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Light yield non-proportionality and energy resolution of \( \text{Lu}_{1.95}\text{Y}_{0.05}\text{SiO}_5:\text{Ce} \) and \( \text{Lu}_2\text{SiO}_5:\text{Ce} \) scintillation crystals

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Abstract

The scintillation response of the new Ce-doped lutetium-yttrium oxyorthosilicate (\( \text{Lu}_{1.95}\text{Y}_{0.05}\text{SiO}_5:\text{Ce} \), LYSO:Ce) crystal was investigated and compared to that of Ce-doped lutetium oxyorthosilicate (\( \text{Lu}_2\text{SiO}_5:\text{Ce} \), LSO:Ce) crystal. The light yield and energy resolution were measured using photomultiplier tube (XP5200B PMT) readout. For 662 keV \( \gamma \)-rays (\( ^{137}\text{Cs} \) source), the LYSO:Ce showed the light yield of 39,900 ph/MeV, which is higher than that of 35,900 ph/MeV obtained for LSO:Ce:Ce. The energy resolution of 8.2% obtained with LYSO:Ce is better than that of 10.6% obtained with LSO:Ce. The non-proportionality of the light yield and energy resolution versus \( \gamma \)-ray energy were measured and the intrinsic resolution of the crystals was calculated. Over the energy range from 16.6 keV to 1274.5 keV, the non-proportionality of about 35% for LYSO:Ce is better than that of about 42% for LSO:Ce. The photofraction was determined for both crystals and compared with the cross-sections ratio calculated using WinXCOM program.

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1. Introduction

Inorganic scintillators play a major role in many fields of radiation detection, including medical imaging, astrophysics, high energy physics and exploring resources like oil. The last decade has seen the introduction of several new high luminosity scintillators, in particular Ce-doped complex oxide crystals, that are promising candidates for these applications [1-4].

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Lu₂SiO₅:Ce (LSO:Ce) [5] and (Lu,Y)₂SiO₅:Ce (LYSO:Ce) [6] 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. Both crystals have the same emission spectra peaking at 420 nm and exhibit the highest light yield up to ~ 30,000 ph/MeV [6, 7].

In this paper, we present the gamma-ray detection properties of LYSO:Ce crystal and compare to those of LSO:Ce crystal. The photoelectron yield, energy resolution as a function of γ-ray energy and the non-proportional response were measured, and the intrinsic resolution of the both crystals was calculated. The estimated photofraction for both samples at 662 keV gamma peak will also be discussed.

1. Methodology

The LYSO:Ce and LSO:Ce crystals with same size of 10×10×2 mm³ were supplied by Proteus Inc. According to the manufacturer, the nominal cerium doped level is 0.2% for LSO:Ce sample and less than 1% for LYSO:Ce sample. The yttrium fraction in LYSO:Ce is about 2.5%.

Photoelectron yield and energy resolution were measured by coupling the crystals to a Photonis XP5200B PMT using silicone grease. In order to maximize light collection, the crystals were wrapped in a reflective, white Teflon tape on all sides (except the one coupled to the PMT). The signal from the PMT anode was passed to a CANBERRA 2005 preamplifier and was sent 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 [8] was used to record energy spectra.

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each γ-ray peak, was measured by Bertolaccini method [9,10]. 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. Results and discussion

3.1. Energy Spectra and Light Yield

Fig. 1 presents a comparison of the energy spectra for 662 keV γ-rays from a 137Cs source measured with LYSO:Ce and LSO:Ce crystals. The energy resolution of 8.2% obtained with LYSO:Ce is better than that of 10.6% obtained with LSO:Ce.

The number of photoelectrons produced by the studied crystals in the XP5200B 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 1 summarizes comparative measurements of photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the studied crystals. The number of photoelectrons measured for both crystals was recalculated to the number of photons assuming the quantum efficiency of 27% for the XP5200B PMT at the peak emission 420 nm for both crystals.

The studied LYSO:Ce showed the light yield of 39,900 ph/MeV. This value is slightly higher than the value of 34,100 ph/MeV measured with 1 cm³ sample in Ref [11]. Despite a comparable light output, LYSO:Ce shows a better energy resolution compared with LSO:Ce. It suggested looking at the non-proportionality of the light yield versus γ-ray energy.
Table 1. Photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the studied crystals as measured with the XP5200B PMT

| Crystal   | Photoelectron yield [phe/MeV] | Light yield [ph/MeV] | Energy resolution [%] |
|-----------|-------------------------------|----------------------|-----------------------|
| LYSO:Ce   | 10,780                        | 39,900               | 8.2                   |
| LSO:Ce    | 9,700                         | 35,900               | 10.6                  |

3.2. Non-proportionality of the Light Yield

Light yield non-proportionality as a function of energy is one of the most important reasons for degradation in energy resolution of established scintillators. The non-proportionality is defined here as the ratio of photoelectron yield measured at specific γ-ray energies relative to the photoelectron yield at the 662 keV γ-peak.

Fig. 2 presents the non-proportionality characteristics of LSO:Ce and LYSO:Ce crystals. Over the energy range from 16.6 keV to 1274.5 keV, the non-proportionality is about 35 % for LYSO:Ce, which is better than that of about 42% for LSO:Ce. The better in proportionality of LYSO:Ce is one of the important reasons behind its energy resolution. It appears so far, that all silicate scintillators (LSO, YSO, GSO or LGSO) exhibit large non-proportionality in the light yield [7, 11-13]. The non-proportionality characteristics of the studied crystals should be reflected in their intrinsic resolutions, as it is known that the non-proportionality in the light yield is a fundamental limitation to the intrinsic energy resolution [14, 15].
Fig. 2. Non-proportionality of the light yield as a function of $\gamma$-ray energy, measured with LYSO:Ce and LSO:Ce crystals. Error bars are within the size of the points.

3.3. Energy Resolution

The energy resolution ($\Delta E/E$) of a full energy peak measured with a scintillator coupled to a PMT can be written as [15]

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

where $\delta_{sc}$ is the intrinsic resolution of the crystal, $\delta_p$ is the transfer resolution and $\delta_{st}$ is the statistical contribution of PMT to the resolution. The statistical uncertainty of the signal from the PMT can be described as

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

where $N$ is the number of the photoelectrons and $\varepsilon$ is the variance of the electron multiplier gain, equal to 0.1 for an XP5200B 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 intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [14, 15] and many effects such as inhomogeneities in the scintillator which can cause local variations in the scintillation light output and non-uniform reflectivity of the reflecting cover of the crystal.

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

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

Fig. 3 presents the measured energy resolution versus energy of $\gamma$-rays for LYSO:Ce and LSO:Ce crystals. Other curves shown in Fig. 3 represent the PMT resolution calculated from the number of photoelectrons and the intrinsic resolution of the crystals calculated from Eq.3. Apparently, the energy resolution for the both crystals is mainly contributed by the intrinsic resolution over the whole energy range from 22 to 1274.5 keV.
Fig. 3. Energy resolution and contributed factors versus energy of LYSO:Ce and LSO:Ce crystals. Error bars are within the size of the points.

Fig. 4. Intrinsic resolution of LYSO:Ce and LSO:Ce crystals versus energy of γ-rays.

Fig. 4 presents a direct comparison of the intrinsic resolution for the studied crystals. Both crystals exhibit a comparable intrinsic resolution, reflected by a non-proportionality of the light yield (see Fig. 2).

To better understand the energy resolution of the studied crystals in γ-ray spectrometry, the contribution of various components to the overall energy resolution were analyzed for 662 keV photopeak, and the results are presented in Table 2. The second column gives $N$, the number of photoelectrons produced in the PMT. The third column gives $\Delta E/E$, the overall energy resolution at 662 keV photopeak. The PMT contribution ($\delta_{st}$) was calculated using Eq.2. From the values of $\Delta E/E$ and $\delta_{st}$, the intrinsic resolution ($\delta_{sc}$) was calculated using Eq.3.

Table 2. Analysis of the 662 keV energy resolution for LYSO:Ce and LSO:Ce crystals

| Detector | $N$ [electrons] | $\Delta E/E$ [%] | $\delta_{st}$ [%] | $\delta_{sc}$ [%] |
|----------|-----------------|-----------------|-------------------|-----------------|
| LSO:Ce   | 6400            | 10.6            | 3.2               | 10.1            |
| LYSO:Ce  | 7140            | 8.2             | 3.1               | 7.6             |
3.4. Photofraction

The photofraction is defined here as the ratio of counts under the photopeak to the total counts of the spectrum as measured at a specific \( \gamma \)-ray energy. The photofraction for LYSO:Ce and YSO:Ce at 662 keV \( \gamma \)-peak is collected in Table 3. For a comparison, the ratio of the cross-sections for the photoelectric effect to the total one was calculated using WinXCom program [16]. LSO:Ce shows slightly higher photofraction than LYSO:Ce in a same trend with the cross-section ratio (\( \sigma \)-ratio) obtained from WinXCom program, reflecting its slightly higher density and effective atomic number (\( Z_{\text{eff}} \)).

Table 3. Photofraction at 662 keV \( \gamma \)-peak for LSO:Ce and LYSO:Ce crystals

| Crystal    | \( Z_{\text{eff}} \) | Density (g/cm\(^3\)) | Photofraction (%) | \( \sigma \)-ratio (%) |
|------------|------------------------|------------------------|-------------------|------------------------|
| LSO:Ce     | 66                     | 7.4                    | 28.3              | 24.0                   |
| LYSO:Ce    | 63.5                   | 7.1                    | 26.1              | 22.6                   |

4. Summary

The scintillation properties of LYSO:Ce and LSO:Ce were studied for \( \gamma \)-ray detection. The energy resolution of 8.2\% for 662 keV \( \gamma \)-rays obtained with LYSO:Ce is better than that of 10.6\% for LSO:Ce. LYSO:Ce showed a better energy resolution compared with LSO:Ce. The reason is a higher photoelectron yield and a lower contribution of intrinsic resolution for LYSO:Ce. It confirms that the intrinsic resolution of the scintillators is correlated with the non-proportionality in the scintillation response.

In conclusion, the main advantages of LYSO:Ce is a high light yield. This together with high density and photofraction make it very promising scintillator for \( \gamma \)-ray detection and PET medical imaging.

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