การแยกชัดพลังงานและเวลาของซินทิลเลเตอร์ผลึกเดี่ยว $Lu_3Al_5O_{12}$, Lu_2SiO_5 และ $LaBr_3$ โดปด้วย Ce^{3+}

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บทคัดย่อ

งานวิจัยนี้ได้ศึกษาเปรียบเทียบสมรรถนะของซินทิลเลเตอร์ผลึกเดี่ยว $Lu_3Al_5O_{12}$, Lu_2SiO_5 และ $LaBr_3$ โดปด้วย Ce^{3+} สำหรับการวัดรังสีแกมมา จากการวัดการแยกชัดพลังงานด้วยหลอดทวีคูณแสงเบอร์ XP5200B สำหรับรังสีแกมมาพลังงาน 662 keV พบว่ามีค่า 3.5% สำหรับ $LaBr_3$:Ce ซึ่งดีกว่าที่ได้จาก LuAG:Ce และ LSO:Ce ซึ่งมีค่าเป็น 6.7% และ 8.3% ตามลำดับ ได้ทำการวัดความไม่เป็นสัดส่วนของยีลด์แสงและการแยกชัดพลังงานที่แปรตามพลังงานรังสีแกมมา ซึ่งนำไปสู่การคำนวณหาค่าการแยกชัดในตัวของผลึก นอกจากนี้ยังได้ทำการวัดการแยกชัดเวลาโคอินซิเดนซ์สำหรับควอนตาประลัยพลังงาน 511 keV โดยทำการทดลองแบบโคอินซิเดนซ์ร่วมกับหัววัดรังสี BaF_2 ซึ่งพบว่า หัววัด $LaBr_3$:Ce, LSO:Ce และ LuAG:Ce ให้ค่าการแยกชัดเวลา เท่ากับ 204 ps, 212 ps และ 583 ps ตามลำดับ

คำสำคัญ : การแยกชัดพลังงานและเวลา / $LaBr_3$:Ce / LuAG:Ce / LSO:Ce / ความไม่เป็นสัดส่วนของยีลด์แสง / การวัดรังสีแกมมา / โฟโตแฟรกชัน / ซินทิลเลเตอร์

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Energy and Timing Resolutions of Ce^{3+} - Doped $Lu_3Al_5O_{12}$, Lu_2SiO_5 and $LaBr_3$ Single Crystal Scintillators

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Abstract

The performances of Ce-doped $Lu_3Al_5O_{12}$ (LuAG:Ce), Lu_2SiO_5 (LSO:Ce) and $LaBr_3$ ($LaBr_3$:Ce) single crystal scintillators were compared for γ-ray detection. For 662 keV γ-rays (^{137}Cs source), an energy resolution of 3.5% obtained for $LaBr_3$:Ce coupled to XP5500B PMT is much better than that of 6.7% and 8.3%, respectively, for LuAG:Ce and LSO:Ce, while its photofraction is worse than that of LuAG:Ce and LSO:Ce. The light yield non-proportionality and energy resolution versus γ-ray energy were measured and the intrinsic resolution of the crystals was calculated. The coincidence timing resolution, obtained in this work for 511 keV annihilation quanta, was 204, 212 and 583 ps, respectively, for $LaBr_3$:Ce-, LSO:Ce- and LuAG:Ce-based detectors in coincidence experiment together with a BaF_2 detector.

Keywords : Energy and Time Resolution / $LaBr_3$:Ce / LuAG:Ce / LSO:Ce / Non-proportionality of Light Yield / γ- Ray Detection / Photofraction / Scintillator

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1. Introduction

Growing interest in the development of new scintillator materials is pushed by increasing number of medical, industrial and scientific applications. During the last two decades, new types of scintillators, in particular, Ce-doped inorganic scintillators were intensively studied and some of them were successfully industrialized, for recent reviews see [1- 4].

$Y_3Al_5O_{12}$:Ce (YAG:Ce) single crystal was reported in the literature as a fast oxide scintillator [5,6]. Isostructural $Lu_3Al_5O_{12}$:Ce (LuAG:Ce) has a higher density (6.67 g/cm^3) than YAG:Ce (4.56 g/cm^3), which is advantageous in the case of high energy gamma-ray detection [7,8]. Its emission spectrum at room temperature (RT) is peaked around 525 nm. The scintillation light yield within 1 μs time gate is about 12,500 ph/MeV and 22,000 ph/MeV, respectively for LuAG:Ce and YAG:Ce crystals [9].

Lu_2SiO_5:Ce (LSO:Ce) [10] and $(Lu,Y)_2SiO_5$:Ce (LYSO:Ce) [11,12] have been developed as promising scintillators for positron emission tomography (PET) due to their desirable properties such as high density, fast decay time and high light output. LSO:Ce has a density of 7.4 g/cm^3 and an emission spectrum at RT is peaked around 410 nm. LSO:Ce exhibits a high light yield up to about 30,000 ph/MeV [13,14].

New Ce-doped $LaCl_3$ [15] and $LaBr_3$ [16] scintillators appeared with attractive properties due to high light output and very good energy resolution. $LaBr_3$:Ce has a density of 5.3 g/cm^3 and an emission spectrum at RT is peaked around 370 nm. $LaBr_3$:Ce exhibits a very high light yield above 60,000 ph/MeV and excellent energy resolution of about 3% for 662 keV γ-rays. The high energy resolution of $LaBr_3$:Ce is confirmed by its good proportionality characteristic and corresponding excellent intrinsic resolution [17].

In this paper, we present the comparative study on energy and timing resolutions of Ce-activated LuAG, LSO and $LaBr_3$ crystals under γ-ray excitation. The light yield non-proportionality and energy resolution versus γ-ray energy were measured and the intrinsic resolution of the crystals was calculated. The estimated photofraction in the pulse height spectra of 320, 662 and 835 keV γ-rays was determined and compared with the ratio of the cross-sections for the photoelectric effect to the total one calculated using the WinXCom program.

2. Experimental procedures

The LuAG:Ce and LSO:Ce crystals with size of 10×10×5 mm^3 were supplied by Crytur Ltd (Czech Republic) and CTI (USA), respectively. The $LaBr_3$:Ce crystal encapsulated in an aluminum can with size of Ø13×13 mm^2 was supplied by Saint-Gobain (France).

Photoelectron yield and energy resolution were measured by coupling the crystals to a Photonis XP5500B PMT using silicone grease. In order to maximize light collection, the crystals were covered with several layers of white Teflon tape in a configuration of a reflective umbrella. The signal from the PMT anode was passed to an ORTEC 113 preamplifier and then to a Tennelec TC244 spectroscopy amplifier. The measurements were carried out with 3 μs shaping time constant in the amplifier. The PC-based multichannel analyzer (MCA), Tukan 8k [18] was used to record energy spectra.

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each γ-peak, was measured by Bertolaccini method [19,20]. In this method the number of photoelectrons is measured by comparing the position of a full

energy peak of γ-rays detected in the crystals with that of the single photoelectron peak from the photocathode, which determines the gain of PMT. The measurements of light yield non-proportionality and energy resolution were carried out for a series of X/γ-rays emitted by different radioactive sources in the energy range from 16.6 to 1274.5 keV.

Coincidence timing measurements were performed using 511 keV annihilation quanta from a ^{22}Na source. Each crystal was coupled to a Photonis XP20D0 PMT. A BaF_2 crystal coupled to a Photonis XP20Y0Q/DA PMT was used as the reference detector. Its time resolution of 128 ps for 511 keV full energy peak selection in side channel was reported [21]. Upon irradiation of the crystals coupled to each PMT with 511 keV annihilation quanta, a signal from each PMT was processed with an ORTEC 935 constant fraction discriminator (CFD). Time spectrum was measured with an ORTEC 566 time to pulse height converter (TPHC) and recorded by the PC-based MCA. All measurements were carried out at RT.

3. Results and discussion

3.1 Light yield and energy resolution

Fig. 1 presents the pulse height spectra of 662 keV γ-rays from a ^{137}Cs source as measured with LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals at RT. The energy resolution of 3.5% obtained with $LaBr_3$:Ce is superior compared to the value of 6.7% and 8.3%, respectively, obtained with LuAG:Ce and LSO:Ce. This is due to a much higher photoelectron yield and very good proportionality of light yield for $LaBr_3$:Ce, see below. Note a higher photofraction in the spectrum obtained with LSO:Ce, as would be expected due to higher effective atomic number and density of the LSO:Ce material.

Photoelectron yield was determined using 662 keV γ-rays from a ^{137}Cs source. $LaBr_3$:Ce exhibits the photoelectron yield of 12,320 phe/MeV, which is much larger than that of 9,990 phe/MeV and 3,730 phe/MeV, respectively, for LSO:Ce and LuAG:Ce. The number of photoelectrons measured for studied crystals was recalculated to the number of photons assuming the quantum efficiency of 29%, 33%, and 18%, respectively, for the XP5500B PMT at the peak emission of $LaBr_3$:Ce (370 nm), LSO:Ce (410 nm) and LuAG:Ce (525 nm). The light yield of about 42,500 ph/MeV, 30,300 ph/MeV and 20,700 ph/MeV was obtained, respectively, for $LaBr_3$:Ce , LSO:Ce and LuAG:Ce.

The energy resolution (ΔE/E) of a full energy peak measured with a scintillator coupled to a photomultiplier can be written as [22]

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_{st})^2 \quad \textbf{(1)}$$

where δ_{sc} is the intrinsic resolution of the crystal, δ_p is the transfer resolution and δ_{st} is the statistical contribution of PMT to the resolution.

The statistical uncertainty of the signal from the PMT can be described as

$$\delta_{st} = 2.355 \times 1/N^{1/2} \times (1 + \varepsilon)^{1/2} \quad \textbf{(2)}$$

where N is the number of the photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.1 for an XP5500B PMT.

The transfer component depends on the quality of optical coupling of the crystal and PMT, homogeneity of quantum efficiency of the photocathode and efficiency of photoelectron collection at the first dynode. The transfer component is negligible compared to the other components of

the energy resolution, particularly in the dedicated experiments [22].

Overall energy resolution and PMT resolution can be determined experimentally. If δ_p is negligible, intrinsic resolution δ_{sc} of a crystal can be written as follows

$$(\delta_{sc})^2 = (\Delta E/E)^2 - (\delta_{st})^2. \quad \textbf{(3)}$$

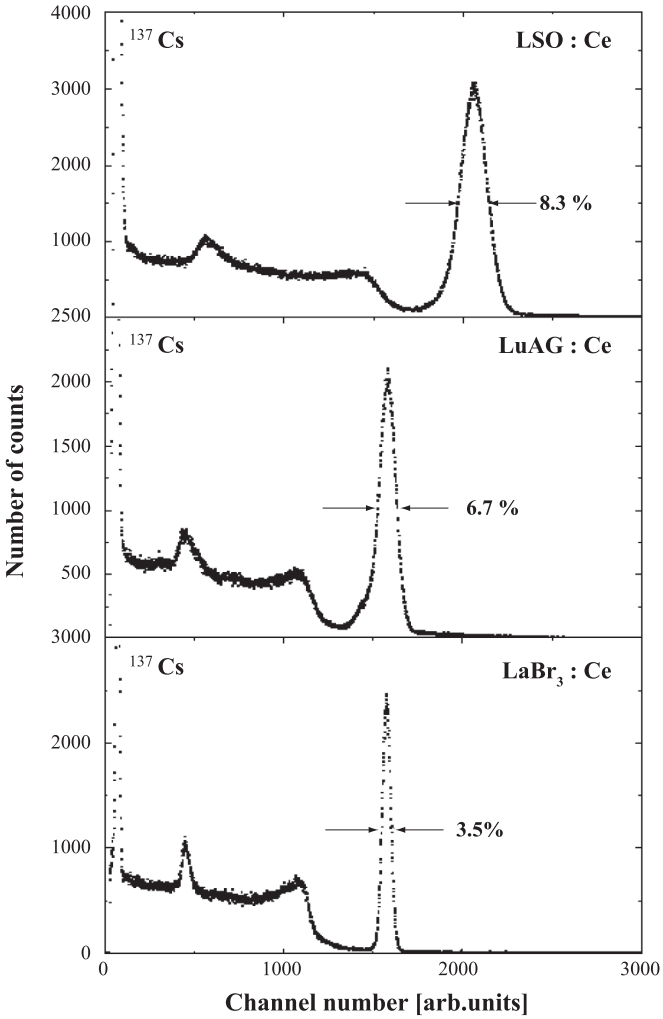

Fig. 1 Pulse height spectra of 662 keV γ - rays from a ^{137}Cs source as measured with LSO:Ce, LuAG:Ce and $LaBr_3$:Ce crystals.

Fig. 2 presents the overall energy resolution (ΔE/E) as a function of γ-ray energy, measured for the studied crystals. Over the energy range from 16.6 to 1274.5 keV, the overall energy resolution of $LaBr_3$:Ce is much better than that of LuAG:Ce and LSO:Ce. Fig.3 presents a direct comparison of the intrinsic resolution for the studied crystals. The results summarizing the photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the studied crystals are presented in Table 1.

Excellent energy resolution of $LaBr_3$:Ce is most likely associated with a lowest statistical error in the number of photoelectrons (δ_{st}) as well as a lowest contribution of intrinsic resolution (δ_{sc}). The energy resolution of LSO:Ce is worse than that of LuAG:Ce in spite of a lower contribution of δ_{st}. The reason is a much higher contribution of δ_{sc}. This result suggested looking at the non-proportionality of light yield versus γ-ray energy, as the non-proportionality of light yield is a fundamental limitation to δ_{sc} of the scintillators [22,23].

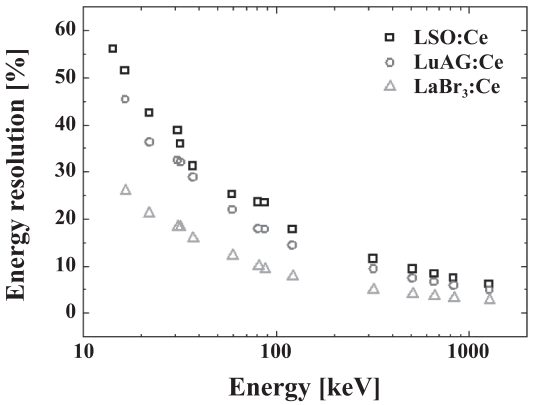

Fig. 2 Overall energy resolution of LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals.

Table 1 Photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the studied crystals

Crystal	Photoelectron yield [phe/MeV]	Light yield [ph/MeV]	Energy resolution [%]		
			ΔE/E	δ_{st}	δ_{sc}
LuAG:Ce	3,730 ± 200	20,700 ± 2,000	6.7	5.0	4.5
LSO:Ce	9,990 ± 500	30,300 ± 3,000	8.3	3.0	7.7
$LaBr_3$:Ce	12,320 ± 600	42,500 ± 4,000	3.5	2.7	2.2

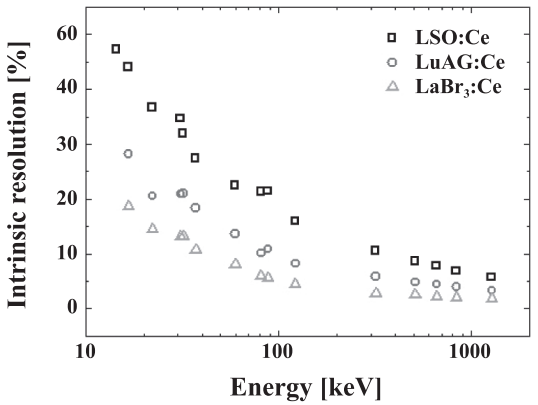

Fig. 3 Intrinsic resolution of LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals.

3.2 Non-proportionality of light yield

Non-proportionality of light yield is defined as the ratio of light yield measured at specific γ-ray energies relative to the light yield at the 662 keV γ-peak. Fig. 4 presents a comparison of the non-proportionality characteristics measured for all studied crystals. The most proportional scintillation response is obtained for $LaBr_3$:Ce with its non- proportionality only about 7% deviation from unity at 16.6 keV, which is much better than that of about 22% and 45%, respectively, for LuAG:Ce and LSO:Ce. The highest proportionality of the light yield for $LaBr_3$:Ce is related to a lowest contribution of δ_{sc}, see Fig. 2.This result confirms that the intrinsic resolution of a scintillator is strongly correlated with the non-proportionality in the scintillation response [22,23].

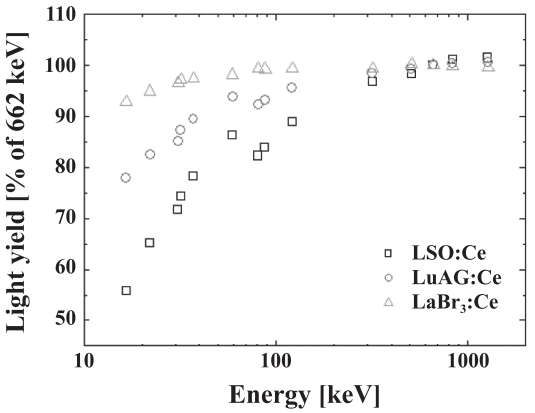

Fig. 4 Non-proportionality of light yield as a function of γ-ray energy for LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals.

3.3 Photofraction

The photofraction is defined here as the ratio of counts under the photopeak to the total counts of the pulse height spectrum as measured at a specific γ-ray energy. The photofraction for LSO:Ce, LuAG:Ce and $LaBr_3$:Ce at 320, 662 and 835 keV γ-rays is collected in Table 2. For a comparison, the ratios of the cross-sections (σ-ratio) for the photoelectric effect to the total one calculated using the WinXCom program [24] are also given. The LSO:Ce exhibits higher photofraction than LuAG:Ce and $LaBr_3$:Ce in a similar trend as the σ-ratio obtained from the WinXCom program. The reason is due to higher effective atomic number (Z_{eff} = 66) and density (ρ = 7.4 g/cm^3) of the LSO:Ce with respect to those of LuAG:Ce (Z_{eff} = 58.9; ρ = 6.67 g/cm^3) and $LaBr_3$:Ce (Z_{eff} = 46.9; ρ = 5.29 g/cm^3). However, the measured photofractions for both LSO:Ce and LuAG:Ce crystals are closer to the σ-ratios than the values for $LaBr_3$:Ce. It may be due to a larger size (a factor of 2.5) of the studied $LaBr_3$:Ce sample.

Table 2 Photofraction for $LaBr_3$:Ce, LuAG:Ce and LSO:Ce crystals

γ energy (keV) **Source**	**320** **^{51}Cr**	**662** **^{137}Cs**	**835** **^{54}Mn**	
Photof. (%)	44.7	16.2	12.7	$LaBr_3$:Ce
σ- ratio (%)	31.3	8.7	5.9	
Photof. (%)	64.8	28.0	22.3	LuAG:Ce
σ- ratio (%)	52.8	19.9	14.2	
Photof. (%)	70.9	35.8	27.5	LSO:Ce
σ- ratio (%)	58.8	24.0	17.4	

3.4 Coincidence Timing Resolution

Fig. 5 presents the coincidence timing spectra measured for LuAG:Ce, $LaBr_3$:Ce and LSO:Ce detectors in combination with a BaF_2 detector. The timing resolution was measured to be 583, 204 and 212 ps, respectively, for LuAG:Ce, $LaBr_3$:Ce and LSO:Ce detectors. The results of the measurements are collected in Table 3. The measured timing resolution, presented in the second column, is corrected for the contribution of the reference BaF_2 detector (128 ps) and shown in the third column. The last column shows the number of photoelectrons corresponding to the 511 keV peak.

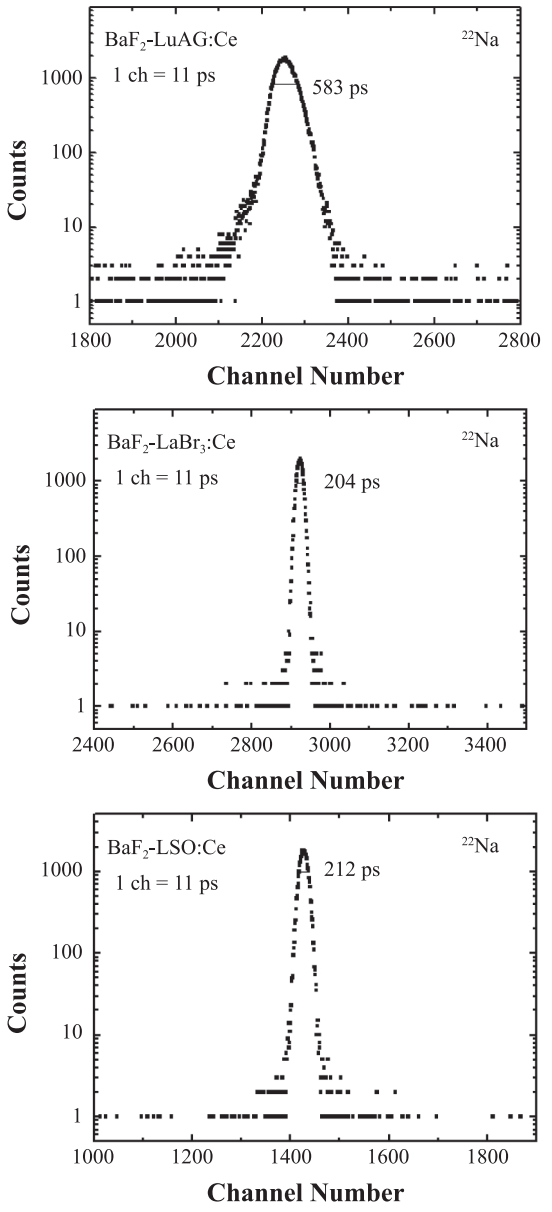

Fig. 5 Timing resolution spectra measured for LuAG:Ce, $LaBr_3$:Ce and LSO:Ce detectors in coincidence with a BaF_2 detector.

Note an excellent timing resolution of 159 ps for $LaBr_3$:Ce detector. No doubt that it is a consequence of very high light yield and a very fast scintillation pulse with decay time of 16 ns for $LaBr_3$:5%Ce [25]. LSO:Ce detector shows timing resolution of 170 ps, which is somewhat worse than that of $LaBr_3$:Ce detector. It is caused by lower light yield and slower scintillation decay (40 ns) of LSO:Ce. The timing resolution of 568 ps for LuAG:Ce detector is much worse with respect to both $LaBr_3$:Ce and LSO:Ce detectors. This is due to a much lower light yield of LuAG:Ce and the fact that only about 47% of its scintillation is emitted (as measured within 1 μs range) with decay time of 61 ns [26].

Table 3 Coincidence timing resolution for $LaBr_3$:Ce, LuAG:Ce and LSO:Ce detectors.

Crystal	Timing resolution, δ_t (ps)		N (phe) at 511 keV
	Measured	Tested detector	
$LaBr_3$:Ce	204 ± 5	159 ± 5	6220 ± 400
LuAG:Ce	583 ± 18	568 ± 18	1290 ± 80
LSO:Ce	212 ± 6	170 ± 9	3600 ± 200

4. Conclusions

The performances among Ce-activated $LaBr_3$, LuAG and LSO scintillators were investigated and compared in γ-ray spectrometry. The high energy resolution of 3.5% for 662 keV γ-rays obtained with $LaBr_3$:Ce is much better than the values of 6.7% and 8.3% obtained, respectively, for LuAG:Ce and LSO:Ce. The high light output and very good proportionality of $LaBr_3$:Ce are the important reasons behind its high energy resolution. It has a potential to replace NaI:Tl as the scintillator of choice for SPECT camera and γ-ray spectrometry. $LaBr_3$:Ce appears to be promising for PET due to its excellent timing resolution, but a relatively low density and photofraction make it less attractive than LSO:Ce and LYSO:Ce.

The main advantages of LSO:Ce are high light yield and detection efficiency for γ-rays. This fact and together with its excellent timing resolution make it an excellent scintillator for PET.

An advantage of LuAG:Ce is its superior energy resolution with respect to LSO:Ce. A drawback of LuAG:Ce is its very intense slow component in the scintillation pulse [27, 28], which is due to retrapping of charge carriers at shallow traps and appearance of the delayed radiative recombination at the Ce^{3+}-emission centers. It points to a chance to enhance its scintillation intensity of fast component determining both the energy and time resolutions, if related shallow traps could be suppressed. This fact together with the considerably fast scintillation decay (~60 ns) and moderate detection efficiency for γ-rays, would make LuAG:Ce the material of choice for γ-ray spectrometry and PET.

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