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Comparison of $LaCl_3$:Ce and NaI(Tl) scintillators in γ-ray spectrometry

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Abstract

The commercial sample of $LaCl_3$:(9 ± 1)%Ce with the size of ∅25 × 25 mm was compared to NaI(Tl). A good light output of 9400 ± 100 photoelectrons per MeV and an energy resolution of 4.2 ± 0.2% for 662 keV γ-rays were measured with the $LaCl_3$:Ce crystal coupled to an XP3212 photomultiplier with bialkali photocathode. Below 122 keV, the energy resolution of $LaCl_3$:Ce was unexpectedly worse than that of NaI(Tl). The estimated photofraction of 17.9% at 662 keV is comparable to 21.4% for NaI(Tl) of ∅25 × 31 mm size. The radioactive background of the natural ^{138}La radioactive isotope was observed in $LaCl_3$:Ce, estimated to be about 1.07 c/s/cm^3 of the crystal. Moreover, the contamination of the crystal by α-emitting isotopes of the uranium series was discovered. The $LaCl_3$:Ce showed a good proportionality of the light yield versus energy within 3% down to 20 keV. The total and intrinsic energy resolutions are discussed.

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Keywords: $LaCl_3$:Ce; NaI(Tl); Scintillators; Gamma spectroscopy

1. Introduction

A high-energy resolution of 3.3% for 662 keV γ-rays of the recently developed $LaCl_3$:Ce crystal [1], reported for small samples below 1 cm^3 [1–3], prompted the present comparison of a larger sample of ∅25 × 25 mm size to NaI(Tl) in γ-ray spectrometry. $LaCl_3$:Ce has an emission maximum at 335 nm, a density of 3.86 g/cm^3 and is hygroscopic. Its effective Z is 59.5, as compared to 50.0 for NaI(Tl). The latter characteristics suggest that the detection efficiency of γ-rays and the photofraction of $LaCl_3$:Ce should be comparable to that of NaI(Tl). The reported high light output of 49000 ph/MeV and a short decay time of the main component of the light pulse of 25 ns allowed the obtaining of a time resolution comparable to that

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of BaF_2, as reported in Ref. [3]. The study of non-proportionality of the light yield versus energy showed a good proportionality down to 20 keV [4].

Most of the measurements were done on small (up to ∅16 × 19 mm) samples. The aim of this work is to compare detection properties of $LaCl_3$:Ce ∅25 × 25 mm, which has a useful size for a potential user with those of NaI(Tl) of a comparable size. Some aspects of intrinsic energy resolution will also be discussed.

2. Samples and setup

A ∅25 × 25 mm sample of $LaCl_3$, doped with $9 \pm 1\%$ of cerium, was delivered by Saint Gobain. It was assembled by the manufacturer in the aluminum case with a front glass window with a transmission of 90% down to 300 nm (short wavelength edge of scintillator emission). The crystal had a milky appearance such that the other flat side of the crystal could not be seen through the window.

An NaI(Tl) crystal with a Tl concentration of 0.048 weight% of ∅25 × 31 mm was delivered by the Institute of Single Crystals, Kharkov, Ukraine. The crystal was encapsulated in the standard way in the aluminum case with a glass window.

In all the measurements, the crystals were coupled to a 50 mm diameter Photonis XP3212 photomultiplier with a blue photocathode sensitivity of 11 μA/lmF. Energy spectra were measured using a preamplifier (ORTEC 113), a spectroscopy amplifier (Tennelec TC244) and a multichannel analyzer (Tukan). The sources were positioned on the axis of the scintillator and the photomultiplier. Moreover, the light output of the crystals was measured separately with the calibrated XP5200 PMT with a quantum efficiency of 26% at 350 nm.

3. Results

3.1. Energy spectra and light yield

Fig. 1 presents the comparison of the energy spectra of 662 keV γ-rays from a ^{137}Cs source, as measured with $LaCl_3$:Ce and NaI(Tl) crystals at a 3 μs shaping time constant. Note a very good energy resolution of $LaCl_3$:Ce of 4.2% in comparison to the 6.5% observed with NaI(Tl). Poorer energy resolution than that reported in Refs. [1–3] could be associated with the larger size of the tested sample, lower quality and its lower light output, see below. The same energy resolution was reported in Ref. [4] for the ∅16 × 19 mm crystal delivered by Saint Gobain. Photofractions in both the spectra correspond to 17.9% and 21.4%, respectively, reflecting the larger volume of the NaI(Tl) crystal.

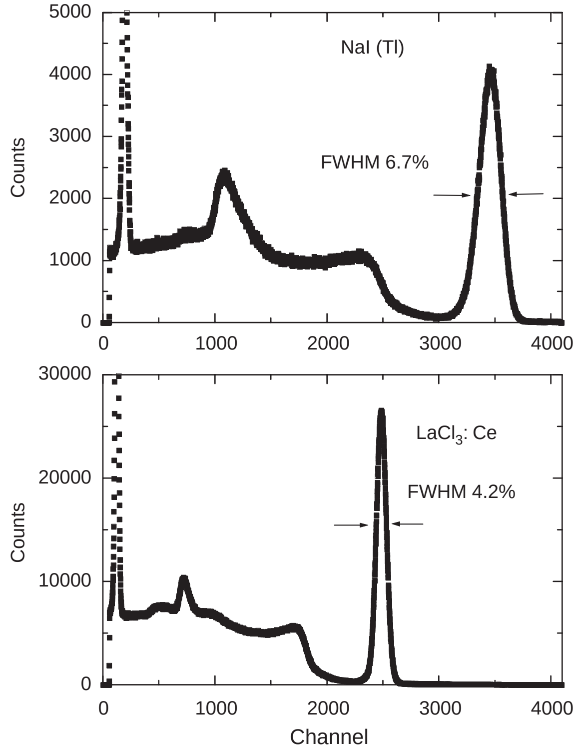

Fig. 1. The comparison of energy spectra of 662 keV γ-rays from a ^{137}Cs source, as measured with $LaCl_3$:Ce and NaI(Tl) crystals.

Fig. 2 presents the energy spectra of γ-rays from a ^{60}Co source. Note a very high-energy resolution of $2.88 \pm 0.09\%$ for the 1.33 MeV peak measured with $LaCl_3$:Ce crystal. Both 1.17 and 1.33 MeV peaks are fully resolved. The peak (1.33 MeV) to valley count ratio between 1.33 and 1.17 MeV is 34 for $LaCl_3$:Ce and 8 for NaI(Tl).

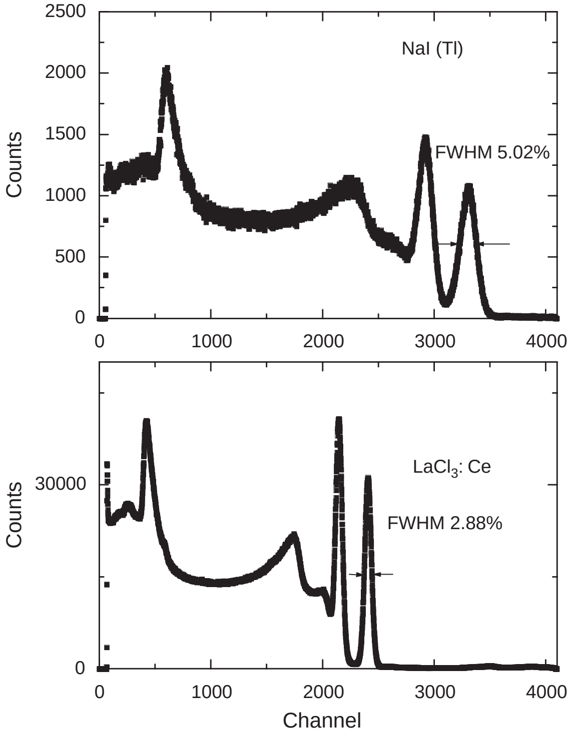

Fig. 2. Comparison of energy spectra for ^{60}Co γ-rays.

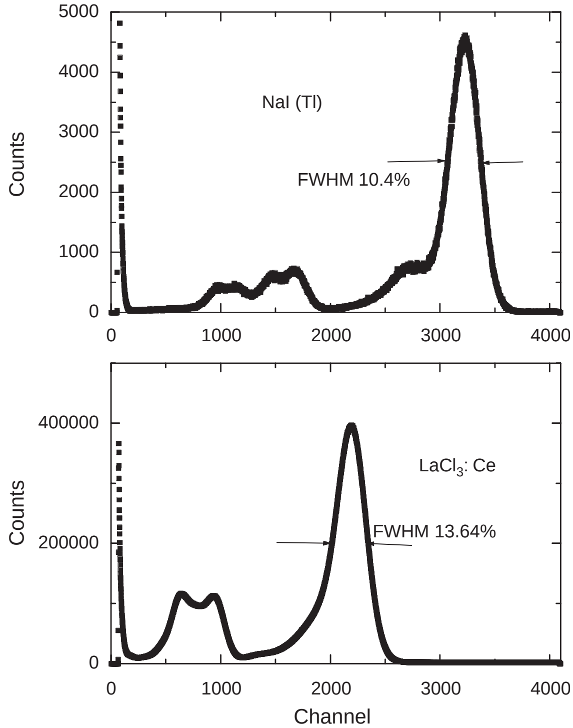

Fig. 3. Energy spectra of 59.6 keV γ-rays from a ^{241}Am source.

Fig. 3 presents the energy spectra of 59.6 keV γ-rays from an ^{241}Am source. An unexpected poorer energy resolution is recorded for the $LaCl_3$:Ce crystal. It suggests the necessity of studying carefully the non-proportionality of the light yield and a contribution of intrinsic resolution versus energy to measured energy resolution.

The number of photoelectrons per energy unit for each γ-peak was measured by the Bertolaccini et al. method [5], used further in Ref. [6]. In this method the number of photoelectrons is measured directly by comparing the position of the full energy peak of γ-rays detected in the crystal with that of the single photoelectron peak. The single photoelectron spectrum was measured without the crystals, before and after measurements of the energy spectra with the $LaCl_3$:Ce and NaI(Tl).

In the measurements with the XP3212, the $LaCl_3$:Ce had a photoelectron output of 9400 ± 100 photoelectrons per MeV (phe/MeV) as measured at 662 keV γ-rays with 3 μs shaping time constant. The tested NaI(Tl) showed a photoelectron yield of 12750 ± 130 phe/MeV.

The light output expressed in photons per MeV (ph/MeV) was determined separately by using a calibrated Photonis XP5200 PMT. These NaI(Tl) crystals have about 15% higher light output than typical crystals usually measured by us with this photomultiplier. The photoelectron yield of 9300 ± 400 phe/MeV measured for $LaCl_3$:Ce corresponds to 35800 ± 2000 ph/MeV at a QE of the peak emission of 26%. For NaI(Tl), the number of photoelectrons was equal to 12800 ± 500 phe/MeV. Note a significantly lower light output from the tested $LaCl_3$:Ce crystal, by about 30%, compared to that quoted for small samples in Refs. [1–3]. In contrast, the tested NaI(Tl) exhibited a very high light output, 20% larger than that measured by Holl et al. [7] for many different samples of NaI(Tl). It is most likely due to quantum efficiency of XP3212 photomultiplier being higher then our standard calibrated XP2020 ∅ for high wavelengths.

3.2. Intrinsic spectra of scintillation

$LaCl_3$:(9 ± 1)%Ce contains an admixture of the natural radioactive isotope ^{138}La with contribution of 0.09% of the total La content. The ^{138}La, with a half-life of 1.05×10^{11} years, decays to ^{138}Ba by electron capture with a characteristic γ line of 1435.8 keV (66.4% of decay intensity) and by β^- decay of a maximum energy of 1044 keV to ^{138}Ce with characteristic γ-rays of 788.8 keV (33.6% of decay intensity). In the internal energy spectrum of $LaCl_3$:Ce, one can expect to observe a continuum spectrum of β-rays summed up with 788.8 keV γ-rays, and X-rays of Ba from EC decay and 1435.8 keV γ-line shifted up by internal summing with Ba X-rays. For a $\varnothing 25 \times 25$ mm crystal of $LaCl_3$:Ce of 3.86 g/cm^3 density, it should give 13.8 decays per second, that is, 1.07 decays per cm^3/s. The spectrum of intrinsic scintillation collected for 114 h is shown in Fig. 4. The 1435 keV γ line is seen around channel 1600.

We detected about 50 counts per second (cps), including laboratory background, at the gain settings shown in Fig. 4. A larger counting rate than expected is because of the peaks to the right of the 1435 keV γ-peak, identified as those due to α-particles originating from a decay of U series isotopes ^{227}Th, ^{223}Ra, ^{219}Rn and ^{215}Po, and ^{214}Po from ^{238}U decay series. Characteristic γ-rays from α-decay were detected by Ge detector. The highest α energy and the corresponding peak at about channel 2800 is at 7686 keV from ^{214}Po of the ^{238}U series. Note that Rn gas and the following isotopes of the series are not in age equilibrium in the sample, since it was heated to the melting point recently. We have also checked for possible rare-earth alpha emitters, but the assignment did not fit the spectrum. Based on this assignment of α particles, we estimate α/γ ratio as 0.33 ± 0.01 for the energy of 7.7 MeV, but for lower energy α-particles, the number may be lower. The observed α counting rate was 0.89 counts/cm^3/s.

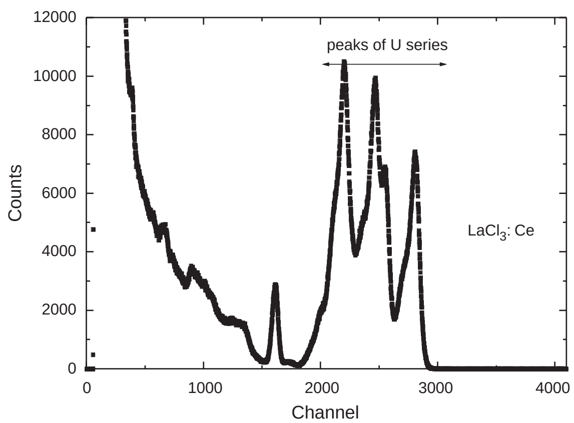

Fig. 4. Energy spectrum of intrinsic scintillation of $LaCl_3$:Ce. 10000 counts correspond to 88 counts per hour. Note strong peaks due to α-particles identified as those coming from a decay of U series.

An additional γ-spectrum taken on a Ge detector showed characteristic lines of ^{138}La and X-rays of ^{138}Ba, according to final states of the ^{138}La decay; the ^{40}K line of 1460 keV was also detected, but with much lower intensity.

The relative abundance of U to La is estimated at 1.4 ppm. In the case of GSO, significant contamination by U series isotopes was connected with the production of the small crystal in the welded crucible of Ir [12]. Note that NaI(Tl) has no naturally radioactive components.

3.3. Non-proportionality of light yield and intrinsic energy resolution versus energy

The non-proportionality curve for $LaCl_3$:Ce and NaI(Tl) is shown in Fig. 5. The non-proportionality is defined here as the phe number yield

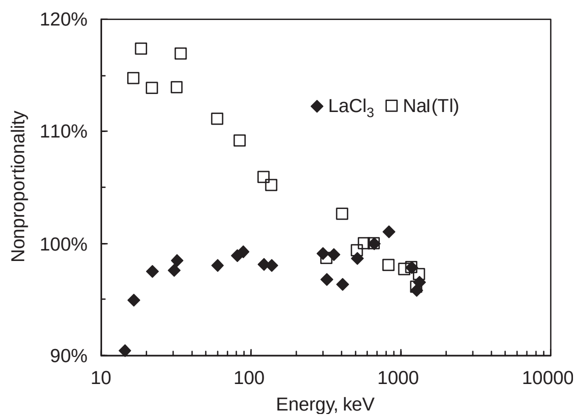

Fig. 5. Non-proportionality of the light yield of $LaCl_3$:Ce and NaI(Tl). The curve error is 3%.

measured at a specific γ-ray energy relative to the phe number yield of the 662 keV γ-peak. $LaCl_3$:Ce is clearly superior to NaI(Tl) in terms of non-proportionality.

The energy resolution, $\Delta E/E$, of the full energy peak measured with a scintillator coupled to a photomultiplier can be written as [8]

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

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

The statistical uncertainty of the signal from the PMT is described as

$$\delta_{st} = 2.35 \times 1/N^{1/2} \times (1+\varepsilon)^{1/2} \tag{2}$$

where N is the number of photoelectrons, and ε is the variance of the electron multiplier gain. In the case of the XP3212, this was determined as 0.12, analyzing the single photoelectron peak.

The transfer component, δ_p, in modern scintillation detectors is negligible compared to the other components of the energy resolution [8].

The energy resolution of $LaCl_3$:Ce and NaI(Tl) measured with the same photomultiplier versus energy is shown in Fig. 6. The curve for NaI(Tl) has a known step-like shape [8], while the energy resolution of $LaCl_3$:Ce is approximately inversely proportional to the square root of the energy as reported recently in Ref. [4]. The curves confirm observations presented in Figs. 1–3. The energy resolution of $LaCl_3$:Ce above 122 keV is superior, while that below 122 keV is poorer as compared to those measured with NaI(Tl) crystals.

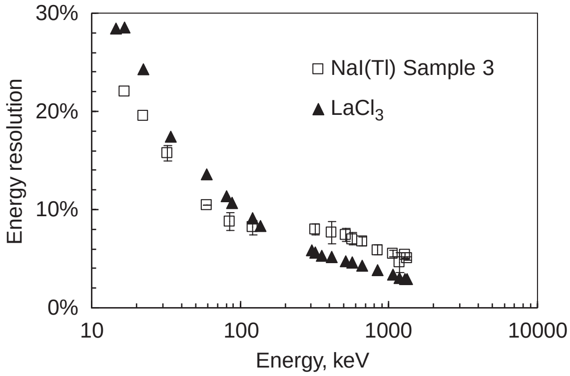

Fig. 6. Energy resolution of $LaCl_3$:Ce and NaI(Tl) versus energy.

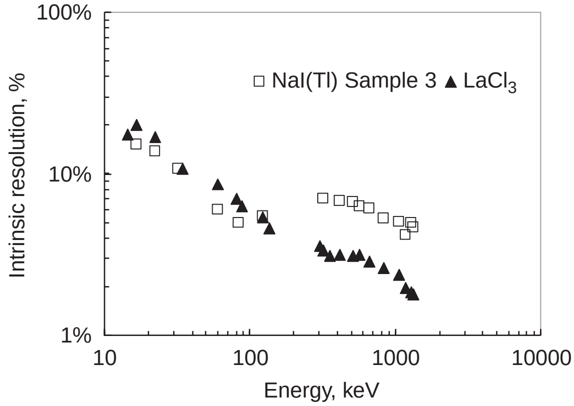

Fig. 7. Intrinsic energy resolution of $LaCl_3$:Ce and NaI(Tl) crystals.

The effect is more pronounced in Fig. 7 , which presents the intrinsic resolution of $LaCl_3$:Ce and NaI(Tl) evaluated by Eqs. (1) and (2). The intrinsic resolution at high energies is almost a factor of two better for $LaCl_3$:Ce, which seems to follow a better proportionality of the light yield, see Fig. 5.

At low energies, the contribution of the intrinsic resolution to the measured energy resolution in $LaCl_3$:Ce is larger than that in NaI(Tl). The point corresponding to a line at 33 keV is of special interest in this respect. It was determined from the Ba X-ray peak resulting from ^{138}La decay observed in the internal background of the $LaCl_3$:Ce crystal, see Section 3.2. The Ba X-rays are produced in the whole volume of the crystal. The line was separated to K_α and K_β components. It fits precisely to the distribution of other points in the low-energy range. This fact excludes a possibility of distortion of the intrinsic resolution in the low-energy region by surface effects in the crystal.

3.4. Photofraction

The ratio of counts under the photopeak (including the escape peak) to the total counts of the spectrum (photofraction) is shown for several energies in Table 1 for $LaCl_3$:Ce and NaI(Tl). For a comparison, the ratio of the cross-sections for the photoelectric effect to the total one including

Table 1
Photofraction for $LaCl_3$:Ce and NaI(Tl)

γ energy (keV) Source	320 ^{51}Cr	412 ^{198}Au	662 ^{137}Cs	835 ^{54}Mn	
Photof. (%)	38.7	27.8	17.9	17.8	$LaCl_3$:Ce
σ ratio (%)	33.8	22.3	9.8	6.7	
Photof. (%)	50.5	30.1	21.4	20.7	NaI(Tl)
σ ratio (%)	38.5	25.9	11.6	8.0	

coherent scattering calculated using XCOM program [9] are given too. The data suggest that both materials show a similar photofraction. However, for the 320 and 412 keV γ-peaks, the σ-ratio is closer to the measured photofraction for $LaCl_3$:Ce, then for NaI(Tl). It may be due to a slightly higher size of the NaI(Tl) crystal, but also due to lower δ-ray production in $LaCl_3$:Ce. Note that the σ-ratio is quite similar in both the crystals.

4. Summary and discussion

The advantages and properties for $LaCl_3$:Ce and NaI(Tl) are summarized in Table 2.

There is no doubt that the main advantage of $LaCl_3$:Ce over NaI(Tl) is its superior energy resolution at energies above 120 keV. This fact and a comparable detection efficiency and photofraction will make it, in the near future , a crystal of choice for precise γ-ray spectrometry. A much faster light pulse allows for high counting rate measurements, several times larger than with NaI(Tl). A high speed of the fast component of the light pulse and a high light output also assure fast timing capabilities of the new crystal.

The observed background of radioactive ^{138}La of about 1.07 counts per cm^3/s is not very serious. However, the observed contamination by U series isotopes should be removed in the further development of the crystal as it limits the application of the crystal in detection of radioactivity traces at high energies.

The very good proportionality of the light yield of $LaCl_3$:Ce, down to 20 keV, resulted in an excellent energy resolution above 120 keV. However, below 120 keV, energy resolution and especially intrinsic energy resolution is worse than that for NaI(Tl). The analysis of the energy resolution of 33 keV X-rays from a radioactive background of ^{138}La excluded a trivial explanation by a surface effect.

However, at low energies below 20 keV, a sharp proportionality drop is seen in Fig. 5. Thus, probably it is responsible for a large deterioration of the intrinsic resolution at low energies. Low-energy X-rays, Auger electrons and numerous secondary electrons (δ-rays) also contribute to create a full energy peak. Such deterioration, in fact, was qualitatively predicted in the simulation of the contribution arising from the γ-ray detection process to the intrinsic resolution based on the measured electron non-proportionality in LSO and NaI(Tl) [10]. In the calculations, δ-rays were not considered. Note that the YAP crystal, which exhibits a good proportionality down to 5.9 keV at 93% [11], show an intrinsic energy resolution better then NaI(Tl), even at 14 keV.Therefore, $LaCl_3$:Ce presents an intermediate case between YAP and LSO. All these crystals do not show any increase in the proportionality curve like NaI(Tl) does; the downward bending starts at about 10, 20 and 100 keV for YAP, $LaCl_3$:Ce and LSO, respectively. The intrinsic energy resolution also scales in the same order.

Table 2
Comparison of properties of $LaCl_3$:Ce and NaI(Tl)

Property	NaI(Tl)	$LaCl_3$:Ce
Light yield	12750 phe/MeV[a]	9400 phe/MeV
Afterglow	Large	Substantial
Radioactivity	None	^{138}La, 1.07 decays/cm^3
Energy resolution at 662 keV	$6.76 \pm 0.15\%$	$4.2 \pm 0.13\%$
Proportionality	+15% at 20 keV	$\pm 3\%$ above 20 keV
α/γ	0.64	0.33 ± 0.01
Density	3.67 g/cm^3	3.86 g/cm^3
Z_{eff}	50	59.5
Photofraction at 662 keV	21.4%	17.9%

[a]This number is unusually high. See end of Section 3.1.

A further improvement of crystal quality will benefit in a higher light output. At present, the tested sample showed about 40% lower light yield than small crystal samples [1–3].

Acknowledgments

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