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Enhanced time response of 1-in. $LaBr_3(Ce)$ crystals by leading edge and constant fraction techniques

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ABSTRACT

We have characterized in depth the time response of three detectors equipped with cylindrical $LaBr_3(Ce)$ crystals with dimensions of 1-in. in height and 1-in. in diameter, and having nominal Ce doping concentration of 5%, 8% and 10%. Measurements were performed at ^{60}Co and ^{22}Na γ-ray energies against a fast BaF_2 reference detector. The time resolution was optimized by the choice of the photomultiplier bias voltage and the fine tuning of the parameters of the constant fraction discriminator, namely the zero-crossing and the external delay. We report here on the optimal time resolution of the three crystals. It is observed that timing properties are influenced by the amount of Ce doping and the crystal homogeneity. For the crystal with 8% of Ce doping the use of the ORTEC 935 CFD at very shorts delays in addition to the Hamamatsu R9779 PMT has made it possible to improve the $LaBr_3(Ce)$ time resolution from the best literature value at ^{60}Co photon energies to below 100 ps.

1. Introduction

High-density scintillators that exhibit fast response and good energy resolution are the preferred crystals in a number of applications such as gamma-ray spectroscopy, medical diagnosis and timing measurements. One of the fastest scintillators commercially available nowadays is $LaBr_3(Ce)$ [1–3]. It has very good timing properties in addition to high γ-ray efficiency and good stopping power. Its energy resolution at 662 keV is measured to be as good as 2.8%, and its photon yield corresponds to 63 photons/keV [3], which is much higher than that of other inorganic scintillators [4].

Due to their superb properties, $LaBr_3(Ce)$-based detectors have been the option of choice since 2005 [5] for the application of the Advanced Time-Delayed (ATD) method, which was introduced as a HPGe-gated β–γ electronic timing technique with ultrafast scintillators [6]. The ATD method allows the measurement of nuclear level lifetimes down to a few picoseconds by using the fast coincidences between the radiation populating and de-exciting a nuclear energy level [6,7]. Nuclear level lifetimes are decisive observables in nuclear physics since they provide access to reduced transition probabilities between nuclear states, and therefore insight into the nuclear structure. Currently $LaBr_3(Ce)$ crystals have become the standard detectors for *fast-timing* spectroscopy [8–11].

The sensibility of the ATD method is directly determined by the time resolution of detectors in use, and therefore the best choice of crystal type ($LaBr_3(Ce)$ in this case), size, shape and doping is required, together with the right selection of the coupling photosensor [12] is required. The optimization of the electronic circuit and the detector operation parameters is also needed to achieve the maximum performance [13,14] of the set-up.

In this paper we report on the time response of three $LaBr_3(Ce)$ detectors equipped with 1-in. cylindrical crystals. The selected photomultiplier tube (PMT) is the Hamamatsu R9779, which is the most performing model available nowadays in the market [12]. The detector time response has been optimized by fine-tuning of the electronics parameters. Three crystals with different nominal Ce concentration were used [15], making it possible to assess the effect of the amount of doping on the time response. This work is part of a broader study of the characteristics of ultrafast scintillator detectors, equipped with crystals of different types, sizes and geometries and coupled to state-of-the-art photomultipliers. The aim is to construct a high performance fast-timing array of $LaBr_3(Ce)$ detectors for γ–γ and β–γ spectroscopy, and in

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† During the preparation of the final version of this paper, our colleague Henryk Mach suddenly passed away. Henryk was an inspirational character for us and for several generations of scientists. He will be greatly missed.

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particular the optimal FAst TIMing array (FATIMA) [16], which will be placed at the focal plane of the SuperFRS at FAIR. This instrument belongs to the HISPEC-DESPEC experiment [17,18] of the Nuclear Structure Astrophysics and Reactions (NUSTAR) collaboration, one of the four pillars of the FAIR project [19].

2. Detector characteristics

2.1. The $LaBr_3(Ce)$ detectors

Three $LaBr_3(Ce)$ crystals with identical shapes were studied in order to characterize their timing properties. The crystals were produced by different manufacturers with distinct Ce dopant concentration, but having the same cylindrical shape with nominal size of 1-in. in height and 1-in. in diameter, and hermetically sealed inside an aluminum housing. The housing had a thin aluminium window at the entrance, and was fitted with a glass light guide at the coupling side to the photosensor. Inside the case several layers of light reflector and shock absorbing material assure the stability of the crystal and minimize photon losses.

Crystals were labelled as A, B and C. Crystal C was the first one produced; it was grown in 2006 as a test crystal. Since it had been reported that the time resolution of $LaBr_3(Ce)$ crystals improves with the amount of Ce doping [20], Crystal C was made with enhanced Ce doping concentration of 10%, while standard crystals were commercially available with 5% doping. Crystal A was produced in the same year as a commercial crystal using the standard crystal growth and production techniques, with a 5% of Ce. Finally, at the end of 2012, Crystal B was produced with an 8% Ce doping, with the idea of finding a good balance between increased doping and homogeneity for medium-sized crystals.

The chosen photomultiplier tube fitting the crystal properties is the Hamamatsu R9779. This tube is optimized for fast timing applications and shows good timing and energy characteristics [12,13]. It is an 8-stage device with a window of 2 in. in diameter, equipped with bialkali photocathode. It has a typical transit time of 20 ns with transit time spread (TTS) of 250 ps, and anode rise time of 1.8 ns [21]. To favour light transmission, crystals were coupled to the PMT by Viscasil silicon grease and wrapped into opaque tape. An example of the anode pulse is plotted in Fig. 1. PMT high voltages in the range of -1200 V to -1300 V were found to be optimal to provide fast-timing response while preserving good energy resolution and linearity as presented in the next sections.

2.2. Energy resolution, linearity and efficiency

In addition to the time response, energy resolution and linearity are also important characteristics to be considered. A non-linear energy response implies high-order polynomial energy corrections, while bad energy resolution could lead to the inability of precise γ-ray energy determination and hamper transition selection.

We report here the energy resolution of Crystals A, B and C at 662 keV (^{137}Cs), given as the ratio between the FWHM of the γ-ray full energy peak and its energy, corrected for non-linearity [13]. The resolution was measured at the PMT bias voltages that provide the best time resolution. The relative energy resolution of Crystals A, B and C, once corrected for non-linearity, is measured to be 3.4%, 3.4% and 4.0% respectively. These values are worse than the number quoted by the manufacturer, 2.8% (at 662 keV), due to the use of the fast R9779 PMT [21], which is optimized for timing measurements, and provides the best timing performance at the expense of slightly worse energy resolution. To quantify this effect Crystal A was also tested with a second PMT, designed for energy measurements (Hamamatsu R6231), yielding 2.9% (at 662 keV), value that matches the specifications. Table 1 summarizes the energy resolution measured for the three crystals with both PMT models (R9779 and R6231) and Fig. 2 illustrates Crystal B energy resolution as a function of the energy in the range from 122 to 1332 keV.

The dependence of the energy resolution on the Ce concentration was assessed as well. It has been shown in [20] that up to 5% of Ce concentration the $LaBr_3(Ce)$ photon yield increases [20] and hence the energy resolution also improves. However, a further Ce increase leads to constant or even lower energy resolution [22]. In our case, Crystals A (5% of Ce) and B (8% of Ce) have the same energy resolution, while Crystal C, with a 10% nominal Ce concentration, shows the worst value among the three, 4.0%. The measured photon yield for Crystal C is ~8% lower than for Crystal A, in accordance with [20], but also the photon yield for Crystal B with 8% Ce doping is higher than for Crystal A by ~10%. Given that the contributions to the intrinsic resolution are the non-proportional response (which is low for $LaBr_3(Ce)$ [2]) and the inhomogeneities, which cause local variations in the scintillation light output [23], the main reason for the worse energy resolution for the highly doped crystal C could be explained by Ce inhomogeneities in the crystal.

Detectors based on high photon yield scintillators such as $LaBr_3(Ce)$ may display non-linear behaviour, even with good light yield proportionality. This may be caused by space-charge effects in the photomultiplier tube [24]. In order to check the energy linearity of our detectors, we have measured the functional relationship between the peak position (signal amplitude) and the real γ-ray energy from a ^{152}Eu source for the three crystals at bias voltages in the range from -900 V to -1700 V. We have found that $LaBr_3(Ce)$ detectors behave

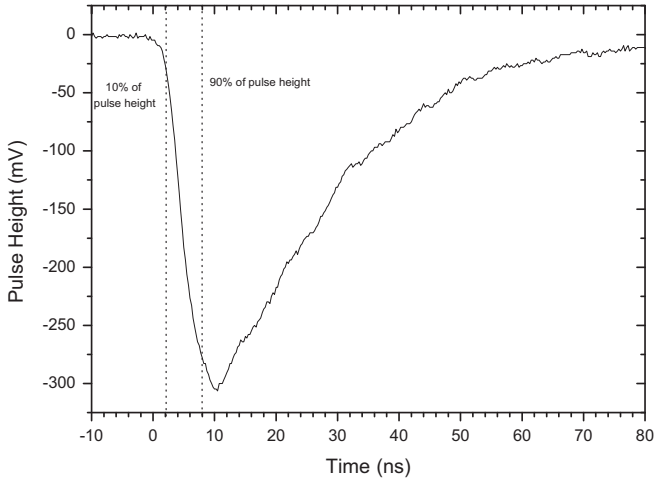

Fig. 1. Anode pulse of Crystal A+Hamamatsu R9779 acquired with 2 Gsa/s 1 GHz oscilloscope. The used source is a standard ^{137}Cs and the PMT bias voltage -1000 V. The anode rise time taken from 10% to 90% of the maximum pulse height is ~6 ns.

Table 1
Relative energy resolution Er of the three crystals (A–C) at ^{137}Cs energy (662 keV). The uncertainty in the values is 0.1%. The crystals were coupled to Hamamatsu R9779 and R6231 PMTs.

Crystal	Ce (%)	PMT	HV (V)	Er (%)
A	5	R9779	1300	3.4
A	5	R6231	1000	2.9
B	8	R9779	1300	3.4
C	10	R9779	1200	4.0