



## ガンマ線検出器試験

所属	甲南大学理工学部	ビームライン	BL01
利用者氏名	宇都宮弘章	利用分野	量子ビーム技術
利用年度	平成26年度	活用技術	ガンマ線利用

### 利用成果の概要

LaBr<sub>3</sub>:CeとCeBr<sub>3</sub>無機シンチレーション検出器は、NaI(Tl)検出器に比べて早いガンマ線応答、向上したエネルギー分解能と検出効率を持ち、高エネルギーガンマ線検出において優れた性能を有している。現在、ブカレスト近郊(ルーマニア)に欧州連合の研究所ELI-NPが建設中であり、利用代表者がconvenerを務める研究グループ(Gamma Above Neutron Threshold)では、巨大共鳴およびピグミー共鳴のガンマ崩壊を研究するためのガンマ線検出器候補として、LaBr<sub>3</sub>:CeとCeBr<sub>3</sub>検出器を検討している。ニュースバル放射光施設で生成するレーザー逆コンプトンガンマ線ビームを用いて、両シンチレーターの高エネルギーガンマ線に対する性能比較評価を行った。

以下のことが判明した。

- (1) エネルギー分解能ではLaBr<sub>3</sub>:Ce検出器がCeBr<sub>3</sub>よりやや優れている。
- (2) エネルギー直線性は同等である。
- (3) LaBr<sub>3</sub>:Ce検出器はよく知られているように<sup>138</sup>La由来の低エネルギーバックグラウンドガンマ線が存在するが、巨大共鳴の高エネルギーガンマ崩壊の検出には干渉しない。CeBr<sub>3</sub>検出器には検出器由来のバックグラウンドガンマ線は存在しない。
- (4) LaBr<sub>3</sub>:Ce検出器とCeBr<sub>3</sub>検出器はどちらもELI-NPプロジェクトのガンマ線検出器として適している。

### <利用目的>

ELI-NPプロジェクトで巨大共鳴のガンマ崩壊を測定するガンマ線検出器の候補であるLaBr<sub>3</sub>:Ce検出器とCeBr<sub>3</sub>検出器の性能比較評価をレーザー逆コンプトンガンマ線を用いて行うことを目的とした。

### <実験方法>

NewSUBARU蓄積リング中の電子ビームとNd:YVO<sub>4</sub>レーザー光子(波長1064nm)の逆コンプトン散乱によってガンマ線ビームを発生させた。3.5"×4"LaBr<sub>3</sub>:Ce検出器と3×3"CeBr<sub>3</sub>検出器の逆コンプトンガンマ線ビームに対する応答関数を測定した。発生させたガンマ線の最大エネルギー(16.93, 15.19, 13.03, 12.30, 10.87, 9.54, 8.29, 7.12, 6.04 MeV)は電子エネルギーの絶対エネルギー較正值[1]から求めた。逆コンプトン散乱によるガンマ線生成過程を組み込んだモンテカルロシミュレーションコードGEANT4を用いて、測定した応答関数を解析した。逆コンプトン散乱のシミュレーションには電子ビームのエミッタンスを含めている。また、標準ガンマ線源(<sup>60</sup>Co, <sup>137</sup>Cs)を用いて2種類のシンチレーション検出器の低エネルギーガンマ線に対するエネルギー分解能を測定した。

### <実験結果>

得られたデータは、一緒に実験を行ったIoana Gheorghe(ELI-NPリサーチアシスタント、ブカレスト大学大学院生)によって解析され、レポートが作成されている。モンテカルロシミュレーションによって測定した応答関数をよく再現することができた。レポートを資料として添付する。

## 文部科学省 [先端研究施設共用促進事業トライアルユース 成果報告]

兵庫県立大学 高度産業科学技術研究所 ニュースバル放射光施設

### <成果の波及効果、今後の見通し>

本実験で得られた結果は、ELI-NPの当該研究グループのTDR(Technical Design report) に反映された。最終的なガンマ線検出器の選択は、業者見積りとELI-NPプロジェクトの国際評価委員会の評価によって決定される。

### 文献

[1] H.Utsunomiya et al., IEEE Transactions on Nuclear Science 61, 1252 (2014).

### <図面等>

お問い合わせ先 兵庫県立大学 高度産業科学技術研究所  
ニュースバル放射光施設 共用促進室  
〒678-1205 兵庫県赤穂郡上郡町光都1-1-2  
TEL : 0791-58-2543 FAX : 0791-58-2504  
E-mail : kyoyo@lasti.u-hyogo.ac.jp  
<http://www.lasti.u-hyogo.ac.jp/NS/>

# Comparative study of energy resolution and linearity of CeBr<sub>3</sub> and LaBr<sub>3</sub>:Ce using Laser Compton Scattered $\gamma$ -ray beams

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## 1 Introduction

The unique energy resolution and intensity parameters of the future ELI-NP Gamma Beam System (GBS) will allow a thorough investigation of the excitation and particle and gamma decay of Giant Resonances. Using the brilliant GBS at ELI-NP, the Gamma Above Neutron Threshold (GANT) research group proposes to study the GDR excitation and the competition of its various decay channels as well as that of direct and statistical decay processes to improve our understanding of the GDR structure. A multi purpose neutron and gamma radiation detection setup consisting in a flexible array up to 40 large volume scintillator detectors and liquid scintillation neutron detectors each is purposed to be developed.

Cerium doped Lanthanum Bromide - LaBr<sub>3</sub>:Ce - detectors are foreseen to be used for gamma ray detection in the multipurpose setup because of their good energy resolution, fast response and linear response with energy. But we are also interested in other types of detectors having the required working parameters. Such is the case of Cerium Bromide - CeBr<sub>3</sub> - scintillation detectors, which are known to have poorer energy resolution than the LaBr<sub>3</sub>:Ce ones, but considerably superior to other scintillation detectors such as NaI.

We report here on our comparative study between the working parameters of Cerium doped Lanthanum Bromide (LaBr<sub>3</sub>:Ce) and Cerium Bromide scintillation detectors investigated with quasimonochromatic  $\gamma$ -ray beams produced by the scattering of laser photons on relativistic electrons (LCS) at the synchrotron radiation facility NewSUBARU.

## 2 Experimental procedure

### 2.1 Detectors

We investigated the energy resolution of two cylindrical scintillation detectors, a 3"  $\times$  3" CeBr<sub>3</sub> detector from Scionix and a 3.5"  $\times$  4" LaBr<sub>3</sub>:Ce detector, BrillLanCe380 89S102/3.5, Saint Gobain. High positive voltage was applied on the LaBr<sub>3</sub>:Ce (630 V) and on the CeBr<sub>3</sub> detector (555 V), the two voltage values being chosen to use the entire ADC input range for minimum gain on the amplifier. Both detectors had photomultiplier tubes dedicated to energy spectroscopy measurements. The LaBr<sub>3</sub>:Ce photomultiplier signal was sent to an Ortec preamplifier and NIM amplifier module with 0.5  $\mu$ s shaping time. The amplifier signal was inserted into a 100 MHz 16 bit ADC digital MCA. The CeBr<sub>3</sub> detector had the TB-5 digital base from AMPTEK, containing a 14 pin photomultiplier base tube, a digital pulse processor with charge sensitive preamplifier and MCA and both (low and high voltage) power supplies.

### 2.2 Energy calibration with standard sources

Standard calibration sources <sup>60</sup>Co, <sup>137</sup>Cs and <sup>152</sup>Eu were used for low energy calibration and for investigating the energy resolution in the energy range of 661 to 1332 keV. Energy spectra of <sup>60</sup>Co and <sup>137</sup>Cs  $\gamma$ -ray sources taken with the LaBr<sub>3</sub>:Ce and the CeBr<sub>3</sub> detectors are displayed in Figure 1. The spectra are taken in the same conditions as the LCS  $\gamma$ -ray beam measurements - the same applied high voltage and the same amplification gain. Relative energy resolution values in full width at half maximum (FWHM) are displayed in Table 1 for different amplification conditions given by the applied high voltage. For the investigated energy range, the energy resolution of the LaBr<sub>3</sub>:Ce detector is superior but comparable to the CeBr<sub>3</sub> one. By increasing with  $\sim$  170 V the applied high voltage chosen to optimize the ADC input range usage on both detectors, the energy resolution of the LaBr<sub>3</sub>:Ce is considerably improved by  $\sim$  10 %, while the CeBr<sub>3</sub> one is remains constant within 1 %.

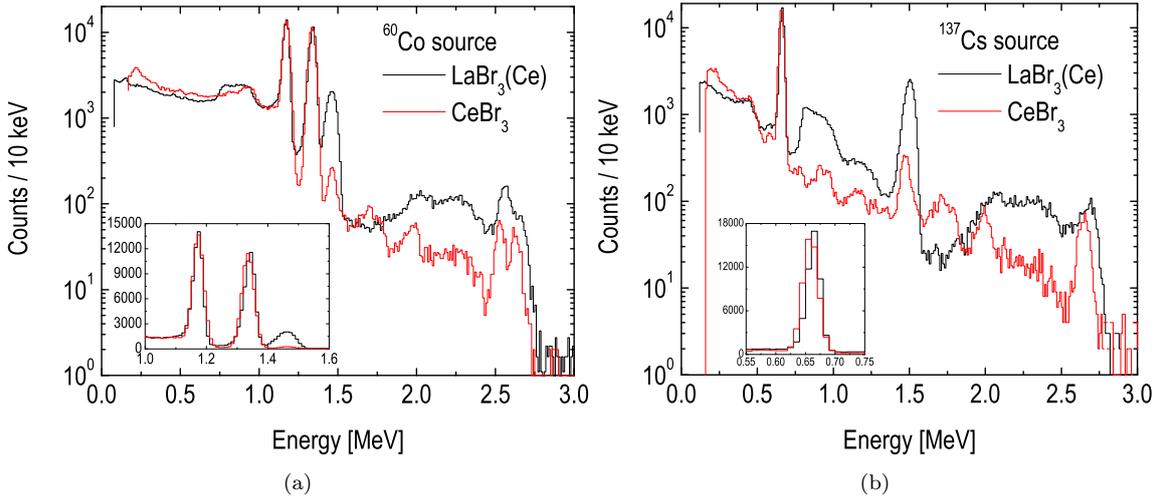


Figure 1: Energy spectra of standard calibration sources (a)  $^{60}\text{Co}$  and (b)  $^{137}\text{Cs}$  recorded with the  $\text{LaBr}_3:\text{Ce}$  detector (black line) and with the  $\text{CeBr}_3$  detector (red line) in the same conditions as the LCS  $\gamma$ -ray beam measurements - the same applied high voltage and the same amplification gain.

Table 1: Relative energy resolution values in FWHM for the  $\text{LaBr}_3:\text{Ce}$  and the  $\text{CeBr}_3$  detectors.  $E_\gamma$  and  $\Delta_E$  are the gamma ray energy and the detector's relative energy resolution in FWHM.

$E_\gamma^{max}$ [keV]	$\text{LaBr}_3:\text{Ce}$		$\text{CeBr}_3$	
	$\Delta_E$ [%]	High voltage [V]	$\Delta_E$ [%]	High voltage [V]
344.29	-	+ 630	5.95	+ 555
661.657	3.53	+ 630	4.39	+ 555
778.900	-	+ 630	4.05	+ 555
963.380	-	+ 630	3.44	+ 555
1173.28	3.30	+ 630	3.51	+ 555
1332.490	3.07	+ 630	3.39	+ 555
661.657	3.48	+ 800	4.45	+ 730
1173.28	2.99	+ 800	3.49	+ 730
1332.490	2.96	+ 800	3.37	+ 730

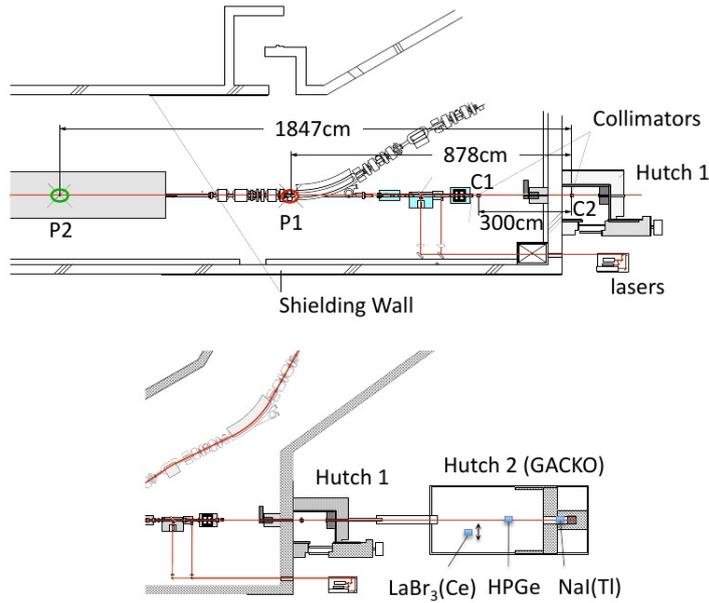
### 2.3 LCS $\gamma$ -ray beam measurements

Quasimonochromatic  $\gamma$ -ray beams with typical energy resolution values of **1.2 - 1.6 % in FWHM** are produced at the synchrotron radiation facility NewSUBARU in the inverse Compton scattering of laser photons from relativistic electrons circulating in a storage ring. For this measurement, a 40 W Nd:YVO<sub>4</sub> laser ( $\lambda = 1064$  nm) and electron beams with energies between 584 and 982 MeV were used to generate LCS  $\gamma$ -ray beams with maximum energies between 6 and 17 MeV. For each energy point, the  $\gamma$ -ray beam energy spectrum was alternatively measured with each scintillation detector.

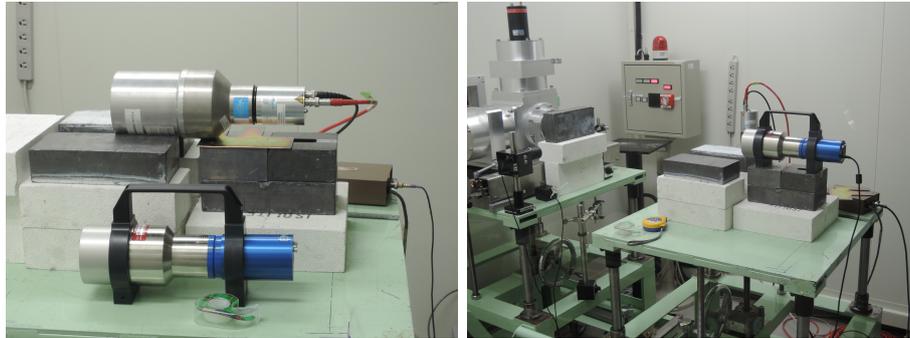
The experimental setup is displayed in Figure 2. The  $3.5'' \times 4''$   $\text{LaBr}_3:\text{Ce}$  and the  $3'' \times 3''$   $\text{CeBr}_3$  detectors were alternatively mounted at the end of the  $\gamma$ -ray beam line in the experimental Hutch 2, GACKO (Gamma collaboration hutch of Konan University), to record the incident LCS  $\gamma$ -ray beams.

Fine tunings of the laser optics and the collimator alignment along the horizontal (x), vertical (y) and rotational ( $\theta$ ) axes were carried out by irradiating the  $\text{LaBr}_3:\text{Ce}$  with LCS  $\gamma$ -ray beams. The optimal alignment was obtained by maximizing the  $\gamma$ -ray flux incident on the detector. After completing the collimator alignment procedure, both detectors were aligned to the gamma ray beam axis with synchrotron radiation. In the case of the NewSUBARU facility, the laser beam is sent head-on against the electron beam, therefore the electron beam axis coincides with both the laser and the  $\gamma$ -ray beam axis.

We measured the energy spectra of LCS  $\gamma$ -ray beams for 9 energy values of the electron beam. Figure 3 shows the energy spectra recorded with both detectors for each LCS  $\gamma$ -ray beam having the maximum energy of: (a) 6.042 MeV, (b) 7.121 MeV, (c) 8.286 MeV, (d) 9.536 MeV, (e) 10.872 MeV, (f) 12.289 MeV, (g) 13.027 MeV, (h) 15.187 MeV and (i) 16.931 MeV. For each energy point we measured for the same amount of time both LCS  $\gamma$ -ray spectra and background synchrotron radiation spectra. The electron beam energies and current, the laser power, the maximum  $\gamma$ -ray beam energies along with the measurement times and count rates for each detector at each energy point are listed in Table 2.



(a)



(b)

(c)

Figure 2: (a) Schematic image of the experimental setup in the  $\gamma$ -ray beam line GACKO at NewSUBARU facility. (b) and (c) Images of the experimental setup consisting in a  $3.5'' \times 4''$   $\text{LaBr}_3:\text{Ce}$  and a  $3'' \times 3''$   $\text{CeBr}_3$  detector. For each energy point, the LCS beam is recorded alternatively using the two detectors which are in turns aligned to the electron beam axis.

A top-up operation was performed for the 982.427 MeV electron beam energy measurements, having the electron beam current constant at 300 mA. For the lower energy values of the electron beam, the electron beam current varied between 288 and 143 mA. A 5 cm thick lead absorber was placed at  $\sim 60$  cm in front of the measuring detector to prevent pile-up events and possible saturation of phototube. The laser power was varied between 4 to 26 % of the maximum 40 W value to obtain detector count rates between 3 and 9 kcps.

We can observe in the LCS  $\gamma$ -ray beam energy spectra shown in Figure 3 that the  $\text{LaBr}_3:\text{Ce}$  detector has a higher full energy deposition probability than the  $\text{CeBr}_3$  detector. The difference between detectors full energy peak - first escape peak ratio is because of the different crystal sizes. Also, the second escape peak is significantly higher in the  $\text{CeBr}_3$  spectra because of the smaller crystal size.

The  $\gamma$ -ray beams are produced at NewSUBARU by the collision of a  $\text{Nd}:\text{YVO}_4$  laser in the first harmonic and electron beams of highly precised energy. The NewSUBARU electron nominal beam energy resolution is of 0.04 % and the uncertainty in the energy calibration over the energy range from 550 to 974 MeV is  $(5.5 - 9.4) \times 10^{-5}$ . Therefore, as will be the case of ELI-NP, the  $\gamma$ -ray beams have a precisely defined, sharp high energy front. After collimation, the  $\gamma$ -ray beam presents a low energy tail with an small energy width of  $\sim 1.2$  to  $1.6$  % in FWHM values, depending on collimator thickness and aperture<sup>1</sup>. An example of incident  $\gamma$ -ray beams along

<sup>1</sup>A complex collimation system consisting of four main collimators is employed at NewSUBARU. A large aperture (2 cm) first collimation stage consisting in a water cooled Copper collimator placed near the interaction point is used for protecting the electron pipes from high flux  $\gamma$ -ray beams. A second collimator stage of large aperture (5 cm) consisting in a 30 cm thick lead block placed at approximately 13 meters from the interaction point is used for absorbing  $\gamma$ -rays scattered at large angles. The fine tuning collimation system consists of two 10 cm thick lead blocks of variable apertures (1 to 6 mm).

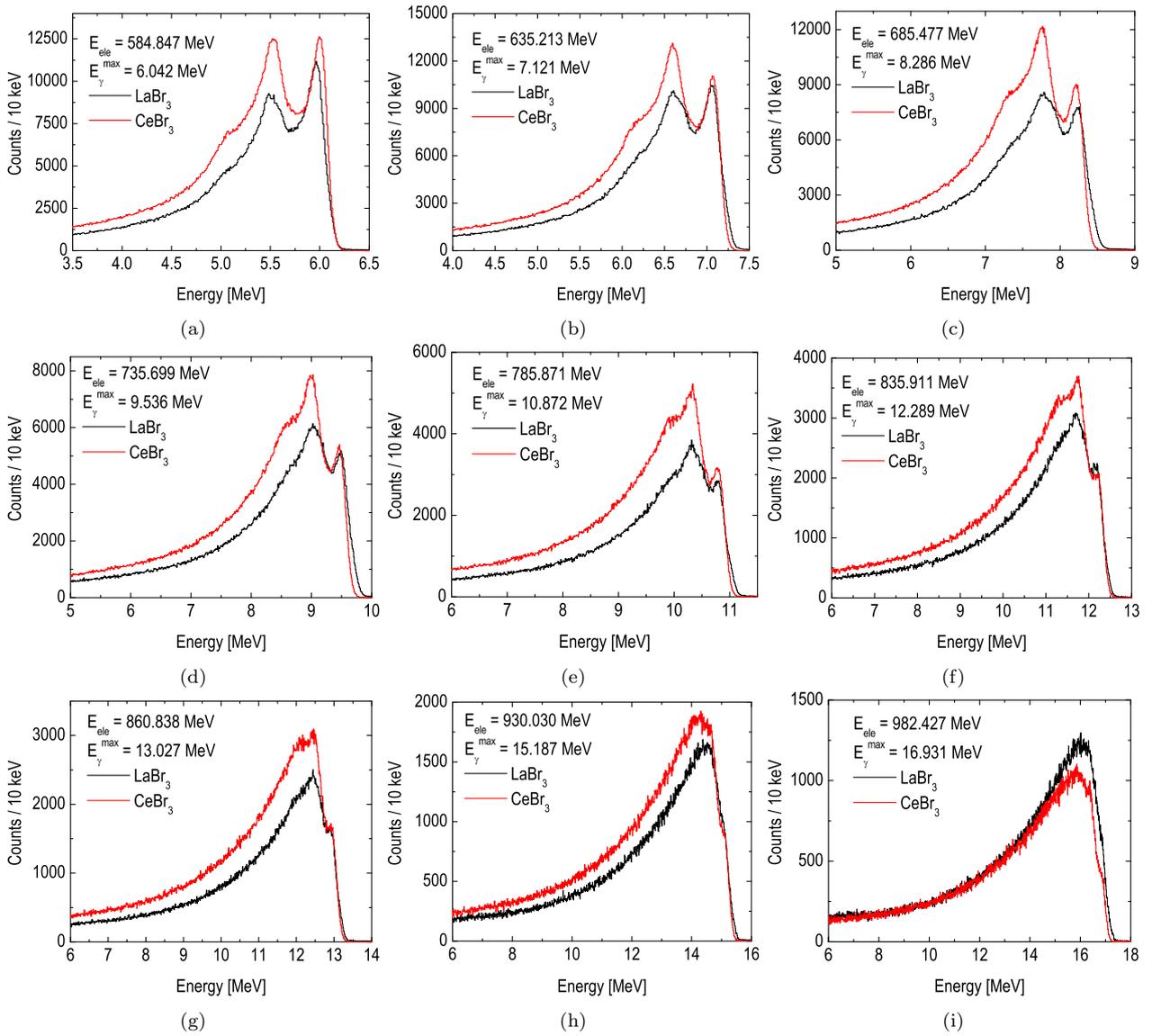


Figure 3: Laser Compton Scattered  $\gamma$ -ray beam spectra recorded with a  $3.5'' \times 4''$   $\text{LaBr}_3$  detector (black line) and with a  $3'' \times 3''$   $\text{CeBr}_3$  (red line). The detectors' response functions have been investigated using LCS  $\gamma$ -ray beams with maximum energies of: (a) 6.042 MeV, (b) 7.121 MeV, (c) 8.286 MeV, (d) 9.536 MeV, (e) 10.872 MeV, (f) 12.289 MeV, (g) 13.027 MeV, (h) 15.187 MeV and (i) 16.931 MeV. See text for details.

with experimental and simulated  $\text{LaBr}_3:\text{Ce}$  detector response is shown in Figure 4 taken from the PRC article published this year "Photoneutron cross sections for samarium isotopes: toward a unified understanding of ( $\gamma$ , n) and (n,  $\gamma$ ) reactions in the rare earth region" by Filipescu et al.

Therefore, the high energy front of the LCS  $\gamma$ -ray beam spectra is given entirely by the energy resolution of the detector and it is the main indicator of energy resolution differences between the  $\text{LaBr}_3:\text{Ce}$  and the  $\text{CeBr}_3$  detector. As can be seen in Figure 3, the LCS  $\gamma$ -ray beam high energy front measured with the  $\text{CeBr}_3$  detector is steeper than the one recorded with the  $\text{LaBr}_3:\text{Ce}$  detector.

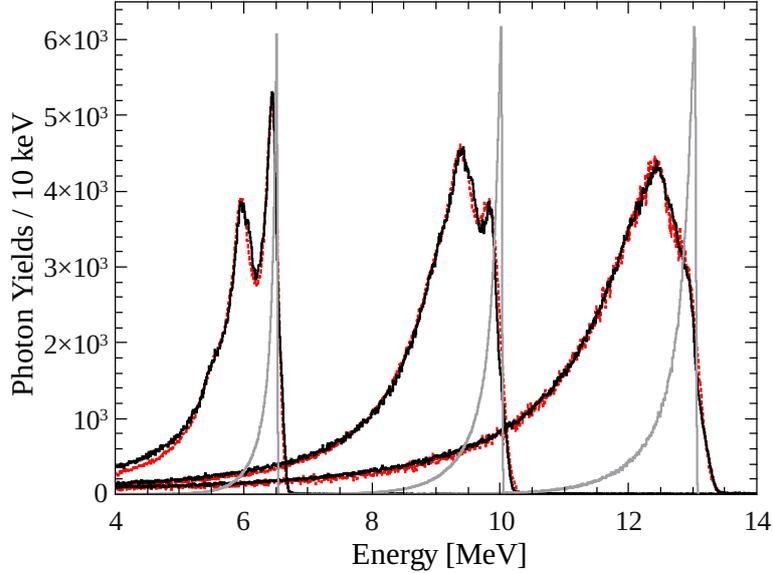


Figure 4: Typical spectra of the NewSUBARU LCS  $\gamma$ -ray beams recorded with the  $\text{LaBr}_3\text{:Ce}$  detector (solid lines), the simulations of the response function (dotted lines) and of the incident  $\gamma$ -ray beam (gray lines). *Figure taken from D.M. Filipescu et al PRC paper (2014).*

Table 2: Details regarding each LCS  $\gamma$ -ray beam measurement with each detector.  $E_\gamma^{max}$ ,  $E_{ele}$ ,  $t_m$  and  $I_{ele}$  represent the LCS  $\gamma$ -ray beam maximum energy, the electron beam energy, the measurement time for each irradiation and the electron beam current.

$E_\gamma^{max}$ [MeV]	$E_{ele}$ [MeV]	$\text{LaBr}_3\text{:Ce}$			$\text{CeBr}_3$			Laser power [W]
		Count rate [kcps]	$t_m$ [s]	$I_{ele}$ [mA]	Count rate [kcps]	$t_m$ [s]	$I_{ele}$ [mA]	
6.042	584.847	7.5	200	143	6.1	300	151	10.4
7.121	635.213	8.9	200	168	6.9	300	163	9.68
8.286	685.477	8.8	200	178	8.0	300	185	8.36
9.536	735.699	7.4	200	203	5.8	300	198	5.84
10.872	785.871	5.4	200	216	4.7	300	224	3.52
12.289	835.911	5.5	200	245	3.8	300	238	2.8
13.027	860.838	4.8	200	255	3.8	300	264	2.36
15.187	930.030	4.7	200	288	3.3	300	279	1.68
16.931	982.427	3.3	300	300	2.3	300	300	<1.68

## 2.4 Linearity of detector response with energy

The response linearity with energy was investigated for both scintillation detectors. The energy calibration was performed using the low energy  $\gamma$ -ray transitions of the  $^{60}\text{Co}$  source and the maximum energy of each LCS  $\gamma$ -ray beam. Although simulations are necessary to obtain the precise spectrum position of the  $\gamma$ -ray beam maximum energy, qualitative information can be obtained by examining the experimental spectra. The energy calibration was performed with a third degree polynomial. The degree of the polynomial was chosen to obtain the most similar correspondance between energy and channel number for both detectors. As can be observed in Figure 5, in the present configurations consisting in photomultiplier tube types, crystal size, applied high voltage and amplification gain, etc., the  $\text{CeBr}_3$  detector in the present configuration has a more linear response than the  $\text{LaBr}_3\text{:Ce}$ .

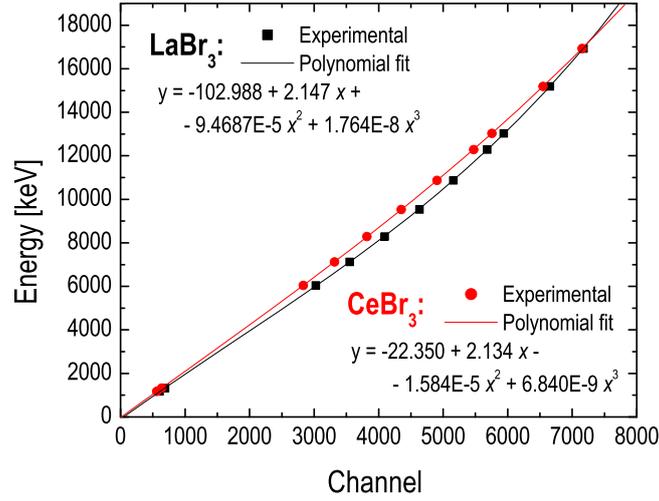


Figure 5: Linearity in the response of a LaBr<sub>3</sub>:Ce detector - black line, and of a CeBr<sub>3</sub> detector to  $\gamma$  rays in the energy range 1173 - 16931 keV. The data for low-energy photons were taken with the standard  $\gamma$ -ray source <sup>60</sup>Co. The data for 6 -17 MeV  $\gamma$  rays were taken with LCS  $\gamma$ -ray beams produced with a Nd:YVO<sub>4</sub> laser. The solid lines are the 3<sup>rd</sup>-order polynomial fits to the data.

## 2.5 Intrinsic radioactivity of LaBr<sub>3</sub>:Ce crystals

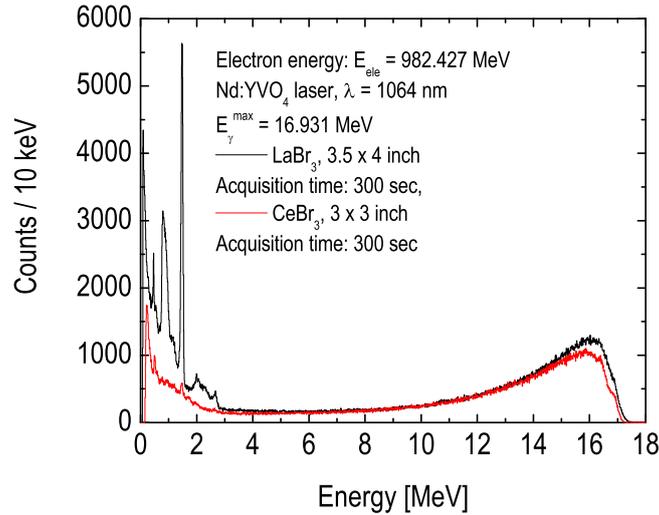


Figure 6: Energy spectra of 16.931 MeV maximum energy LCS  $\gamma$ -ray beam recorded with the LaBr<sub>3</sub>:Ce detector (black line) and with the CeBr<sub>3</sub> detector (red line). The large volume LaBr<sub>3</sub>:Ce detector presents a considerable intrinsic radioactivity component at energies between 0.7 - 2.5 MeV.

Since the LaBr<sub>3</sub> crystals are grown from natural lanthanum, the LaBr<sub>3</sub>:Ce detector contains 0.09 % of the radioactive isotope <sup>138</sup>La. With a half life of  $1.02 \times 10^{11}$  years, <sup>138</sup>La decays either by electron capture (65.5 %) to <sup>138</sup>Ba or by  $\beta^-$  (34.4 %) to <sup>138</sup>Ce. Also a small amount of contamination with radioactive <sup>227</sup>Ac is present in the scintillator. The radioactive decay products of these impurities contribute to the natural background spectrum of the LaBr<sub>3</sub>:Ce scintillation detector. The prominent peak at 1436 keV which can be clearly seen in Figure 1(b) results from the electron capture branch of <sup>138</sup>La. The group of peaks at energies between 1.8 MeV and 2.8 MeV are  $\alpha$ -decays from the <sup>227</sup>Ac decay chain. Additional features in the spectrum can be accounted for by decay products of the <sup>238</sup>U and <sup>232</sup>Th decay chains. We also show in Figure 6 an energy spectra of a 16.931 MeV maximum energy  $\gamma$ -ray beam measured with the LaBr<sub>3</sub>:Ce and with the CeBr<sub>3</sub> detector, without subtracting background.

### 3 Conclusions

We have investigated the energy resolution and response linearity for two types of scintillation detectors, a large volume  $\text{LaBr}_3\text{:Ce}$  detector and a  $\text{CeBr}_3$  one. While for an investigation made with nuclei emitted gamma rays of precisely determined energy the difference in the energy resolution of the two detectors is obvious,  $\text{LaBr}_3\text{:Ce}$  detector being superior to the  $\text{CeBr}_3$  one, when investigating with an  $\sim 1\%$  energy resolution  $\gamma$ -ray beam and considering the ability to observe separately the full energy, first and second escape peaks, the two detectors appear to have similar energy resolution. When investigating the detector response to the sharp high energy front characteristic to LCS  $\gamma$ -ray beams produced at NewSUBARU, the  $\text{CeBr}_3$  detector in the previous configuration has slightly better resolution than the  $\text{LaBr}_3\text{:Ce}$  one. The linearity of response with energy is also comparable for the two detectors in the present configuration. A clear disadvantage of the  $\text{LaBr}_3\text{:Ce}$  detector is its intrinsic radioactivity, which is not present in the  $\text{CeBr}_3$  crystal.

We conclude that both detectors are suitable to be used in the multidetector array proposed by the Gamma Above Neutron Threshold ELI-NP Research Group.