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Improved scintillation proportionality and energy resolution of LaBr₃:Ce at 80 K

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ABSTRACT

Using highly monochromatic synchrotron X-rays in the energy range from 10.5 to 100 keV, the temperature dependence of the photon nonproportional response and energy resolution of LaBr₃ scintillators doped with 5% Ce³⁺ were studied at 80, 295, and 450 K. Improvement of the proportionality and better energy resolution was observed on lowering the temperature. This effect suggests that the already outstanding energy resolution of LaBr₃:Ce can be improved even further. It may also provide new clues to better understanding of the processes that cause nonproportionality of inorganic scintillator response.

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1. Introduction

The widespread use of inorganic scintillators for applications in science and society is the driving force behind the search of new high performance compounds. Both novel and known phosphors are constantly suggested as promising scintillators [1–4]. The most important requirement imposed on new scintillators is a high energy resolution (R) for gamma-ray detection. R is defined as the Full Width at Half Maximum (FWHM) ΔE over the energy E of the full absorption peak in so-called pulse height spectra [5]. There are two key factors that determine the energy resolution, i.e., Poisson statistics in the number of detected photons (R_M) and the nonproportionality of the light yield of scintillators with gamma-ray energy (R_{nPR}) [6]:

$$\left(\frac{\Delta E}{E}\right)^2 = R^2 = R_M^2 + R_{nPR}^2 \quad (1)$$

$$R_M = 2.35 \sqrt{(1 + \text{var}(M)) / N_{phe}^{PMT}} \quad (2)$$

where $\text{var}(M)=0.28$ is the contribution from the fractional variance in the gain of the Hamamatsu R6231-100 photomultiplier tube [7] (PMT) and N_{phe}^{PMT} is the number of photoelectrons produced in the PMT by the scintillation photons from LaBr₃:Ce.

Nonproportionality means that the total light output of a scintillator is not precisely proportional to the energy of the absorbed gamma-ray photon. This has a deteriorating effect on energy resolution [6]. For example, based on Poisson statistics alone LaBr₃:Ce, which was discovered in 2001 [8], should display an energy resolution of 2.1% when 662 keV gamma-ray photons are detected with a standard type PMT but, however in reality it is about 2.8%. Because the light yield and the PMT performance is already close to the optimal value we need to reduce R_{nPR} in order to improve the energy resolution, and for that we wish to understand the physical causes of nonproportionality.

Nonproportionality is due to electron-hole recombination losses during the scintillation process [9]. It is currently believed that those losses occur inside parts of the ionization track where the ionization density is high [10–12]. That density increases when the gamma-ray energy decreases. The scintillation yield per energy unit in LaBr₃:Ce scintillator at 10 keV energy is for example 15% smaller than at 662 keV [5]. The origin of this decrease in efficiency, i.e., the true cause of electron-hole recombination losses, and the related deterioration in energy resolution is not known. It is a mystery to the scintillation community as to why some scintillators reveal poor proportionality while others appear reasonably good [13]. To elucidate the true origin of nonproportionality, accurate data on the scintillation light output of LaBr₃ doped 5% Ce³⁺ as function of energy and temperature were measured.

Attempts to measure the photon nonproportional response (photon-nPR) as a function of temperature were made before [14] by changing the temperature of both the scintillator and PMT over

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a relatively narrow temperature range from $-30\text{ }^{\circ}\text{C}$ to $+60\text{ }^{\circ}\text{C}$. In our experiments the PMT remains at room temperature while the scintillator temperature can be changed. Thus, we do not need to consider the effect of temperature on the characteristics of the PMT, and we measure only the intrinsic properties of the scintillator.

2. Experimental

To measure the pulse height spectra at many finely spaced energy values between 10.5 and 100 keV, experiments were carried out at the X-1 beamline at the Hamburger Synchrotronstrahlungslabor (HASY-LAB) synchrotron radiation facility in Hamburg, Germany. A highly monochromatic pencil X-ray beam in the energy range of 10.5–100 keV was used as an excitation source. A tunable double Bragg reflection monochromator using a Si[5 1 1] set of silicon crystals providing an X-ray resolution of 1 eV at 10.5 keV rising to 20 eV at 100 keV was used to select the X-ray energies. The beam spot size was set by a pair of precision stepper-driven slits, positioned immediately in front of the cryostat chamber. For all measurements, a slit size of $50 \times 50\text{ }\mu\text{m}^2$ was used. A description of the experimental setup used for nonproportionality and energy resolution measurements can be found in Refs. [15,16]. To record scintillation pulse height spectra as a function of temperature, a $\text{LaBr}_3\text{:Ce}$ sample was fixed at the bottom of a parabolic-like stainless steel cup covered with reflective Al foil mounted onto the cold finger of a liquid nitrogen bath cryostat. The cup directed the scintillation light through the quartz window towards a PMT situated outside the cryostat chamber. The Hamamatsu R6231-100 PMT at -680 V remained at room temperature and about 20% of the emitted scintillation light was observed. To collect as much of the PMT charge pulse as possible, the shaping time of an Ortec 672 spectroscopic amplifier was set at $10\text{ }\mu\text{s}$. The detailed study of $\text{LaBr}_3\text{:Ce}$ time profiles versus temperature can be found in Ref. [17], but even at 80 K slow component decay time does not exceed 710 ns. The temperature of the crystal was controlled by two thermocouples attached to different parts of the sample holder.

3. Results and discussion

We define the photon nonproportional response (photon-nPR) of a scintillator at X-ray energy (E_X) as the light output per MeV observed at energy E_X divided by the light output per MeV observed at $E_X=662\text{ keV}$. Fig. 1 shows the photon-nPR as a function of E_X at 80, 295 and 450 K. The shape of the response curves has been discussed by us in detail previously in Ref. [5]. Note that for $\text{LaBr}_3\text{:Ce}$ shown in Fig. 1 we observe discontinuities in the photon-nPR curves not only at the lanthanum K-electron binding energy $E_{\text{KLa}}=38.925\text{ keV}$, but also at the bromine K-electron binding energy $E_{\text{KBr}}=13.474\text{ keV}$. But the most important observation is that the photon-nPR reveals strong temperature dependence. At 100 keV the values of photon-nPR at all three temperatures are almost the same as 98.8%. The situation is quite different at low energy. At 10.5 keV the photon-nPR at 80 K is 90.7%, at 295 K it falls to 86.1% and at 450 K it has decreased to 82.1%. Therefore, a clear improvement in photon-nPR occurs as the temperature decreases from 450 to 80 K. Since the typical error in photon-nPR does not exceed 1% at 10.5 keV these are statistically significant improvements. This result indicates that the electron-hole recombination losses in the high ionization density part of the ionization track in $\text{LaBr}_3\text{:Ce}$ have a strong temperature dependence.

Fig. 1. Photon nonproportional response of $\text{LaBr}_3\text{:Ce}$ as a function of X-ray energy (E_X) curve at (1) 80 K, (2) 295 K, and (3) 450 K.

Fig. 2. Energy resolution of the X-ray photopeak as a function of X-ray energy for $\text{LaBr}_3\text{:Ce}$ inside the cryostat at 80 K (curve 1) and at 450 K (curve 2). Curve (3) is obtained with $\text{LaBr}_3\text{:Ce}$ mounted on the PMT at 295 K and Curve (4) is the resolution derived from (3) for $\text{LaBr}_3\text{:Ce}$ inside the cryostat at 295 K.

The resolution R of the X-ray photopeaks is plotted on a double-log scale as shown in Fig. 2 as a function of E_X . Ideally when only R_M contributes to the energy resolution, a straight line with slope -0.5 is expected, see Eq. (2) and Ref. [18]. Due to a relatively small contribution of R_{nPR} to R , a slight deviation of the experimental values from a straight line can be observed as a step-like increase of R at energy E_{KLa} . This situation was discussed in detail by us in Ref. [5]. Curves (1) and (2) in Fig. 2 shows data obtained at 80 and 450 K with the $\text{LaBr}_3\text{:Ce}$ crystal mounted inside the cryostat. For these data, improvement of energy resolution is clearly seen with decrease of the scintillation crystal temperature. At 100 keV, the energy resolution at 80 K is 1.8% smaller than that at 450 K. With a decrease of X-ray energy the resolution difference increases reaching 10% at 10.5 keV. The $N_{\text{phe}}^{\text{PMT}}$ produced in the PMT by $\text{LaBr}_3\text{:Ce}$ 5% mounted inside the cryostat with 100 keV X-rays at 80 K was 479 ± 4 photoelectrons while it

was 463 ± 4 photoelectrons at 450 K. This confirms the high temperature stability of the scintillation light yield of $\text{LaBr}_3\text{:Ce}$ 5% [17]. The improved energy resolution at 80 K is therefore attributed to better proportionality.

To further demonstrate that the improved energy resolution is due to improved proportionality we collected data obtained at room temperature by direct optical coupling of the $\text{LaBr}_3\text{:Ce}$ 5% scintillation crystal to the photocathode of a Hamamatsu R6231-100 PMT, see curve (3) shown in Fig. 2. The number of photoelectrons N_{phe}^{PMT} produced in the PMT by 100 keV X-ray detection was 2254 ± 10 photoelectrons whereas $N_{phe}^{PMT} = 485 \pm 4$ when the same crystal is mounted inside the cryostat. The data from Fig. 1 and Eqs. (1) and (2) provides us with $N_{phe}^{PMT}(E_X)$, $R_M(E_X)$, and $R_{nPR}(E_X)$ at 295 K. Assuming that $R_{nPR}(E_X)$ is an intrinsic contribution from the scintillator that is not affected by the photon collection efficiency we can calculate $R(E_X)$ at room temperature for a scintillator mounted inside the cryostat, see curve (4) shown in Fig. 2. Note that the curve (4) falls nicely in between curves (1) and (3). The increasingly better proportionality observed in Fig. 1 when temperature decreases is therefore fully consistent with the improvement of energy resolution observed in Fig. 2.

4. Conclusion

Summarizing, we have found that by reducing the temperature of $\text{LaBr}_3\text{:Ce}^{3+}$ scintillator from 450 to 295 K and then to 80 K its photon proportionality improves and as a consequence a better energy resolution for X-ray photon detection is obtained. This improvement evidences that the yet unknown parameters responsible for the nonproportional response of $\text{LaBr}_3\text{:Ce}$ depend on temperature. Apparently, with decreasing temperature, the amount of electron-hole recombination losses in the dense parts of the ionization track decreases and as a consequence the efficiency of the scintillator increases. Recent simulation data in Ref. [11] suggests that electron and hole mobilities are important parameters that determine nonproportionality, and possibly our finding is a manifestation of changing mobilities with changing temperature. It is known that electron mobility is strongly temperature dependent due to phonon scattering [19]. Although, the cause of proportionality improvement is not yet established in this work, our results do demonstrate that nonproportionality is a property that can be

improved. If it can be improved by changing temperature one may also hope to improve it by other means like activator concentration, crystal quality, co-dopants, etc.

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