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Comparison of Lu$_3$Al$_5$O$_{12}$:Pr$^{3+}$ and Bi$_4$Ge$_3$O$_{12}$ scintillators for gamma-ray detection

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ABSTRACT

The scintillation properties of Lu$_3$Al$_5$O$_{12}$:Pr$^{3+}$ (LuAG:Pr) single crystal grown by the Czochralski method with praseodymium concentration of 0.19 mol% were investigated. For a comparison, a good quality Bi$_4$Ge$_3$O$_{12}$ (BGO) single crystal grown by Bridgemen method was also studied. The light yield and energy resolution were measured using photomultiplier tube (XP5200B PMT) readout. Moderate light yield of 15,900 photons per MeV was measured for the LuAG:Pr(0.19%) crystal. For 662 keV gamma rays ($^{137}$Cs source), an energy resolution of 6.5% obtained for LuAG:Pr(0.19%) is much better than that of 9.0% obtained for BGO. The light yield non-proportionality and energy resolution versus energy of gamma rays were measured and the intrinsic resolution of the crystals was determined after correcting the measured energy resolution for PMT statistics. The LuAG:Pr(0.19%) showed a good proportionality of the light yield within 5% over the energy range from 1274.5 keV down to 32 keV, which is much better than that of 14% for BGO. The photofraction was determined at 320 and 662 keV for both crystals and compared with the ratio of the cross-sections for the photoelectric effect to the total one calculated using WinXCOM program.

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1. Introduction

Research and development of new scintillator materials is mainly triggered by the growing needs of modern medical imaging and high-energy physics research. A number of Ce$^{3+}$-doped oxides based scintillators have been intensively studied, Gd$_2$SiO$_4$:Ce (GSO;Ce) (Takagi and Fukazawa, 1983), Lu$_2$SiO$_4$:Ce (LSO;Ce) (Melcher and Schweitzer, 1992), (Lu$_2$Y$_2$SiO$_4$:Ce (LYSO:Ce) (Cooke et al., 2000; Kimble et al., 2002), Lu$_2$SiO$_4$:Y$_2$O$_3$:Ce (LPS:Ce) (Pidig et al., 2003), and LuAlO$_3$:Ce (LuAP:Ce) (Lempicki et al., 1995; Wojtowicz et al., 1998), Lu$_3$Al$_5$O$_{12}$:Ce (LuAG:Ce) (Nikl et al., 2000) and Lu$_3$Al$_5$O$_{12}$:Pr (LuAG:Pr) (Drozdzowski et al., 2006). These scintillators exhibit desirable properties for γ-ray detection; high stopping power, high light output and fast scintillation decay.

Pr$^{3+}$-doped Lu$_3$Al$_5$O$_{12}$ (LuAG:Pr) single crystal was recently reported as a new, even faster and efficient scintillator (Ogino et al., 2006a,b; Nikl et al., 2005). The density of LuAG is of about 6.67 g/cm$^3$ and its effective atomic number is of about 59. LuAG:Pr has a moderate light yield of about 19,000 photons/MeV (ph/MeV) and a very good energy resolution of 4.6% (both values observed at 662 keV gamma excitation; Drozdzowski et al., 2008). These properties, and together with a fast scintillation decay time of 20 ns make LuAG:Pr an attractive scintillator for γ-ray detection. It was found that the light yield of LuAG:Pr increases with Pr concentration from 0.1 to 0.22 mol%, peaks at about 0.22–0.24 mol%, and then decreases (Ogino et al., 2006a; Yanagida et al., 2011). The decrease of the light yield with increasing Pr concentration from 1.5 to 10 mol% was also reported (Drozdzowski et al., 2009).

In this paper, we report on the detection properties of γ-rays for LuAG:Pr (0.19%) crystal and compared with those of BGO crystal over energy range from 32 to 1274.5 keV. The light yield non-proportionality and energy resolution versus energy of γ-rays were measured and the intrinsic resolution of the crystals was determined after correcting the measured energy resolution for PMT statistics. The estimated photofraction in the pulse height spectra of 320 and 662 keV γ-rays was determined for both crystals and compared with the ratio of the cross-sections for the photoelectric effect to the total one calculated using WinXCOM program.

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2. Experimental

The LuAG:Pr crystal was grown by Czochralski method at Crytur Ltd (Czech Republic) with Pr\(^{3+}\) concentration of 0.19 mol % in the crystal [Pejchal et al., 2008]. Plate of 7 x 7 x 1 mm\(^3\) was cut and polished for all measurements. For a comparison, the same size (7 x 7 x 1 mm\(^3\)) plate of high quality BGO single crystal grown by Bridgman method at the Shonan Institute of Technology (Fujisawa, Japan) was also measured.

Photoelectron yield and energy resolution were measured with a Photonis XP5200B photomultiplier tube (PMT) having a high blue photocathode sensitivity of 12.5 μA/mV. In order to maximize light collection, the samples were optically coupled by silicone grease to the PMT and covered with several layers of white Teflon (PTFE) tape in a configuration of a reflective umbrella. The signal from the PMT anode was passed to a Canberra 2005 preamplifier and then to a Tennelec TC243 spectroscopy amplifier. The measurements were carried out with 4 μs shaping time constant in the amplifier. The PC-based multichannel analyzer (MCA), Tukan 8k (Guzik et al., 2006) was used to record pulse height spectra. Gaussian functions were fitted to the full energy peak, using procedures in the MCA, to determine their position and energy resolution. It included also the analysis of complex double peaks, characteristic of Kα-rays and those exhibiting an escape peak.

The photoelectron yield, expressed as a number of photoelectrons released from the PMT photocathode per MeV (ph/eV) of energy deposited in the crystal, was determined by means of a single photoelectron method [Bertolacini et al., 1988; Muszynski et al., 1997]. 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 PMT photocathode.

The measurements of light yield non-proportionality and energy resolution were carried out for a series of γ-rays emitted by different radioactive sources (\(^{24}\)Am, \(^{133}\)Ba, \(^{51}\)Cr, \(^{137}\)Cs, \(^{58}\)Co, and \(^{22}\)Na) in the energy range from 32.1 to 1274.5 keV. All measurements were carried out at room temperature (RT).

3. Results and discussion

3.1. Energy spectra and photoelectron yield

Fig. 1 presents the pulse height spectra of 662 keV γ-rays from \(^{137}\)Cs source as measured with LuAG:Pr (0.19%) and BGO crystals at RT under identical operating conditions, whereas the values of photoelectron yield and energy resolution are summarized in Table 1. The energy resolution of 6.5% obtained with LuAG:Pr (0.19%) is much better than that of 9.0% observed with BGO. This is due to much higher photoelectron yield and very good proportionality of light yield for LuAG:Pr (0.19%), see below. Worse energy resolution for a studied LuAG:Pr (0.19%) than that reported in the literature for crystals with higher Pr concentration of about 0.23 mol% [Drozdowski et al., 2008; Swiderski et al., 2000] could be associated with its lower photoelectron yield, see below. In contrast, a good energy resolution of 9.0% observed for studied BGO crystal at RT is better than that of 10.0% reported for the 29 x 4 mm BGO crystal manufactured by Bicron (Moszynski et al., 2004). It could be associated with a higher photoelectron yield obtained for the studied BGO, see below. Note a higher photofraction in the spectra obtained with BGO crystal, as would be expected due to a higher effective atomic number and density of the BGO material.

Fig. 2 presents the pulse height spectra of 320 keV γ-rays from a \(^{51}\)Cr source as measured with LuAG:Pr(0.19%) and BGO crystals under identical operating conditions. The energy resolution of 9.8% obtained with LuAG:Pr(0.19%) is superior compared to the value of 13.3% obtained with BGO. Note the Kα-rays escape peak well separated from photopeaks for BGO crystal, which is due to its good energy resolution and high energy of bismuth Kα-rays of about 76 keV.

In the measurements with the XP5200B PMT, the studied LuAG:Pr(0.19%) sample shows a photoelectron yield of 3660 ± 200 ph/eV as measured at 662 keV γ-rays. Note a significantly lower photoelectron yield from the studied LuAG:Pr (0.19%) crystal, by about 20–40%, compared to that quoted in the literature [Drozdowski et al., 2008; Swiderski et al., 2006] for the crystals with higher Pr concentration of about 0.23 mol%. The photoelectron yield of 3660 ± 200 ph/eV measured for LuAG:Pr (0.19%) corresponds to a light yield of 15,500 ± 1600 ph/MeV, assuming 23% effective quantum efficiency (QE) of PMT for Pr\(^{3+}\) 5d → 4f emission (300–440 nm, peaks at 310 and 370 nm). This value is in agreement with the light yield measured for LuAG:Pr crystals with Pr concentration of 0.10, 0.18, and 0.22 mol% [Yanagida et al., 2011]. This result, and together with the decreasing light yield of LuAG:Pr crystals at higher Pr concentrations [Drozdowski et al., 2009], agrees with the conclusions of Ogino et al. [Ogino et al., 2006a] that the light yield of LuAG:Pr first increases with concentration, peaks at about 0.22–0.24% Pr, and decreases.

In the measurements with the XP5200B PMT, the studied BGO shows a photoelectron yield of 1810 ± 100 ph/eV/MeV. This value corresponds to a light yield of 8600 ± 800 ph/MeV at a QE of 21% for the BGO peak emission (480 nm). Note a significantly higher photoelectron yield, by about 25%, compared to that quoted for a 29 x 4 mm BGO crystal [Moszynski et al., 2004]. However, the scintillation light loss in BGO is rather large [Drozdowski et al., 2010]. To obtain an intrinsic yield Y0, which would be observed in the absence of scintillation light loss inside the material by optical absorption and photon scattering, we employ a simple two-ray

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Photoelectron yield (ph/eV)</th>
<th>Energy resolution (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LuAG:Pr</td>
<td>3660 ± 200</td>
<td>6.5 ± 0.2</td>
</tr>
<tr>
<td>BGO</td>
<td>1810 ± 100</td>
<td>9.0 ± 0.3</td>
</tr>
</tbody>
</table>

Table 1: Photoelectron yield and energy resolution (at 662 keV γ-rays) measured with LuAG:Pr(0.19%) and BGO crystals.
Fig. 2. Pulse height spectra of 320 keV γ-rays from a $^{51}$Cr source measured with LuAG:Pr (0.19%) and BGO crystals.

(*2R*) model proposed by Wojtowicz et al. (Wojtowicz et al., 2006), and assuming a loss coefficient $\mu = 0.8$ cm$^{-1}$ (Drozdowski et al., 2010) for the studied BGO. The intrinsic yield $Y_0 = 1960$ phe/MeV obtained for the studied BGO is almost two times larger than that reported for a $2 \times 2 \times 10$ mm$^3$ pixel BGO (Drozdowski et al., 2010). It indicates a high quality of the studied BGO crystal grown by the Bridgman method.

![Image of pulse height spectra](image)

Fig. 3. Overall energy resolution of LuAG:Pr and BGO crystals. Error bars are not shown for clarity of the figure.

$$\frac{\Delta E}{E} = \delta_0^2 + \delta_{t}^2 + (\delta_{nl}^2),$$  \hspace{1cm} (1)

where $\delta_0$ is the intrinsic resolution of the crystal, $\delta_{t}$ is the transfer resolution and $\delta_{nl}$ is the statistical contribution of PMT to the resolution.

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

$$\delta_{nl} = 2.355 \times 1/N^{1/2} \times (1 + e)^{1/2},$$  \hspace{1cm} (2)

where $N$ is the number of the photoelectrons and $e$ is the variance of the electron multiplier gain, equal to 0.1 for an XP5200B PMT.

Overall energy resolution and PMT resolution can be determined experimentally. If $\delta_{t}$ is negligible, intrinsic resolution $\delta_0$ of a crystal can be written as follows

$$\delta_0^2 = \left(\frac{\Delta E}{E}\right)^2 - \delta_{nl}^2.$$  \hspace{1cm} (3)

Fig. 4. Intrinsic resolution of LuAG:Pr and BGO crystals. Error bars are not shown for clarity of the figure.

The energy resolution ($\Delta E/E$) of a full energy peak measured with a scintillator coupled to a PMT can be written as (Moszynski et al., 2002)

$$\frac{\Delta E}{E} = \frac{\delta_{0}^2 + \delta_{t}^2 + \delta_{nl}^2}{E},$$

where $\delta_0$ is the intrinsic resolution of the crystal, $\delta_{t}$ is the transfer resolution and $\delta_{nl}$ is the statistical contribution of PMT to the resolution.

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

$$\delta_{nl} = 2.355 \times 1/N^{1/2} \times (1 + e)^{1/2},$$

where $N$ is the number of the photoelectrons and $e$ is the variance of the electron multiplier gain, equal to 0.1 for an XP5200B PMT.

Overall energy resolution and PMT resolution can be determined experimentally. If $\delta_{t}$ is negligible, intrinsic resolution $\delta_0$ of a crystal can be written as follows

$$\delta_0^2 = \left(\frac{\Delta E}{E}\right)^2 - \delta_{nl}^2.$$
Fig. 5. Non-proportionality of light yield (normalized to the value at 662 keV) as a function of γ-ray energy for LuAG:Pr and BGO crystals. Error bars are not shown for clarity of the figure.

(large contribution of δρ). The intrinsic resolution of 4.1% obtained for the studied LuAG:Pr (0.19%) is slightly worse than the value of about 3% reported for the LuAG:Pr crystals with Pr concentration of 0.18 and 0.23 mol% (Yanagida et al., 2011; Swiderski et al., 2009), which reflects to a lower quality of the studied crystal. It could be attributed to the inhomogeneities of dopant and some defects in the crystal.

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. The data presented in Fig. 5 exhibit a high proportional scintillation response of LuAG:Pr (0.19%). Over the energy range from 1274.5 keV down to 32 keV, the non-proportionality in its light yield is about 5% which is much better than that of about 14% for BGO. The higher proportionality of LuAG:Pr with respect to BGO is reflected in its lower value of intrinsic resolution, see Fig. 4. The proportionality behavior obtained in this measurement for a small LuAG:Pr sample (7 × 7 × 1 mm³) is about the same (decrease less than 6% down to 2 keV) as the ones measured for larger samples 10 × 10 × 5 mm³ (Swiderski et al., 2009) and 10 × 10 × 10 mm³ (Drozdowski et al., 2008). These results demonstrate that the non-proportionality response of LuAG:Pr crystal is not strongly affected by the crystal size. The high proportionality of LuAG:Pr is one of the main reasons (in conjunction with its high light yield) behind its good energy resolution.

3.3. Photofraction

The photofraction is defined here as the ratio of counts under the photopeak (including X-ray escape peak) to the total counts of the spectrum as measured at a specific γ-ray energy. The photofraction for LuAG:Pr and BGO at 320 and 662 keV γ-rays is collected in Table 3. For a comparison, the ratio of the cross-sections for the photoelectric effect to the total one calculated using the WinXCom program (Gerward et al., 2004) are also given. The data indicate that BGO shows higher photofraction than LuAG:Pr for both γ-rays energies in a same trend with the cross-section ratio (σ-ratio) obtained from the WinXCom program. It is due to higher effective atomic number (75 vs 59) and density (7.13 vs 6.67 g/cm³) of BGO with respect to LuAG:Pr.

4. Conclusion

The scintillation properties of small size LuAG:Pr (0.19%) and BGO crystals were studied and compared for γ-ray detection. Energy resolution of 6.5% at 662 keV γ-rays obtained with LuAG:Pr (0.19%) is much better than that of 9.0% for BGO. An advantage of BGO is its high density and effective atomic number, which results in a higher photofraction. The studied LuAG:Pr (0.19%) has moderate light yield of 15,900 ph/MeV and very good proportionality of light yield. Its relatively high value of the intrinsic resolution indicates that quality improvement is possible yet. On the other hand, a drawback of LuAG:Pr crystal is its very intense slow component in the scintillation decay (Niki et al., 2005; Drozdowski et al., 2008; Swiderski et al., 2009), which is due to retrapping of charge carriers at shallow traps and appearance of the delayed radiative recombination at the P³⁺-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 already recognized excellent energy resolution, fast (20 ns) scintillation decay and considerably high detection efficiency for γ-rays, would make LuAG:Pr the material of choice for γ-ray spectrometry and PET imaging.

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References


