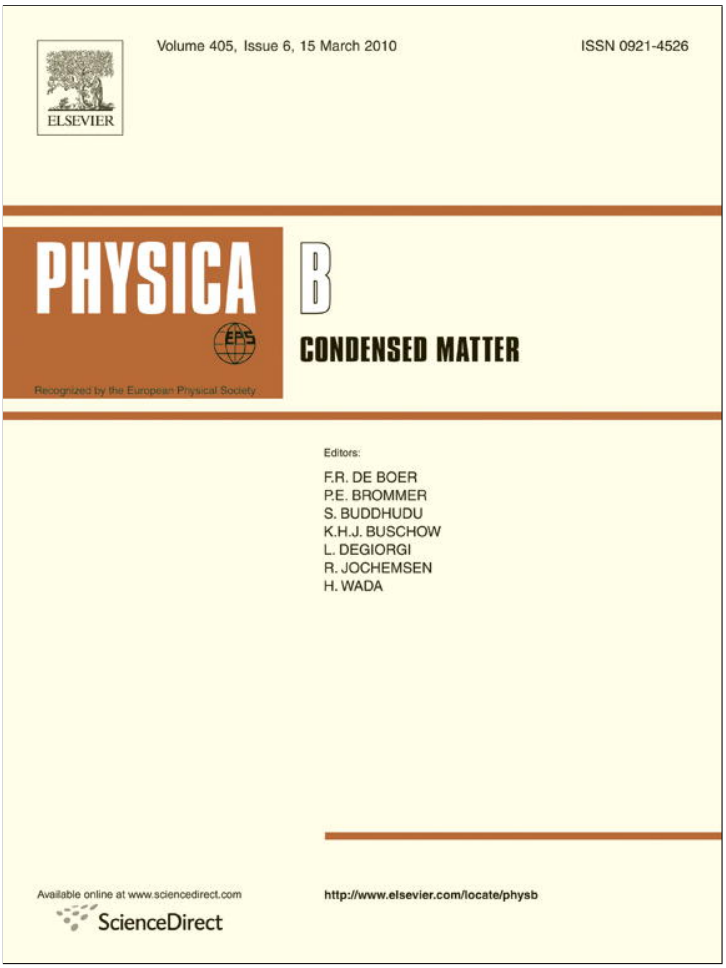

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Scintillation yield of $Bi_4Ge_3O_{12}$ (BGO) pixel crystals

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ABSTRACT

BGO crystals have been grown using the Czochralski method by the West Pomeranian University of Technology in Szczecin, Poland. We report the measurements of scintillation yield and low temperature thermoluminescence, performed on polished $2\times2\times10\,mm^3$ pixel and $2\times2\times2\,mm^3$ cube samples. The yield of the pixels placed horizontally on the PMT window, as well as of the cubes, is about twice higher than the yield of the pixels placed vertically. Such a distinct light loss is interpreted in the frame of a simple two-ray model, describing quantitatively the scintillation yield of pixel samples depending on the measurement geometry. Related to this effect, the common overuse of "BGO units" for expressing the light output of any scintillator is discussed. Thermoluminescence glow curves consist of several peaks attributed to the existence of traps, which turn out to be responsible for a slight decrease of the scintillation yield measured at room temperature in a short time window. To improve the performance of BGO crystals, efforts should be aimed at reducing both the internal light losses and the trap concentration, with the main emphasis laid on the former.

1. Introduction

$Bi_4Ge_3O_{12}$ (BGO) was originally proposed as a potential high-energy X-ray and gamma-ray scintillator by Weber and Monchamp [1]. A first detailed investigation of its basic scintillation properties was performed by Nestor and Huang [2]. In comparison with the well-known NaI:Tl such strong points of BGO as its higher density and gamma absorption coefficient, better mechanical and chemical durability, and non-hygroscopicity were pointed out. The intrinsic 300 ns luminescence of Bi^{3+} ions ($^3P_1\rightarrow{}^1S_0$) at 480 nm was also regarded as an advantage, since it eliminated the problems with non-uniform distribution of luminescence centers, prevalent in doped crystals. Further studies on BGO demonstrated its high radiation hardness [3,4] and absence of afterglow [5]. All these features caused that in spite of only average values of light output, energy resolution, and scintillation decay time, crystals of BGO were successfully implemented in such devices as, inter alia, medical PET scanners [6,7], HEP electromagnetic calorimeters [8], and complex detectors for astrophysics [9,10]. Although some modern scintillators introduced in recent years are likely to replace BGO in those applications sooner or later, there is still a scientific interest in this material, resulting in several papers published per year (e.g. [11–13]). Besides, BGO is commonly used as a kind of a reference pattern at determining the yield of other scintillators, i.e. the yield of an examined crystal is compared to the yield of a well-characterized BGO sample and then expressed as "*x*% of BGO".

In the current work we investigated a set of polished $2\times2\times10\,mm^3$ pixel and $2\times2\times2\,mm^3$ cube crystals of BGO, cut from larger boules grown with the conventional Czochralski method by the Laboratory of Crystal Growth at the West Pomeranian University of Technology (WPUT) in Szczecin, Poland. For a comparison, two 5-years-old BGO $2\times2\times10\,mm^3$ pixels from Photonic Materials Limited (PML), Bellshill, Scotland, were also studied. An overview of all utilized samples is presented in Table 1. Our main attention was focused on the scintillation yield of BGO, its dependence on the crystal size and possible reasons for any light losses. The choice of a $2\times2\times10\,mm^3$ pixel, itself useful for small animal PET scanners, for the basic sample dimension provided an opportunity to compare the scale of internal loss of scintillation light in BGO with such representatives of newer generations of materials as LuAP:Ce and LuYAP:Ce, for which a similar research had been performed before [14,15].

2. Experiment

Pulse height spectra necessary to determine photoelectron yields and energy resolutions were collected at room temperature (RT) under 0.511 and 1.274 MeV gamma excitation from a ^{22}Na source. The pulsed output signal from a Hamamatsu R2059 photomultiplier (PMT) was processed by a Canberra 2005

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Table 1
An overview of the studied BGO samples.

ID	Shape	Dimensions	Manufacturer
K01	pixel	$2\times2\times10\,mm^3$	West Pomeranian University of Technology, Szczecin, Poland
K02			
K11	cube	$2\times2\times2\,mm^3$	
K12			
N37	pixel	$2\times2\times10\,mm^3$	Photonic Materials Limited, Bellshill, Scotland
N38			

integrating preamplifier, a Canberra 2022 spectroscopy amplifier, and a multichannel analyzer. Positions of so-called photopeaks (PP) in the spectra, corresponding to full energy scintillations, were used to evaluate the yields of particular samples, expressed as numbers of photoelectrons released from the PMT photocathode per unit (1 MeV) of energy deposited in the crystal. A detailed description of this offset-corrected procedure was given by Wojtowicz et al. [14]. To improve the light collection efficiency the samples were coupled to the quartz window of the PMT with Viscasil grease and covered with several layers of Teflon tape.

A typical set-up consisting of an X-ray tube operated at 44 kV and 4 mA, a 0.5 m Acton Research Corporation SpectraPro-500 monochromator, a Hamamatsu R928 photomultiplier, and an APD Cryogenics Inc. closed-cycle helium cooler with a Lake Shore 330 programmable temperature controller, was used to measure low temperature thermoluminescence (TL). Prior to the TL runs, the samples were exposed for 10 min to X-rays at 10 K. The glow curves were recorded between 10 and 300 K at a heating rate of about 0.15 K/s.

3. Results and discussion

3.1. Pulse height spectra

Fig. 1 shows a representative set of three pulse height spectra of BGO, recorded with a $2\times2\times10\,mm^3$ pixel in two measurement geometries (i.e. vertically and horizontally placed on the PMT window) and a $2\times2\times2\,mm^3$ cube. One easily notices that the yield of the pixel is about twice higher in horizontal geometry, being close to the yield of the cube. This observation agrees with the conclusions of Dujardin et al. [16] that light output depends mostly on the height of the crystal, less on its length or width.

The values of photoelectron yield and energy resolution (at 0.511 MeV) of all studied samples, derived from their pulse height spectra, are summarized in Table 2. Although there are differences between individual specimens of the same size grown by the same laboratory, some general conclusions can be drawn. With respect to the energy resolution R and to the yield Y_H measured in horizontal geometry, the crystals from WPUT do not give way to the crystals from PML. The latter turn out to be better in vertical geometry, displaying higher values of Y_V than the former. In other words, a so-called "V2H" coefficient, introduced by Balcerzyk et al. [17] and defined as a ratio of Y_V to Y_H, is higher for the pixels from PML ("V2H"=0.56–0.60) than for the ones from WPUT ("V2H"=0.43–0.49).

For a detailed characterization of the decrease of scintillation yield Y of BGO with increasing sample height h we employ a simple two-ray ("2R") model proposed by Wojtowicz et al. [14]. This approach, already applied for such scintillators as LuAP:Ce and LuYAP:Ce [14,15], LuAG:Pr [18], BaF_2:Ce [19], $LaBr_3$:Ce [20], and $CeBr_3$ [21], introduces two parameters: an intrinsic yield Y_0 and a loss coefficient μ, describing the yield, which would be observed in the absence of any losses and the light loss inside the material caused by optical absorption and photon scattering, respectively. The square and circle data symbols in Fig. 2 correspond to the photoelectron yields measured in horizontal (h=0.2 cm) and vertical (h=1 cm) geometry, whilst the solid curves result from fitting the following "2R" equation:

$$Y(h)=Y_0\frac{1-e^{-2\mu h}}{2\mu h} \quad (1)$$

separately for each pixel and provide the values of Y_0 and μ. These values make the difference between the crystals from the two manufacturers more clear: although the pixels from WPUT are characterized by higher intrinsic yields (Y_0=1010–1030 phe/MeV) than the ones from PML (Y_0=896–1010 phe/MeV), their loss parameters are significantly larger (μ=1.15–1.46 cm^{-1} vs. μ=0.74–0.89 cm^{-1}). Thus BGO from PML is currently a better choice for PET cameras (vertical geometry) and any applications, in which large sizes of detectors are needed, since its lower intrinsic yield is more than compensated by the smaller loss coefficient. We note, however, that the loss parameters of the crystals from WPUT are not extraordinarily large. For a comparison, from identical measurements performed on $2\times2\times10\,mm^3$ pixels of Lu(Y)AP:Ce(Mo) [14,15] the following values have been obtained: μ=1.0–2.9 cm^{-1} (LuAP:Ce), μ=0.86–2.7 cm^{-1} (LuYAP:Ce), and

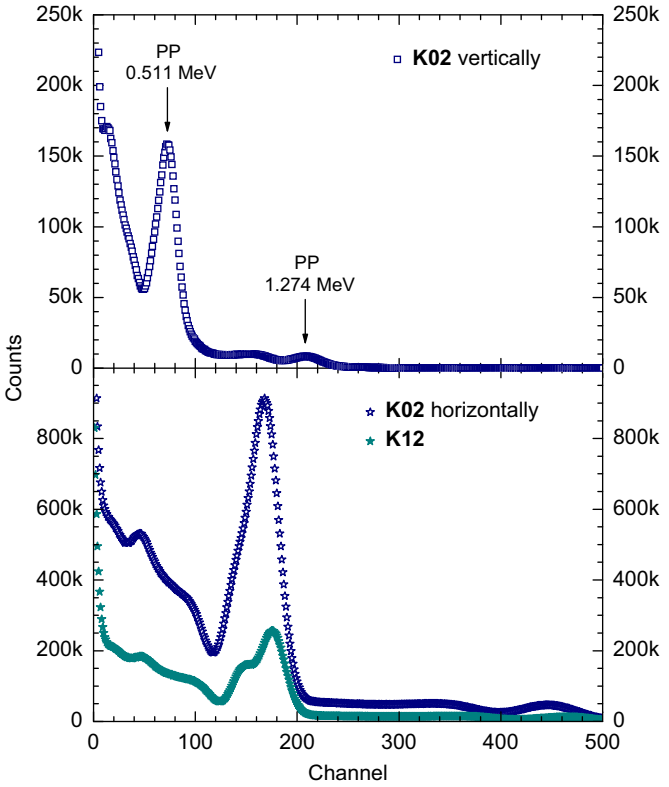

Fig. 1. Pulse height spectra of the $2\times2\times10\,mm^3$ pixel (K02) and the $2\times2\times2$ mm^3 cube (K12) of BGO.

Table 2
Parameters describing the properties of the studied BGO crystals (Y_V and Y_H are the photoelectron yields displayed by the sample placed on the PMT window vertically and horizontally, respectively; R the energy resolution at 0.511 MeV; "V2H" the ratio between Y_V and Y_H; Y_0 the intrinsic photoelectron yield; μ the loss parameter; "TL/(TL+ssRL)" the fraction of the total excitation energy that has been accumulated in traps; $Y_{0,tf}$ the intrinsic photoelectron yield of a trap-free material).

ID	Y_V (phe/MeV)	Y_H (phe/MeV)	R (%)	"V2H"	Y_0 (phe/MeV)	μ (cm^{-1})	"TL/(TL+ssRL)"	$Y_{0,tf}$ (phe/MeV)
K01	328 ± 17	766 ± 39	27.0 ± 1.4	0.43	1010	1.46	0.022	1030
K02	404 ± 21	828 ± 42	23.1 ± 1.2	0.49	1030	1.15	0.022	1050
K11		851 ± 43	20.0 ± 1.0					
K12		858 ± 43	18.9 ± 1.0					
N37	469 ± 24	776 ± 39	30.9 ± 1.6	0.60	896	0.74		
N38	471 ± 24	847 ± 43	23.8 ± 1.2	0.56	1010	0.89	0.006	1020

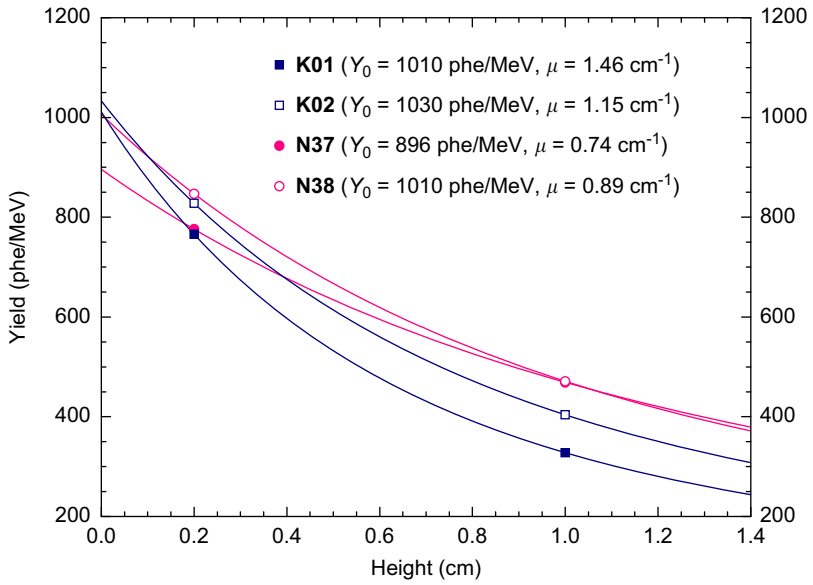

Fig. 2. Photoelectron yield of BGO as a function of sample height. Error bars are not shown for clarity of the figure.

μ=1.4–2.0 cm^{-1} (LuAP:Ce,Mo). Nevertheless, a reduction of the loss coefficient in BGO from WPUT is desired and should be achievable, at least to the level of the PML samples, by optimizing the growth procedures.

The preceding data give us an opportunity to raise the issue of so-called "BGO units". It is a common approach that in order to determine the yield of an examined crystal, one records its pulse height spectrum and subsequently a pulse height spectrum of a BGO reference sample. By comparing the photopeak positions one expresses the sought yield as "x% of BGO". To prove how risky such practice is, let us consider the LuAP:0.15%Ce $2 \times 2 \times 10$ mm^3 pixel from [15] as the studied crystal (Y_H=2470 phe/MeV, Y_V=704 phe/MeV) and the BGO N38 pixel as the reference one (Y_H=847 phe/MeV, Y_V=471 phe/MeV). Based on the comparison of the measurements in horizontal geometry the yield of LuAP:Ce is 292% of BGO, whereas in case of vertical geometry it is 149% of BGO. This large discrepancy clearly demonstrates the imperfection of the "BGO units" (if one still insisted on using these units anyway, the value related to horizontal geometry should be chosen for being less affected by internal losses). Therefore, we advise not to use the "BGO units" at all and instead to specify the yields in numbers of photoelectrons or photons per 1 MeV, always mentioning the sizes of the samples.

3.2. Thermoluminescence

Glow curves of three BGO crystals (two from WPUT and one from PML) are presented in Fig. 3. They reveal the existence of three major traps peaking between 70 and 170 K, and one (WPUT) or two (PML) minor traps above 170 K. Since the trap distributions in the studied samples are quite similar, we suppose that at least the three major traps are a genuine feature of the BGO host and thus occur in the material regardless of its manufacturer. The trap parameters, i.e. trap depths E, frequency factors s, and initial concentrations n_0, derived from the glow curve fits based on the classic first-order model of Randall and Wilkins [22], are listed in Table 3. These parameters, however, need to be corrected due to the presence of thermal quenching of the Bi^{3+} luminescence in BGO. In order to introduce adequate corrections we follow the approach of Petrov and Bailiff [23,24], which is suitable for the case of internal quenching described by the formula:

$$I(T) = \frac{I_0}{1 + C \exp\left(-\frac{W}{k_B T}\right)} \tag{2}$$

where I is the luminescence intensity at any temperature, I_0 the luminescence intensity in the absence of quenching, W the quenching activation energy, and C the constant. The energy correction itself is equal to

$$\Delta E = \frac{CW}{C + \exp\left(\frac{W}{k_B T}\right)} \tag{3}$$

whereas the corrected trap parameters are expressed as

$$E_{corr} = E + \Delta E \tag{4}$$

$$\ln s_{corr} = \ln s + \frac{\Delta E}{k_B T_{max}} \tag{5}$$

Based on the quenching curve published by Gironnet et al. [11] the values of W=0.0598 eV and C=19.1 have been evaluated. The corrected trap depths and frequency factors are singled out in Table 3.

A simple estimation of the potential influence of the detected traps on the scintillation yield of BGO exploits our specific mode of TL measurement, in which the steady-state radioluminescence (ssRL) during the irradiation prior to the TL runs is also recorded. In this way it is possible to calculate for each crystal a "TL/(TL+ssRL)" ratio, indicating the fraction of the total excitation energy that has been accumulated in traps. The values of these ratios, given in Table 2, point out that the contribution from traps is relatively small, particularly in BGO from PML, but still not negligible. Such estimation, however, can be illusive, because it does not consider the thermal range of the trap activity. Therefore, for a precise evaluation of the role of the traps in the scintillation of BGO and primarily their effect on the scintillation yield at room temperature we use the equation developed by Wojtowicz et al. [25], expressing the yield Y as a function of the trap lifetime τ

$$Y = Y_{tf}\left(a + b\frac{\tau_{rad}}{\tau_{rad}-\tau}\left(1+\frac{\tau}{\tau_{rad}}\left(\exp\left(-\frac{2.35\tau_{sh}}{\tau}\right)-1\right)\right)\right) \quad (6)$$

where Y_{tf} is the yield of an ideal trap-free material (regarding the traps detected in TL; possible non-radiative recombination centers are neglected in this approach), a and b are the relative contributions from the direct and trap-mediated scintillation components, respectively (a+b=1), τ_{rad} the radiative lifetime of the emitting ion, and τ_{sh} the electronic shaping time. In accordance with Eq. (6) the aY_{tf} level of the scintillation yield is always preserved, whereas the contribution from bY_{tf} strongly depends on the trap lifetime τ and, following the well-known Arrhenius formula:

$$\frac{1}{\tau} = s\exp\left(-\frac{E}{k_B T}\right) \quad (7)$$

where E is the trap depth, s the frequency factor, k_B the Boltzmann constant, on temperature T. At low temperature the yield is decreased to aY_{tf}, because the trap-mediated component bY_{tf} is completely stored in the traps due to their very long lifetime. At elevated temperature the short lifetime makes the traps ineffective and the entire yield of $(a+b)Y_{tf}=Y_{tf}$ is observed. If there are N different types of traps in the studied material, Eq. (6) assumes the form

$$Y = Y_{tf}\left(a + \sum_{i=1}^{N} b_i K_i\right) \quad (8)$$

where

$$K_i = \frac{\tau_{rad}}{\tau_{rad}-\tau_i}\left(1+\frac{\tau_i}{\tau_{rad}}\left(\exp\left(-\frac{2.35\tau_{sh}}{\tau_i}\right)-1\right)\right) \quad (9)$$

Since we are mostly interested in the scintillation yield of BGO at room temperature, we calculate the values of K_i for the four traps detected in the K02 pixel, using Eq. (9) and Eq. (7) with the parameters from Tables 2 and 3 (including the corrections for thermal quenching), and additionally: T=300 K, τ_{rad}=300 ns (the RT decay time constant of the Bi^{3+} luminescence), τ_{sh}=10 μs. The results listed in Table 4 show that the value of $\sum_{i=1}^{4} b_i K_i$ sum is very

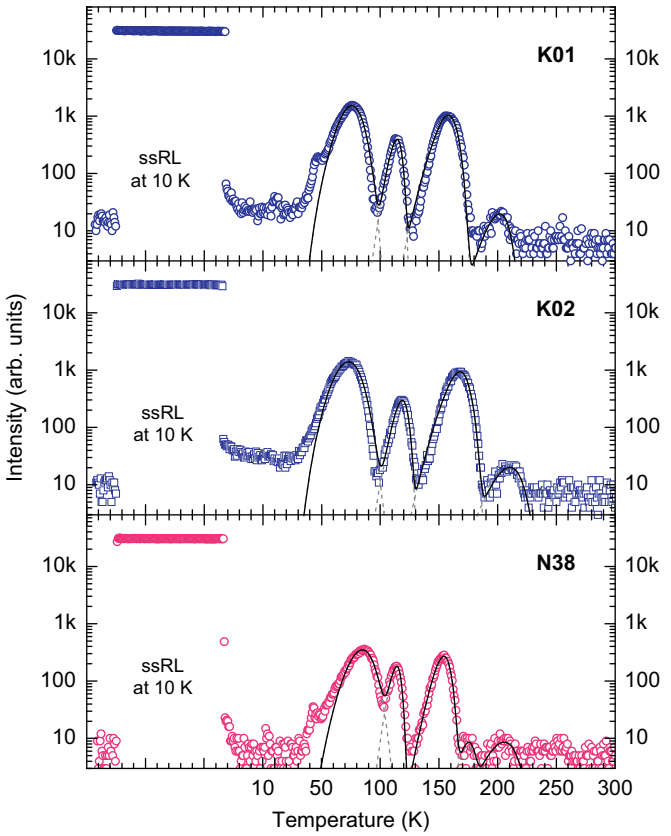

Fig. 3. Glow curves recorded with the 2 × 2 × 10 mm^3 BGO pixels (K01, K02, and N38) at a heating rate of 0.15 K/s following a 10 min X-ray irradiation.

Table 3
Parameters of traps detected in the studied BGO crystals, derived from first-order glow curve fits and corrected for thermal quenching (T_{max} is the temperature, at which the glow curve peaks; E the trap depth from fit; ΔE the temperature-dependent correction of depth; E_{corr} the corrected trap depth; s the frequency factor from fit; s_{corr} the corrected frequency factor; n_0 the initial trap concentration; n_0 is in the same units as TL intensity and s is in s^{-1}).

ID	peak no.	T_{max} (K)	n_0	E (eV)	ΔE (eV)	E_{corr} (eV)	ln s	ln s_{corr}
K01	1	76	216,000	0.0503	1.24×10^{-4}	0.0504	3.55	3.57
	2	115	29,600	0.253	2.61×10^{-3}	0.256	22.4	22.7
	3	158	133,000	0.278	0.0114	0.289	16.5	17.3
	4	201	2900	0.402	0.0226	0.425	19.1	20.4
K02	1	73	219,000	0.0401	8.73×10^{-5}	0.0402	2.12	2.13
	2	119	26,000	0.232	3.19×10^{-3}	0.235	19.1	19.4
	3	168	130,000	0.291	0.0141	0.305	16.1	17.1
	4	210	3820	0.331	0.0246	0.356	13.9	15.3
N38	1	85	55,500	0.0568	3.28×10^{-4}	0.0571	3.53	3.57
	2	114	12,500	0.277	2.47×10^{-3}	0.279	24.9	25.2
	3	155	28,000	0.335	0.0106	0.346	21.5	22.3
	4	174	732	0.459	0.0157	0.475	26.8	27.8
	5	206	1720	0.299	0.0237	0.323	1.25	2.59

Table 4
Influence of traps detected in the K02 BGO pixel on its room temperature scintillation yield (n_0 the initial trap concentration; b the relative contribution from the trap-mediated scintillation component, calculated as $n_{0i}/\sum_{i=1}^{4} n_{0i}$ multiplied by "TL/(TL+ssRL)"; K the factor describing the role of traps, see Eq. (9)).

peak no.	n_0	b	K at 300 K	$b \cdot K$ at 300 K
1	219,000	0.013	4.1×10^{-5}	5.3×10^{-7}
2	26,000	1.5×10^{-3}	0.50	7.5×10^{-4}
3	130,000	7.6×10^{-3}	4.6×10^{-3}	3.5×10^{-5}
4	3820	2.2×10^{-4}	1.1×10^{-4}	2.4×10^{-8}
total		0.022		7.9×10^{-4}

low, hence the scintillation yield displayed by the crystal at room temperature is indeed decreased to the aY_{tf} level. We note that a somewhat larger enhancement of yield would be observed in hypothetical trap-free BGO from WPUT ("TL/(TL+ssRL)"=0.022, $Y_{0,tf}$=1050 phe/MeV, the K02 sample) than in trap-free BGO from PML ("TL/(TL+ssRL)"=0.006, $Y_{0,tf}$=1020 phe/MeV, the N38 sample).

4. Conclusions

With respect to the scintillation yield measured in horizontal geometry there is no significant difference between BGO from WPUT and from PML. Although the pixels from WPUT have slightly better intrinsic yields, the values of their loss parameter are much higher, which decreases their yields measured in vertical geometry stronger than in case of the pixels from PML. The contribution of the trap-mediated scintillation component, although not large by itself, is also higher in the crystals from WPUT. Therefore, efforts should be aimed at reducing primarily the internal light losses (decreasing the loss parameter), but also the trap concentration (decreasing the "TL/(TL+ssRL)" ratio).

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