

Scintillation Properties of LuAG:Ce, YAG:Ce and LYSO:Ce Crystals for Gamma-Ray Detection

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Abstract—The scintillation properties of LuAG:Ce and YAG:Ce crystals were compared with LYSO:Ce crystal under γ -ray excitation. Light yield non-proportionality and energy resolution were measured with a Photonis XP5500B PMT. The energy resolution, obtained in this work for 662 keV γ -rays, was 6.7, 7.0 and 8.7%, respectively, for LuAG:Ce, YAG:Ce and LYSO:Ce detectors. The values reflect the influence of the light yield non-proportionality on the measured energy resolution. A fast component in the scintillation decay of LuAG:Ce crystal is faster than that of the YAG:Ce crystal, whereas the relative intensity of a fast component for YAG:Ce crystal is higher than that of LuAG:Ce crystal. The coincidence time resolution, obtained in this work for 511 keV annihilation quanta, was 660, 583 and 222 ps, respectively, for YAG:Ce, LuAG:Ce and LYSO:Ce detectors in coincidence experiment together with a BaF₂ detector. Time resolution was also discussed in terms of a number of photoelectrons and decay time of the light pulse.

Index Terms—Coincidence time resolution, decay time constant, energy resolution, LuAG:Ce, LYSO:Ce, non-proportionality, YAG:Ce.

I. INTRODUCTION

RESEARCH and development of new scintillator materials is mainly triggered by the growing needs of modern medical imaging and high energy physics. During the last two decades, new types of scintillators, in particular, Ce-doped inorganic scintillators were intensively studied and some of them were successfully developed for commercial production, for recent reviews see [1]–[4].

Ce³⁺-doped Y₃Al₅O₁₂ (YAG:Ce) single crystal was reported in the literature as a fast oxide scintillator [5], [6]. The density of YAG is 4.56 g/cm³ and its effective atomic number is 35. The emission spectrum at room temperature (RT) is peaked around 540 nm. Isostructural Lu₃Al₅O₁₂ (LuAG:Ce) has a higher density of 6.67 g/cm³ and effective atomic number is 58.9, which is advantageous in the case of high energy gamma-ray detection [7], [8]. The emission spectrum at RT is peaked around 525 nm.

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In both YAG:Ce and LuAG:Ce single crystals grown from high temperature melt (Czochralski and Bridgman techniques), the presence of the Y_{Al} and Lu_{Al} antisite defects (Y and Lu cations localized in octahedral sites of the Al cations) was proved both by experiments and theoretical calculations [9]–[11]. Such defects create trapping centers [12], [13] and the emission centers in near-UV region [14], [15]. The presence of defect—related shallow traps in these crystals could explain the degradation in their light yield and timing properties [16].

Ce³⁺-doped Lu₂SiO₅ (LSO:Ce) [17] and (Lu,Y)₂SiO₅ (LYSO:Ce) [18], [19] 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 density of 7.4 g/cm³ and effective atomic number is 66. LYSO:Ce with 10% atomic of Y has density of 7.1 g/cm³ and effective atomic number is 65. The emission spectra of LSO:Ce and LYSO:Ce at RT are peaked around 420 nm. In contrast to YAG:Ce and LuAG:Ce, the scintillation decay of LSO:Ce or LYSO:Ce shows only one component with the decay time somewhat longer with respect to the photoluminescence (PL) one (35 ns), reflecting the lack of shallow electron traps in silicates with respect to aluminum perovskite and garnet hosts [20].

In this work we present the scintillation properties of LuAG:Ce and YAG:Ce crystals compared with a LYSO:Ce crystal using photomultiplier tube (PMT) readout and under γ -ray excitation. The scintillation decay profiles of LuAG:Ce and YAG:Ce crystals were measured to investigate a contribution of slow components. Coincidence timing measurements were performed to discuss time resolution in terms of a number of photoelectrons and decay time of the light pulse.

II. EXPERIMENTAL DETAILS

The LuAG:Ce and YAG:Ce crystals used in this study were supplied by Crytur Ltd. (Czech Republic), with the size of 10 × 10 × 5 mm³. The LYSO:Ce crystal was supplied by Saint-Gobain (France), with the same size of 10 × 10 × 5 mm³. According to the manufacturer, the yttrium fraction in LYSO:Ce is about 10%.

A. Gamma-Ray Spectrometry

Photoelectron yield and energy resolution were measured by coupling the crystals to a Photonis XP5500B PMT with silicone grease. In order to maximize light collection, the crystals were wrapped in reflective, white Teflon tape on all sides (except the one coupled to the PMT). The signal from the PMT anode

was passed to an ORTEC 113 preamplifier and sent to a Tenelec TC244 spectroscopy amplifier. The measurements were carried out with 3 μ s and 12 μ s shaping time constants in the amplifier. The PC-based multichannel analyzer (MCA), Tukan 8 k [21] was used to record energy spectra. Gaussian functions were fitted to full energy peaks using procedures in the analyzer to determine their positions and FWHMs. It included also the analysis of complex double peaks, characteristic of K X-rays and those exhibiting an escape peak.

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each γ -peak, was measured by Bertolaccini method [22], [23]. 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.

B. Decay Time Measurements

The scintillation decays of LuAG:Ce and YAG:Ce crystals were measured by means of the delayed coincidence method [24], using a “fast-slow” coincidence setup as well as a “slow-slow” coincidence setup to determine the contribution of slow decay components [25]. Each tested sample was coupled to a Photonis XP2020Q PMT with silicone grease. Only the sides of the tested samples were covered with Teflon tape, leaving one surface opened to a Hamamatsu R5320 PMT (“fast-slow” coincidence), which was placed opposite to the XP2020Q at a distance of about 10 cm. The tested samples were irradiated with 662 keV γ -rays from a ^{137}Cs source, and their scintillation photons were detected with the R5320 PMT. Anode signals from two PMTs were sent to two ORTEC 935 constant fraction discriminators (CFD) used as the time pick-off units for an ORTEC 566 time-to-amplitude converter (TAC). Dynode signals from the PMTs were passed to ORTEC 113 preamplifiers and amplified with 0.5 μ s shaping time constants in the Tenelec TC 244 amplifiers. Two ORTEC 551 timing single channel analyzers (tSCA) were used to select the events, corresponding to the full energy peak of 662 keV γ -rays and the single photoelectron peak detected in the XP2020Q (start signal) and the R5320 (stop signal), respectively. Time spectra were recorded by the PC-based MCA (Tukan 8 k).

To measure the contribution of slow decay components we used a simplified “slow-slow” coincidence setup. The R5320 PMT was replaced by an R5600U PMT for its very low noise level in order to reduce the number of accidental coincidences. The start and stop input signals for the TAC were generated directly from ORTEC 551 tSCAs instead of commonly used CFDs.

C. Timing Measurements

Coincidence timing measurements with the LuAG:Ce, YAG:Ce and LYSO:Ce crystals were performed using 511 keV annihilation quanta from a ^{22}Na source. Each tested sample was coupled to a Photonis XP20D0 PMT. A BaF₂ 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 the side channel was reported in [26]. 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 CFD. Time spectra were measured with an ORTEC 566 TAC and recorded by the PC-based MCA (Tukan 8 k). In all the measurements, the fast-slow coincidence setup was used for a precise selection of the required energy windows. In addition to time resolution measurement, the number of photoelectrons produced in the PMT by the tested crystals was registered.

III. RESULTS

A. Photoelectron Yield and Energy Resolution

Fig. 1 presents the energy spectra of 662 keV γ -rays from a ^{137}Cs source measured with LuAG:Ce, YAG:Ce and LYSO:Ce detectors. The energy resolution of $6.7 \pm 0.3\%$ obtained with LuAG:Ce detector is comparable with the value of $7.0 \pm 0.3\%$ obtained with YAG:Ce detector. However, despite larger photoelectron yield [see below], LYSO:Ce detector exhibits much worse energy resolution of $8.7 \pm 0.4\%$. Note the lowest photofraction in the spectrum measured with the YAG:Ce detector, as would be expected due to a lower effective atomic number and density of the YAG:Ce crystal. In this respect, YAG:Ce is recommended for spectrometry of X-rays and low energy γ -rays.

The number of photoelectrons produced by the tested crystals in the XP5500B PMT was determined by relating the position of the full energy peak of 662 keV γ -rays to the position of the single photoelectron peak. Table I summarizes comparative measurements of photoelectron yield and energy resolution ($\Delta E/E$) of the tested crystals coupled to the XP5500B PMT, as measured at 3 μ s shaping time constant in the spectroscopy amplifier. Both LuAG:Ce and YAG:Ce samples showed comparable photoelectron yield and energy resolution. LYSO:Ce showed much worse energy resolution compared with both samples, which is not reflected by its much higher photoelectron yield.

Different energy resolutions observed with the tested samples suggested looking at non-proportionality of light yield versus γ -ray energy. The non-proportionality is defined here as the ratio of photoelectron yield measured at specific γ -ray energy relative to the photoelectron yield at 662 keV γ -peak.

Fig. 2 presents a comparison of the non-proportionality characteristics measured for the tested crystals. Over the energy range from 1274.5 keV down to 16.6 keV, both LuAG:Ce and YAG:Ce crystals exhibit a common non-proportionality of about 20%, which is much better than that of about 45% for LYSO:Ce crystal. This result shows that non-proportionality is influenced by the host crystal properties, as the doping agent is the same in all the tested crystals. Moreover, in our previous study [27] we have observed a significant difference of the non-proportionality curves measured for LuAG:Ce and LuAG:Pr crystals. It indicates that non-proportionality is also affected by the doping agents.

Common non-proportionality characteristics of the tested LuAG:Ce and YAG:Ce crystals should be reflected in the same intrinsic resolutions. Intrinsic resolution of a crystal is mainly associated with non-proportional response of the scintillator [28]. It is also affected by many effects such as inhomogeneities

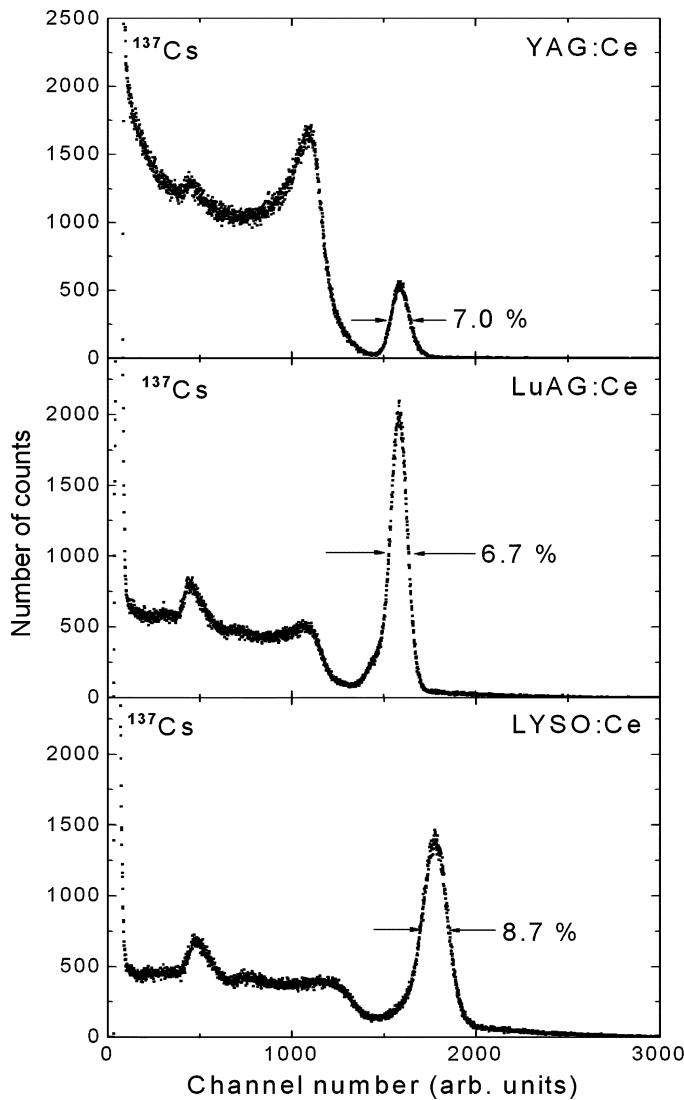


Fig. 1. Energy spectra of 662 keV γ -rays from a ^{137}Cs source measured with LuAG:Ce, YAG:Ce and LYSO:Ce detectors.

TABLE I
PHOTOELECTRON YIELD AND ENERGY RESOLUTION FOR THE 662 KEV
PHOTOPEAK OF THE TESTED CRYSTALS COUPLED TO THE XP5500B PMT,
MEASURED WITH 3 μs SHAPING TIME CONSTANT

Crystal	Photoelectron yield (phe/MeV)	$\Delta E/E$ (%)
LuAG:Ce	3720 ± 200	6.7 ± 0.3
YAG:Ce	3750 ± 200	7.0 ± 0.3
LYSO:Ce	7030 ± 400	8.7 ± 0.4

in the scintillator and non-uniform reflectivity of a reflecting cover of a crystal.

The energy resolution ($\Delta E/E$) of a full energy peak measured with a scintillator coupled to a photomultiplier can be written as [29]

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

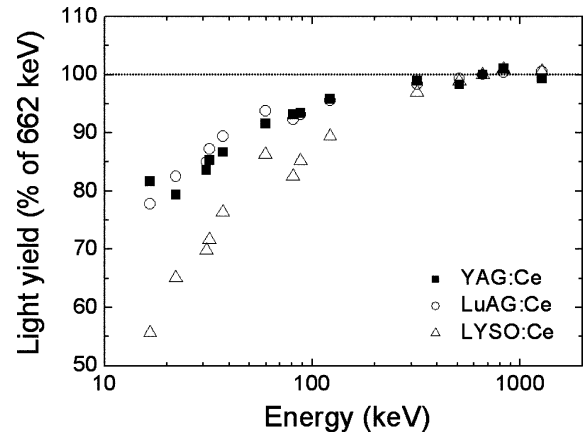


Fig. 2. Non-proportionality of the light yield as a function of γ -ray energy, measured with LuAG:Ce, YAG:Ce and LYSO:Ce crystals. Error bars are within the size of the points.

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

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

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

where N is the number of photoelectrons and ε is the variance of 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 [29].

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 :

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

Fig. 3 presents the intrinsic resolution calculated using (3) for the tested crystals. At energies above 60 keV, both LuAG:Ce and YAG:Ce crystals exhibit comparable intrinsic resolution, reflected in their common non-proportionality curves. For LYSO:Ce crystal, much poorer intrinsic resolution is also associated with its non-proportionality characteristic, influencing its inferior measured energy resolution. The relevant data of energy resolution at 662 keV photopeak for the tested crystals are presented in Table II.

In order to investigate a contribution of slow components in scintillation response for LuAG:Ce and YAG:Ce crystals, the photoelectron yield was also measured with 12 μs shaping time constant in the amplifier. Data measured with 3 μs and 12 μs (see Table III) indicate that LuAG:Ce crystal has more contribution from slow components compared with YAG:Ce crystal. This result should be reflected in a much higher contribution of

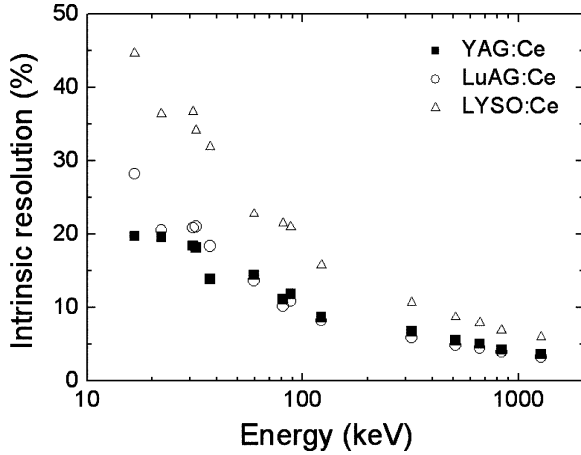


Fig. 3. Intrinsic resolution of LuAG:Ce, YAG:Ce and LYSO:Ce crystals versus energy of γ -rays. Error bars are within the size of the points.

TABLE II
THE ENERGY RESOLUTION AT 662 KEV PHOTOPEAK FOR THE TESTED CRYSTALS COUPLED TO THE XP5500B PMT, MEASURED WITH $3 \mu\text{s}$ SHAPING TIME CONSTANT

Crystal	Energy resolution (%)		
	$\Delta E/E$	δ_{st}	δ_{sc}
LuAG:Ce	6.7 ± 0.3	4.9 ± 0.3	4.5 ± 0.3
YAG:Ce	7.0 ± 0.3	4.9 ± 0.3	5.0 ± 0.3
LYSO:Ce	8.7 ± 0.4	3.6 ± 0.4	7.9 ± 0.4

TABLE III
THE PHOTOELECTRON YIELD FOR 662 KEV PHOTOPEAK FOR LUAG:CE AND YAG:CE CRYSTALS COUPLED TO THE XP5500B PMT, MEASURED WITH $3 \mu\text{s}$ AND $12 \mu\text{s}$ SHAPING TIME CONSTANTS

Crystal	Shaping time constant	Photoelectron yield (phe/MeV)	Photoelectron ratio
			$N(12\mu\text{s}) / N(3\mu\text{s})$
LuAG:Ce	$3 \mu\text{s}$	3720 ± 200	1.40
	$12 \mu\text{s}$	5200 ± 300	
YAG:Ce	$3 \mu\text{s}$	3750 ± 200	1.01
	$12 \mu\text{s}$	3800 ± 200	

slow components in the scintillation pulse for LuAG:Ce crystal compared with YAG:Ce crystal (see below).

B. Scintillation Decay Times

Scintillation decays of LuAG:Ce and YAG:Ce crystals measured at $1 \mu\text{s}$ range using the fast-slow coincidence setup are shown in Fig. 4. The fit by a two-exponential function, $I(t)$, shows that the initial decay in LuAG:Ce gets noticeably faster (61 ns decay time) than that of YAG:Ce (96 ns decay time). Moreover, the intensity of the fast component for LuAG:Ce ($I_f = 47\%$) is considerably lower than that for YAG:Ce ($I_f = 75\%$), which points to a much higher contribution of slow component for LuAG:Ce with respect to YAG:Ce in agreement with [16].

In order to investigate the contribution of slow components for LuAG:Ce and YAG:Ce crystals, we measured their scintillation decay time spectra using the slow-slow coincidence setup in the TAC range of $20 \mu\text{s}$ and $10 \mu\text{s}$, for LuAG:Ce and YAG:Ce,

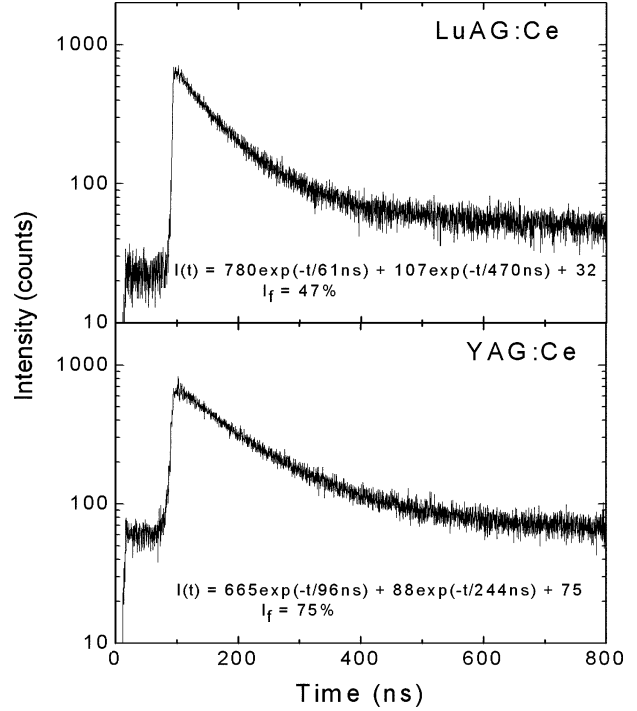


Fig. 4. Scintillation decay spectra of LuAG:Ce and YAG:Ce crystals under excitation with 662 keV γ -rays from a ^{137}Cs source as measured within $1 \mu\text{s}$ range. Experimental data are fitted by the function $I(t)$ displayed in the figure.

respectively. The scintillation decay time profiles obtained in these measurements are shown in Fig. 5. The fitting results are summarized in Table IV.

These measurements indicate that the content of slow components in the scintillation decay of LuAG:Ce crystal is much higher than that of YAG:Ce crystal. Such slow components usually arise due to retrapping of free charge carriers at shallow traps and results in the delayed recombination at the Ce^{3+} emission centers [16]. This result supports higher concentration and deeper thermal depth of antisite defect-related traps in LuAG host with respect to YAG host [12], [30], [31].

C. Coincidence Timing Resolution

Coincidence timing measurements with LuAG:Ce, YAG:Ce and LYSO:Ce crystals were performed using a ^{22}Na source. The results of the measurements are collected in Table V. The measured time resolution, presented in the second column, is corrected for the contribution of the reference detector and collected in the third column. The last column shows the number of photoelectrons corresponding to the 511 keV peak. Note the superior time resolution of $181 \pm 6 \text{ ps}$ for LYSO:Ce detector. No doubt that it is the effect of a large number of photoelectrons and a faster decay time constant (41 ns) as well as a single exponential light pulse for the LYSO:Ce crystal [32].

Despite a reasonably faster decay time of the fast component of scintillation for LuAG:Ce compared to YAG:Ce (61 ns vs 96 ns), time resolution is only slightly better. The reason for this is a poor intensity of the fast component for LuAG:Ce. The rest of scintillation light is mostly emitted within long decay modes (above $1 \mu\text{s}$), which do not contribute to timing properties.

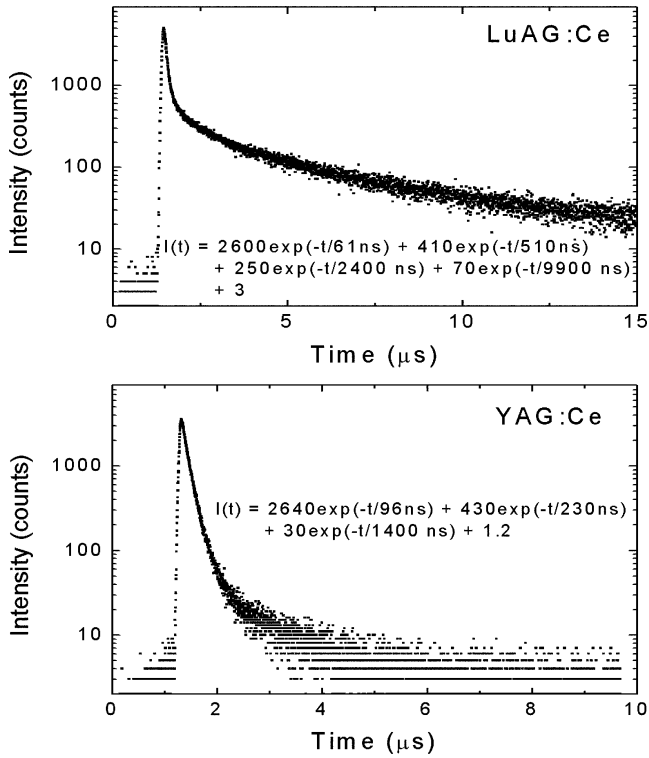


Fig. 5. Scintillation time profiles of LuAG:Ce and YAG:Ce crystals under excitation with 662 keV γ -rays from a ^{137}Cs source as measured in the time range of 20 μs and 10 μs , respectively. Experimental data are fitted by the function $I(t)$ displayed in the figure.

TABLE IV
DECAY TIME CONSTANTS AND THEIR INTENSITIES MEASURED FOR LUAG:CE
AND YAG:CE CRYSTALS

Decay mode	LuAG:Ce		YAG:Ce	
	τ (ns)	I (%)	τ (ns)	I (%)
1	61 \pm 4	12 \pm 2	96 \pm 3	68 \pm 5
2	510 \pm 50	13 \pm 2	230 \pm 10	22 \pm 3
3	2400 \pm 100	35 \pm 3	1400 \pm 100	10 \pm 2
4	9900 \pm 500	40 \pm 5	-	-

TABLE V
TIME RESOLUTION MEASURED WITH YAG:CE, LUAG:CE AND LYSO:CE
CRYSTALS COUPLED TO THE XP20D0 PMT FOR 511 KEV ANNIHILATION
QUANTA

Detector	Time resolution, δ_t (ps)		N(phe) at 511 keV
	Measured	Tested detector ^a	
YAG:Ce	660 \pm 20	647 \pm 20	1360 \pm 80
LuAG:Ce	583 \pm 18	568 \pm 18	1290 \pm 80
LYSO:Ce	222 \pm 6	181 \pm 6	3840 \pm 220

^acorrected for the contribution of the BaF₂ reference detector of 128 ps

Time resolution is approximately proportional to $(\tau/N)^{1/2}$, where τ is the decay time constant of the fast component in the light pulse and N is the number of photoelectrons [33]. The normalized time resolution, $\delta_t[N/\tau]^{1/2}$, should be constant for crystals with negligible rise time of the scintillation pulse. Thus, it is interesting to investigate the effect of rise time on the timing resolution for the tested crystals. The results of calculations are shown in Table VI. Interestingly, both LuAG:Ce and LYSO:Ce

TABLE VI
NORMALIZED TIME RESOLUTION CALCULATED FOR YAG:CE, LUAG:CE
AND LYSO:CE CRYSTALS COUPLED TO THE XP20D0 PMT FOR 511 KEV
ANNIHILATION QUANTA

Detector	δ_t (ps)	τ (intensity ^a) ns (%)	N ^b (phe) at 511 keV	$\delta_t(N/\tau)^{1/2}$ (ns·phe) ^{1/2}
YAG:Ce	647 \pm 20	96 (75%)	1020 \pm 60	2.11
LuAG:Ce	568 \pm 18	61 (47%)	606 \pm 40	1.79
LYSO:Ce	181 \pm 6	41 (100%)	3840 \pm 220	1.75

^aintensity of the fast component measured within 1 μs time range in TAC

^bcorrected for the intensity of the fast component

crystals exhibit a good agreement of the normalized time resolution, which is comparable with the value of 1.75 [ns·phe]^{1/2} and 1.62 [ns·phe]^{1/2}, respectively, for LuAG:Pr and LSO:Ce, obtained in our previous work [27]. In contrast, the normalized time resolution for the YAG:Ce crystal is larger than that of LYSO:Ce and LuAG:Ce crystals. It suggests to be the negative effect of a slow rise time in the scintillation pulse for YAG:Ce crystal, about 5 ns, obtained in [34]. Similar effect was observed for LaBr₃ crystal with 5% Ce doping [35], where normalized time resolution of 3.70 (ns·phe)^{1/2} was measured [27].

IV. CONCLUSION

In this work, the scintillation properties of LuAG:Ce and YAG:Ce crystals were studied and compared with LYSO:Ce crystal. The LuAG:Ce and YAG:Ce crystals show comparable energy resolution and intrinsic resolution due to common non-proportionality of the scintillation response as well as comparable photoelectron yield (at 3 μs shaping time constant in the amplifier). Interestingly, despite the larger photoelectron yield (by almost about 50%), energy resolution for LYSO:Ce detector significantly degrades as compared with LuAG:Ce and YAG:Ce detectors. The reason is much higher contribution of intrinsic resolution for LYSO:Ce crystal, reflected by a large non-proportionality response. It confirms that non-proportionality in the scintillation response is influenced by the host crystal materials.

The YAG:Ce crystal may be used for light charged particles, X-rays and low energy γ -rays, but it is not suitable for γ -rays above 300 keV due to its low effective atomic number and moderate density which limit the photopeak detection efficiency.

The measured coincidence time resolution for LuAG:Ce detector is much inferior compared with LYSO:Ce detector. It is the effect of a much lower number of photoelectrons in its fast component as measured with a blue-sensitive XP20D0 PMT, which mismatch to the peak emission at 525 nm for LuAG:Ce crystal. On the other hand LuAG:Ce crystal exhibits a very intense slow component (more than 53%) in the scintillation decay within 1 μs range, which is due to retrapping of free charge carriers at shallow traps and results in the delayed radiative recombination at the Ce³⁺-emission centers. It points to a chance to enhance its scintillation intensity of the fast component responsible for the timing resolution, if related shallow traps could be removed. This together with a suitable photodetector such as Si-APD and operated at low temperature to reduce its noise, further improvement of energy and time resolution for LuAG:Ce

could be achieved, and make it the scintillator of choice for medical applications.

Overall, the present measurements confirm that LYSO:Ce is a very promising scintillator for PET. It has high light output, high density and shows excellent timing resolution. LuAG:Ce appears to be also promising for PET, but its relatively low light output and poor timing resolution make it less attractive than LYSO:Ce and LSO:Ce. The drawback of LYSO:Ce, LuAG:Ce and other Lu-based scintillator materials (LSO:Ce, LuYAP, LuAP, etc.) consists in an intrinsic radioactivity ($\sim 300 \text{ counts} \cdot \text{s}^{-1} \cdot \text{cm}^{-3}$) due to the 2.6% natural abundance of ^{176}Lu radioactive isotope (half life $\sim 2.2 \times 10^{10}$ years) which makes them unacceptable for SPECT where there is no coincidence method to reject the intrinsic radiation.

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