Comparative studies of Lu$_3$Al$_5$O$_{12}$:Ce and Y$_3$Al$_5$O$_{12}$:Ce scintillators for gamma-ray detection

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The performance of Lu$_3$Al$_5$O$_{12}$:Ce (LuAG:Ce) and Y$_3$Al$_5$O$_{12}$:Ce (YAG:Ce) scintillators were compared under γ-ray excitation using photomultiplier tube (PMT) readout. 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 and 7.0%, respectively, for LuAG:Ce and YAG:Ce detectors. A fast component in the scintillation decay of LuAG:Ce crystal is faster than that of 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 583 and 660 ps, respectively, for LuAG:Ce- and YAG:Ce-based detectors in coincidence experiment together with a BaF$_2$ scintillation detector. Normalized coincidence time resolution was also discussed in terms of a number of photoelectrons and decay time of the light pulse.

1 Introduction

Growing interest in the development of new scintillator materials is pushed by increasing the 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 Refs. [1–4]).

Ce$^{3+}$-doped Y$_3$Al$_5$O$_{12}$:Ce (YAG:Ce) single crystal was reported in the literature as a fast oxide scintillator [5, 6]. The density of YAG:Ce is about 4.56 g/cm$^3$ and its effective atomic number is 35. The emission spectrum at room temperature (RT) is peaked around 540 nm. Isostructural Lu$_3$Al$_5$O$_{12}$:Ce (LuAG:Ce) has a higher density of 6.67 g/cm$^3$ 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. In both YAG:Ce and LuAG:Ce single crystals grown from high temperature melt (Czochralski and Bridgman techniques), the presence of 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 centres [12, 13] and the emission centres 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].

In this work we have performed a comparative study on energy resolution and timing properties of LuAG:Ce and YAG:Ce crystals using photomultiplier tube (PMT) readout and γ-ray excitation. The scintillation decay profiles of LuAG:Ce and YAG:Ce crystals were measured to investigate the 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.

2 Experimental details

2.1 γ-Ray spectrometry

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 mm$^3$. 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 Tennelec TC 244 spectroscopy amplifier. The measurements were carried out with 3 and 12 μs shaping time constants in the amplifier. The PC-based multichannel analyzer (MCA), Tukan 8k [17] 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 [18, 19]. 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.

2.2 Decay time measurements The scintillation decays of LuAG:Ce and YAG:Ce crystals were measured by means of the delayed coincidence method [20], using a ‘fast–slow’ coincidence setup as well as a ‘slow–slow’ coincidence setup to determine the contribution of slow decay components [21]. 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 137Cs source, and their scintillation photons were detected with the 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 137Cs 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 μs shaping time constant in the Tennelec 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 8k). The decay times and fractional intensities of the decay components contributed in the scintillation pulses were determined by fitting the measured time spectra with a sum of exponential components (FluoFit software package from PicoQuant GmbH).

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.

2.3 Timing measurements Coincidence timing measurements with the LuAG:Ce and YAG:Ce crystals were performed using 511 keV annihilation quanta from a 22Na 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 Ref. [22]. 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 8k). 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.

3 Results and discussion
3.1 Photoelectron yield and energy resolution

Figure 1 presents the energy spectra of 662 keV γ-rays from a 137Cs source measured with LuAG:Ce and YAG:Ce detectors. The energy resolution of 6.7 ± 0.3% obtained with the LuAG:Ce detector is comparable with the value of 7.0 ± 0.3% obtained with the YAG:Ce detector. Note the lower photoelectron fraction 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 full energy peak of 662 keV γ-rays to the single photoelectron peak. Table 1 summarizes comparative measurements of photoelectron yield and energy resolution (ΔE/E) of the tested crystals coupled to the XP5500B PMT, as measured at 3 μs shaping time constant in the spectroscopy amplifier. Both LuAG:Ce and YAG:Ce samples showed comparable photoelectron yield and energy resolution.

Table 1

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Photoelectron yield (phe/MeV)</th>
<th>ΔE/E (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LuAG:Ce</td>
<td>3720 ± 200</td>
<td>6.7 ± 0.3</td>
</tr>
<tr>
<td>YAG:Ce</td>
<td>3750 ± 200</td>
<td>7.0 ± 0.3</td>
</tr>
</tbody>
</table>

In our previous work [23], much poorer intrinsic resolution (40%) for LSO:Ce. It indicates that non-proportionality is influenced by the doping agents and the host crystal properties.

Comparative photoelectron yield and energy resolution observed with the tested samples suggested looking at non-proportionality of light yield versus γ-ray energies.

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.

Figure 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 crystals exhibit a common non-proportionality of about 20%. Moreover, in our previous study [23] we have observed a significant difference of the non-proportionality curves measured for LuAG:Pr and LSO:Ce crystals. Over the same energy range, the non-proportionality curves measured for LuAG:Pr and YAG:Ce crystals exhibit a common non-proportionality characteristic (40%), influencing its inferior measured energy resolution (8.2%).

The energy resolution (ΔE/E) of a full energy peak measured with a scintillator coupled to a photomultiplier can be written as [25]

\[
\left(\frac{\Delta E}{E}\right)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_a)^2,
\]

where \(\delta_{sc}\) is the intrinsic resolution of the crystal, \(\delta_p\) is the transfer resolution and \(\delta_a\) is the PMT contribution to the resolution.

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

\[
\delta_a = 2.355 \times \frac{1}{N^{1/2}} \times (1 + \epsilon)^{1/2},
\]

where \(N\) is the number of the photoelectrons and \(\epsilon\) 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 [25].

Overall energy resolution and PMT resolution can be determined experimentally. If \(\delta_p\) is negligible, intrinsic resolution \(\delta_{sc}\) of a crystal can be written as follows:

\[
(\delta_{sc})^2 = \left(\frac{\Delta E}{E}\right)^2 - (\delta_a)^2.
\]

Figure 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. The relevant data of energy resolution at 662 keV photopeak for the tested crystals are presented in Table 2. In our previous work [23], much poorer intrinsic resolution of LSO:Ce (7.5%) is also associated with its high non-proportionality characteristic (40%), influencing its inferior measured energy resolution (8.2%).

In order to investigate the contribution of slow components in scintillation response for LuAG:Ce and YAG:Ce crystals, the photoelectron yield was also measured with a 12 μs shaping time constant in the amplifier. Data...
Figure 3 Intrinsic resolution of LuAG:Ce and YAG:Ce crystals versus energy of γ-rays. Error bars are within the size of the points.

Table 2 The energy resolution at 662 keV photopeak for the tested crystals coupled to the XP5500B PMT measured with 3 μs shaping time constant.

<table>
<thead>
<tr>
<th>crystal</th>
<th>energy resolution (%)</th>
<th>δE/E</th>
<th>δsc</th>
</tr>
</thead>
<tbody>
<tr>
<td>LuAG:Ce</td>
<td>6.7 ± 0.3</td>
<td>4.9 ± 0.3</td>
<td>4.5 ± 0.3</td>
</tr>
<tr>
<td>YAG:Ce</td>
<td>7.0 ± 0.3</td>
<td>4.9 ± 0.3</td>
<td>5.0 ± 0.3</td>
</tr>
</tbody>
</table>

Table 3 The photoelectron yield for 662 keV photopeak for LuAG:Ce and YAG:Ce crystals coupled to the XP5500B PMT measured with 3 and 12 μs shaping time constants.

<table>
<thead>
<tr>
<th>crystal</th>
<th>shaping time constant (μs)</th>
<th>photoelectron yield (phe/MeV)</th>
<th>photoelectron ratio [N(12 μs)/N(3 μs)]</th>
</tr>
</thead>
<tbody>
<tr>
<td>LuAG:Ce</td>
<td>3</td>
<td>3720 ± 200</td>
<td>1.40</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>5200 ± 300</td>
<td></td>
</tr>
<tr>
<td>YAG:Ce</td>
<td>3</td>
<td>3750 ± 200</td>
<td>1.04</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>3800 ± 200</td>
<td></td>
</tr>
</tbody>
</table>

measured with 3 and 12 μs, and the results reported in Table 3, 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 slow components in the scintillation pulse for LuAG:Ce crystal compared with YAG:Ce crystal (see below).

3.2 Scintillation decay times Scintillation decays of LuAG:Ce and YAG:Ce crystals measured at 1 μs range using the fast–slow coincidence setup are shown in Fig. 4.

The decay times and fractional intensities of the contributed components in the scintillation pulses were obtained by fitting the measured spectra with a double – exponential function

\[ I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) + B, \]

where \(\tau_1\) is the decay time of the fast component, \(\tau_2\) is the decay time of the slow component, \(A_1\) and \(A_2\) are the corresponding initial intensities of the pulse shape components and \(B\) is a time independent background intensity. The results for both crystals are presented in Fig. 4. Note that for LuAG:Ce the fast component with decay time of 61 ns and intensity of 47% is followed by a slower component, with a 470 ns decay time and intensity of 53%. In contrast, the scintillation decay of YAG:Ce exhibits a fast component of 96 ns contributing with high intensity of 75% and a slow component of 244 ns with intensity of 25%. It points to a much higher contribution of slow component for LuAG:Ce with respect to YAG:Ce in agreement with Ref. [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 and 10 μs, for LuAG:Ce and YAG:Ce, respectively. The scintillation decay time profiles obtained in these measurements are shown in Fig. 5. Note that for LuAG:Ce the two additional components with decay times around 2.4 and 9.9 μs were found from the best fit of the measured curve while for YAG:Ce the only one additional component with decay time around 1.4 μs was found from the fit.

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 centres [16]. This result supports higher concentration and deeper thermal depth of antisite defect-related traps in LuAG host with respect to YAG host [12, 26–28].

3.3 Coincidence timing resolution Coincidence timing measurements with LuAG:Ce and YAG:Ce crystals were performed using a $^{22}$Na source. Figure 6 presents the coincidence time spectra obtained for 511 keV annihilation quanta with the tested crystals. The results of the measurements are collected in Table 4. 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.

Despite a comparable number of photoelectrons, considerably better time resolution for LuAG:Ce with respect to YAG:Ce can be attributed to its faster decay time (61 ns versus 96 ns) of the fast component.

Time resolution is approximately proportional to $(\tau/\mathcal{N})^{1/2}$, where $\tau$ is the decay time constant of the fast component in the scintillation pulse and $\mathcal{N}$ is the number of photoelectrons [29].

The normalized time resolution, $\delta_t(\mathcal{N}/\tau)^{1/2}$, 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 5. The third column shows the decay time and intensity of the fast component as measured within 1 μs time range. The number of photoelectrons (presented in Table 4) is corrected for the intensity of the fast component, which determines timing properties, and collected in the fourth column.

<table>
<thead>
<tr>
<th>crystal</th>
<th>time resolution, $\delta_t$ (ps)</th>
<th>$\mathcal{N}$ (phe) at 511 keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>YAG:Ce</td>
<td>660 ± 20</td>
<td>647 ± 20</td>
</tr>
<tr>
<td>LuAG:Ce</td>
<td>583 ± 18</td>
<td>568 ± 18</td>
</tr>
</tbody>
</table>

$^a$corrected for the contribution of the BaF$_2$ reference detector of 128 ps.
Table 5 Normalized time resolution calculated for YAG:Ce and LuAG:Ce crystals coupled to the XP20D0 PMT for 511 keV annihilation quanta.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>$\delta_1$ (ps)</th>
<th>$\tau$ (intensity)</th>
<th>$N^0$(phe)</th>
<th>$\delta_1$($N/\tau$)$^{1/2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>YAG:Ce</td>
<td>647 ± 20</td>
<td>96 (75%)</td>
<td>1020 ± 60</td>
<td>2.11</td>
</tr>
<tr>
<td>LuAG:Ce</td>
<td>568 ± 18</td>
<td>61 (47%)</td>
<td>606 ± 40</td>
<td>1.79</td>
</tr>
</tbody>
</table>

$^a$intensity of the fast component measured within 1 µs time range in TAC; $^b$corrected for the intensity of the fast component.

LuAG:Pr and LSO:Ce, obtained in our previous work [23]. In contrast, the normalized time resolution for the YAG:Ce crystal is larger than that of LSO: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 Ref. [30]. Similar effect was observed for LaBr$_3$ crystal with 5% Ce doping [31], where a normalized time resolution of 3.7 [ns · phe]$^{1/2}$ was measured in Ref. [23].

4 Conclusions In this work, the scintillation properties of LuAG:Ce and YAG:Ce crystals were compared with respect to energy and coincidence time resolutions. 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 amplifier). Interestingly, despite the larger photoelectron yield (by almost about 50%) for LSO:Ce, its energy resolution obtained in Ref. [23] significantly degrades as compared with LuAG:Ce and YAG:Ce detectors. The reason is much higher contribution of intrinsic resolution for LSO:Ce crystal, is reflected by a large non-proportionality response. It confirms that non-proportionality in the scintillation response is influenced by the host crystal materials as well as by the doping agents.

The YAG:Ce crystal may be used for light charged particles and X-rays/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 crystal (583 ps) is much inferior compared with the value of 210 ps obtained in Ref. [23] for LSO:Ce crystal. 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 in the scintillation decay, which is due to retrapping of free charge carriers at shallow traps and results in the delayed radiative recombination at the Ce$^{3+}$-emission centres. 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, similarly to LuAG:Ce single crystalline films [28], grown at significantly lower temperature as compared with single crystals. With a further improvement of crystal quality to enhance its scintillation performance, excellent energy and time resolution for LuAG:Ce detector could be achieved by using with a suitable photodetector (e.g. green-sensitive PMT, Si-APD), and make it the scintillation detector of choice for medical applications.

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References


