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Luminescence and Scintillation Properties of Ce-Doped LYSO and YSO Crystals

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Abstract. The luminescence and gamma-ray detection properties of the new cerium-doped rare-earth scintillator lutetium-yttrium oxyorthosilicate ($Lu_{1.95}Y_{0.05}SiO_5$:Ce, LYSO:Ce) were investigated and compared to those of cerium-doped yttrium oxyorthosilicate (Y_2SiO_5:Ce, YSO:Ce) crystal. The light yield and energy resolution were measured using photomultiplier tube (PMT) readout. The non-proportionality of the light yield and energy resolution versus γ-ray energy were measured and the intrinsic resolution of the crystals was calculated. In spite of significant progress in light yield and luminescence properties, the energy resolution of LYSO:Ce appears to still suffer from an excess variance in the number of scintillation photons. The mass attenuation coefficient of LYSO:Ce and YSO:Ce for 662 keV gamma rays was also measured by transmission method and compared with the theoretical values calculated by WinXCom program.

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].

Lu_2SiO_5:Ce (LSO:Ce) [5] and $(Lu,Y)_2SiO_5$: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 luminescence and gamma-ray detection properties of LYSO:Ce crystal, and compare to those of YSO: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. The mass attenuation coefficient of LYSO:Ce and YSO:Ce for 662 keV gamma rays was also measured by transmission method and compared with the theoretical values calculated by WinXCom program.

Medthodology

The LYSO:Ce crystal with size of $10\times10\times2$ mm^3 was supplied by Photonic Materials. The YSO:Ce crystal with size of $10\times10\times5$ mm^3 was supplied by CTI. According to the manufacturer, the nominal cerium doped level is 0.2% for YSO:Ce sample and less than 1% for LYSO:Ce sample. The yttrium fraction in LYSO:Ce is about 2.5%.

UV excitation and emission spectra were obtained using a Hitachi F2500 fluorescence spectrophotometer. The measurements of the emission spectra were made using excitation wavelengths corresponding to the excitation bands found for the studied crystals. The excitation spectra were recorded at the emission wavelength 420 nm and at 500 nm to study the Ce2 site. All measurements were performed at room temperature.

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 TC244 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. 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 [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.

The total mass attenuation coefficients were determined by measuring the transmission of 662 keV gamma rays through studied crystals of known thickness. A narrow-beam setup in transmission geometry was used in this experiment, for more details see [11].

Results and Discussion

UV Excitation and Emission Spectra. Typical LYSO:Ce and YSO:Ce luminescence spectra are shown in Fig. 1. The spectra are normalized to the maximum peak for a better comparison. The excitation peaks at 270, 297 and 365 nm correspond to Ce1 site and following [12] lead to emission at 397 and 427 nm. The excitation bands at 326 and 375 nm are related to the Ce2 site [13] lead to emission at 460 nm. At the first glance, the luminescence spectra of both crystals are quite similar. However, when looking at the excitation spectra measured at 420 nm, the LYSO:Ce samples exhibits less pronounced peaks at 270 and 297 nm. It appears that the LYSO:Ce sample shows less intense band at 326 nm, which is related to Ce2 site.

Energy Spectra and Light Yield

Fig. 2 presents a comparison of the energy spectra for 662 keV γ-rays from a ^{137}Cs source measured with LYSO:Ce and YSO:Ce crystals. The energy resolution of 8.2% obtained with LYSO:Ce is better than that of 9.2% obtained with YSO:Ce. Note a higher photofraction in the spectrum measured with LYSO:Ce, as would be expected due to a higher effective atomic number and density of the LYSO:Ce crystal.

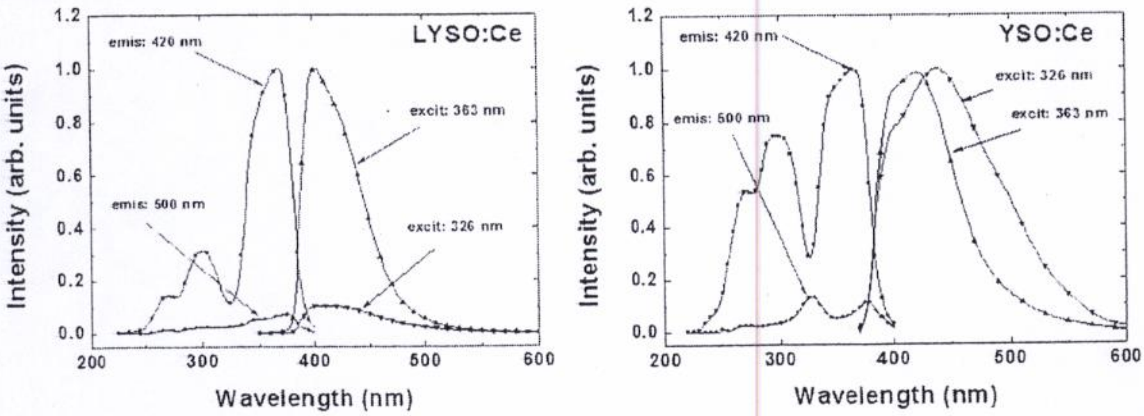

Fig. 1 UV excitation (left curves) and emission (right curves) spectra for LYSO:Ce and YSO:Ce crystals.

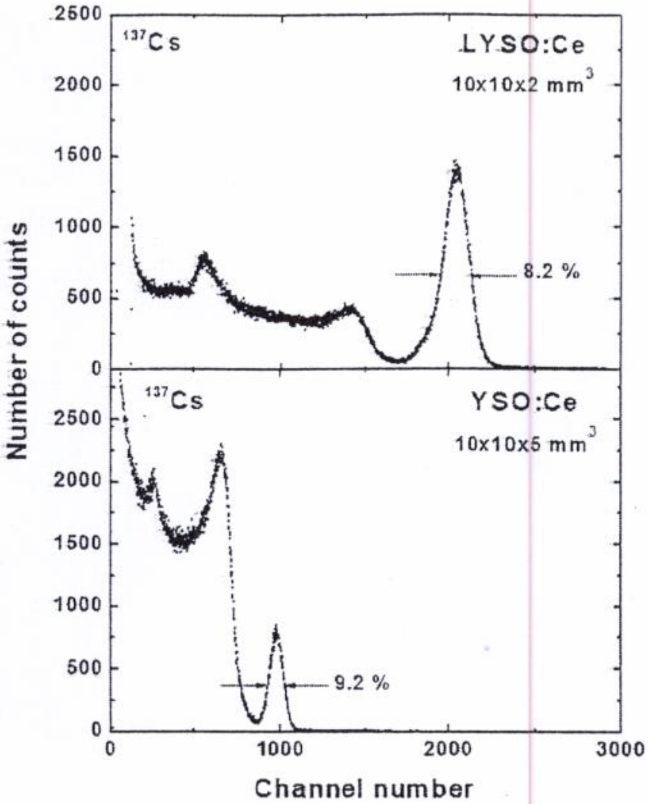

Fig. 2 Energy spectra of 662 keV γ - rays from a ^{137}Cs source measured with LYSO:Ce, and YSO:Ce crystals.

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 coupled to the XP5200B PMT, as measured at 4 μs shaping time constant in the spectroscopy amplifier. 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.

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 ± 1,100	39,900 ± 4,000	8.2 ± 0.3
YSO:Ce	4,340 ± 400	16,100 ± 1,600	9.2 ± 0.4

Note a significantly lower light yield of 16,100 ph/MeV for the studied YSO:Ce crystal, by about 30% compared with a small sample (3×3×20 mm^3) in Ref [14]. 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 cm^3 sample in Ref [15]. Interestingly, despite a much higher light output (a factor of 2), LYSO:Ce shows little gain in energy resolution compared with YSO:Ce. It suggested looking at the non-proportionality of the light yield versus γ-ray energy.

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 [16]. 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.3 presents the non-proportionality characteristics of YSO:Ce and LYSO:Ce crystals. Both crystals exhibit comparable non-proportional scintillation response curves, which is about 35% over the energy range from 1274.5 keV down to 22 keV. It appears so far, that all silicate scintillators (LSO, YSO, GSO or LGSO) exhibit large non-proportionality in the light yield [7, 13-15]. 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 [16,17].

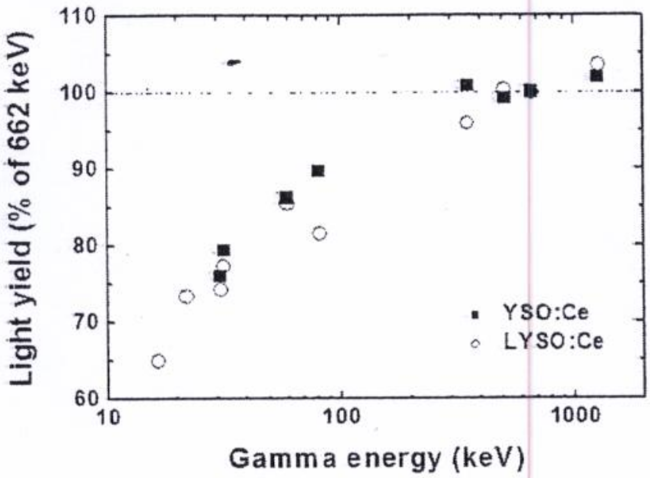

Fig. 3 Non-proportionality of the light yield as a function of γ-ray energy, measured with LYSO:Ce and YSO:Ce crystals. Error bars are within the size of the points.

Energy Resolution

The energy resolution (ΔE/E) of a full energy peak measured with a scintillator coupled to a PMT can be written as [17]

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

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

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

where N is the number of the photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.1 for an 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 transfer component is negligible compared to the other components of the energy resolution, particularly in the dedicated experiments [17].

The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [16,17] 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 δ_p is negligible, intrinsic resolution δ_{sc} of a crystal can be written as follows

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

Fig.4 presents the measured energy resolution versus energy of γ-rays for LYSO:Ce and YSO:Ce crystals. Other curves shown in Fig. 4 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.

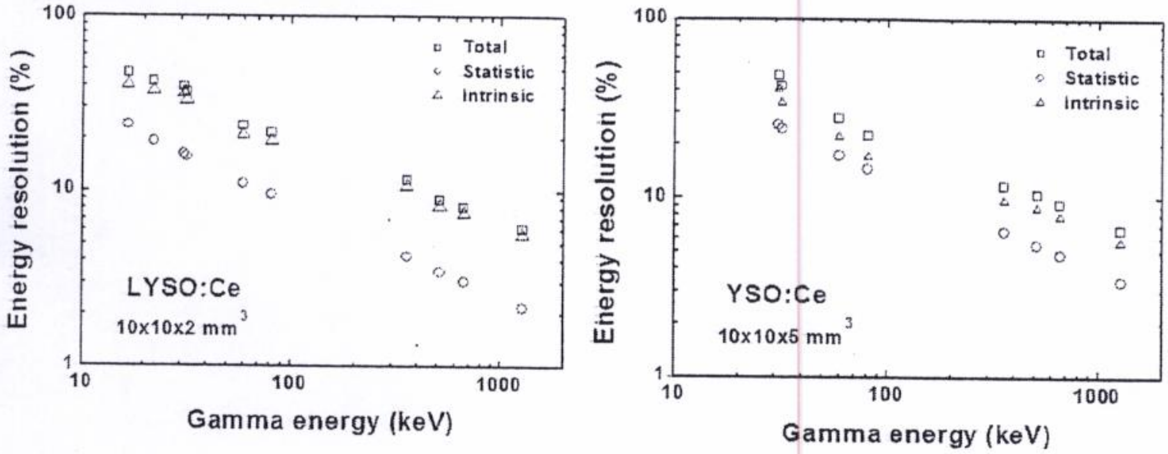

Fig. 4 Energy resolution and contributed factors versus energy of LYSO:Ce and YSO:Ce crystals. Error bars are within the size of the points.

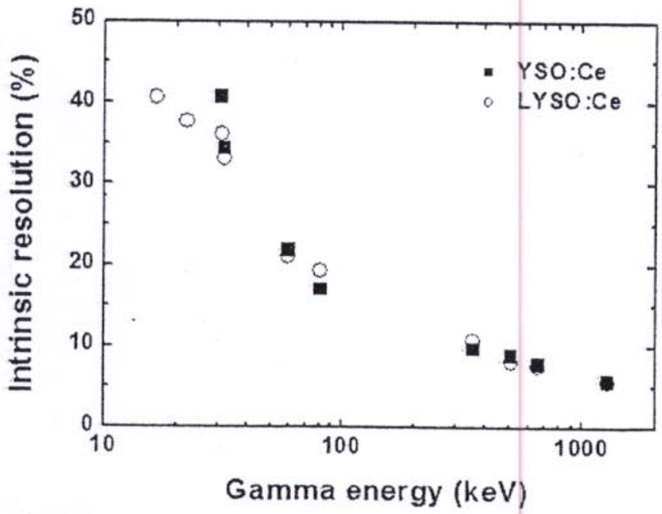

Fig. 5 Intrinsic resolution of LYSO:Ce and YSO:Ce crystals versus energy of γ-rays.

Fig. 5 presents a direct comparison of the intrinsic resolution for the studied crystals. Both crystals exhibit a comparable intrinsic resolution, reflected by a common non- proportionality of the light yield (see Fig. 3).

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 ΔE/E, the overall energy resolution at 662 keV photopeak. The PMT contribution (δ_{st}) was calculated using Eq.2. From the values of ΔE/E and δ_{st}, the intrinsic resolution (δ_{sc}) was calculated using Eq.3. The photoelectron yield of

LYSO:Ce is almost a factor of two higher than that of YSO:Ce. However, there is a little progress in energy resolution, as this is reflected in a large contribution of intrinsic resolution to the overall energy resolution for both studied crystals.

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

Detector	N [electrons]	$\Delta E/E$ [%]	δ_{st} [%]	δ_{sc} [%]
YSO:Ce	2870 ± 300	9.2 ± 0.4	4.8 ± 0.2	7.9 ± 0.3
LYSO:Ce	7140 ± 70	8.2 ± 0.3	3.1 ± 0.1	7.6 ± 0.3

Photofraction

The photofraction is defined here as the ratio of counts under the photopeak (including the escape peak) to the total counts of the spectrum as measured at a specific γ-ray energy. The photofraction for LYSO:Ce and YSO:Ce at 662 keV γ-peak is collected in Table 3. For a comparison, the ratio of the cross-sections for the photoelectric effect to the total one calculated using WinXCom program [18] are given too. The data indicate that LYSO:Ce shows much higher photofraction than YSO:Ce in a same trend with the cross-section ratio (σ-ratio) obtained from WinXCom program. The reason is due to much higher effective atomic number (Lu vs Y) and density (7.11 g/cm^3 vs 4.45 g/cm^3) of the LYSO:Ce crystal.. However, the σ-ratio is closer to the measured photofraction for LYSO:Ce than for YSO:Ce. It may be due to a larger size (a factor of 2.5) of YSO: Ce sample.

Table 3 Photofraction at 662 keV γ-peak for YSO:Ce and LYSO:Ce crystals.

Crystal	Z_{eff}	Density (g/cm^3)	Photofraction (%)	σ- ratio (%)
YSO:Ce	35	4.5	5.8 ± 0.6	3.1
LYSO:Ce	65	7.1	26.1 ± 2.6	22.6

Total Mass Attenuation Coefficient

A parallel beam of monoenergetic γ-rays is attenuated in absorber according to the Lambert-Beer law:

$$I = I_o \exp(-\mu_m \rho t), \tag{4}$$

where I_o and I are incident and transmitted intensities of gamma rays, respectively, μ_m is the mass attenuation coefficient, ρ is the density of the absorber, and t is the thickness of the absorber. The product $\mu_m\rho$ is called the linear attenuation coefficient. Theoretical values of the mass attenuation coefficients of mixture have been calculated by WinXCom program.

Table 4 shows the experimental $(\mu_m)_{ex}$ and theoretical $(\mu_m)_{th}$ values of the mass attenuation coefficients for YSO:Ce and LYSO:Ce crystals at 662 keV γ-rays. The agreement between experiment and theory is within the experimental uncertainty.

Table 4 Total mass attenuation coefficient (cm^2/g) at 662 keV γ-rays for YSO:Ce and LYSO:Ce crystals.

Crystal	$(\mu_m)_{ex}$	$(\mu_m)_{th}$	RD*(%)
YSO:Ce	7.46×10^{-2}	7.45×10^{-2}	0.13
LYSO:Ce	9.27×10^{-2}	8.92×10^{-2}	3.92

* Relative difference between $(\mu_m)_{ex}$ and $(\mu_m)_{th}$

Summary

The luminescence and scintillation properties of the new LYSO:Ce crystal were investigated and compared to YSO:Ce crystal. The two distinct Ce1 and Ce2 luminescence mechanisms previously identified in LSO:Ce are also present in both studied crystals, which is significantly reduced of Ce2 site for LYSO:Ce sample.

In spite of a much higher light yield, the energy resolution of LYSO:Ce is slightly superior than that of YSO:Ce. The main reason is due to a high contribution of intrinsic resolution, reflected by a large non-propportionality in the light yield, which seems to be a common feature in all silicate based scintillators. Moreover, inhomogeneities of Ce-doped and some defects in the crystals could affect the energy resolution, and the crystalline quality of these samples could be further improved. The experimental results of total mass attenuation coefficients are in good agreement with the theoretical values, calculated by WinXCom.

In conclusion, the main advantages of LYSO:Ce are high light yield, high density and photofraction which make it very promising scintillator for γ-ray detection and PET medical imaging.

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