Preliminary investigation of scintillator materials properties: $\text{SrI}_2\text{:Eu}$, CeBr_3 and GYGAG:Ce for gamma rays up to 9 MeV

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Abstract

In this work we measured the performance of a 2" x 2" cylindrical tapered crystal of $\text{SrI}_2\text{:Eu}$, a 2" x 3" cylindrical sample of CeBr_3 and a 2" x 0.3" cylindrical sample of GYGAG:Ce. These scintillators are prototypes in volume or material and were provided by the Lawrence Livermore National Laboratory and by the Institut de Physique Nucléaire d'Orsay. The gamma-ray energy resolution was measured in the energy range of 0.1 – 9 MeV using different sources. Each scintillator was scanned along x, y and z axes, using a 400 MBq collimated ^{137}Cs source. Owing to the GYGAG:Ce thickness, it was not possible to obtain the value of the energy resolution at 9 MeV and to scan the crystal along the z axis. The 662 keV full energy peak position and its FWHM were measured relative to the full energy peaks positions produced by a non-collimated ^{88}Y source. The signals of the detectors were additionally digitized and compared, up to 9 MeV, using a 12 bit LeCroy 600 MHz oscilloscope.

1. Introduction

In the last 10 years several new scintillator materials have been discovered. The lanthanum halides, $\text{LaBr}_3\text{:Ce}$ and $\text{LaCl}_3\text{:Ce}$, showed excellent performance and are now available in large volumes ($V > 1000 \text{ cm}^3$) [1-19]. Small samples (i.e. 1"x 1") of CeBr_3 and $\text{SrI}_2\text{:Eu}$ crystals appeared a few years later and there is still an intense R&D work on such detectors as the produced volume is constantly increasing over time [20-29]. The development of ceramic scintillator materials offers the possibility to have high-resolution gamma-ray spectroscopy at low cost. Transparent ceramic scintillators, such as GYGAG:Ce, allow gamma-ray spectroscopy performance superior to the most common scintillators [29-34].

For high energy gamma spectroscopy, large volume scintillators are required. We have obtained inch-scale samples of three new scintillators for evaluation of their properties, not yet well-established, for high energy gamma spectroscopy. These scintillator materials have very promising properties, as shown in table I. They could be good substitutes for NaI:Tl , the most used scintillator for gamma-ray detection and spectroscopy, as they all offer better energy resolution at 662 keV, higher light yield and higher density. The three studied scintillators could also represent a reasonable alternative for $\text{LaBr}_3\text{:Ce}$.

$\text{SrI}_2\text{:Eu}$ has the best energy resolution ($< 3\%$) among the studied detectors, and contains no internal radioactivity, but it is characterized by a slow signal and self-absorption [25-29]. The study of a large volume crystal is then important to understand how much the energy resolution is affected from this phenomenon. Furthermore, while the long decay time

constant of $\text{SrI}_2:\text{Eu}$ could be a critical aspect in case of high-rate experiments, it makes this material a good candidate for being used as a second stage crystal in a phoswich telescope.

CeBr_3 is characterized by an energy resolution that is a little worse than that of $\text{LaBr}_3:\text{Ce}$, but with the convenient advantage of having no internal radioactivity. This could be a good detector for low background experiments and for medical applications, such as PET (position emission tomography), in which a large number of detectors, providing good timing resolution, are involved.

GYGAG:Ce is a transparent ceramic oxide and therefore it is not affected by the problems of the crystal growth. It is not hygroscopic, nor does it contain internal radioactivity and it could be realized potentially in any dimension and shape.

Even though all three scintillator detectors studied in this work could be excellent substitutes of $\text{LaBr}_3:\text{Ce}$, at the moment, only few detailed studies on large volume $\text{SrI}_2:\text{Eu}$ and CeBr_3 or in general on GYGAG:Ce scintillators [20-34] are available in literature.

Material	Light Yield [ph/keV]	Emission [nm]	En. Res. [%]	ρ [g/cm ³]	Ref.
NaI:TL	38	415	6-7	3.7	[35]
CsI:TL	52	540	6-7	4.5	[35]
$\text{LaBr}_3:\text{Ce}$	63	360	3	5.1	[19]
$\text{SrI}_2:\text{Eu}$	80	480	3-4 (4)	4.6	[33]
GYGAG:Ce	40	540	<5 (5.2)	5.8	[29-34]
CeBr_3	45	370	3.5-4 (4.4)	5.2	[21]

Table I. The scintillation properties are compared. The scintillation light yield, the principal emission wavelength, the energy resolution at 662 keV and the density are listed in columns 2, 3, 4 and 5 respectively. The values of the energy resolution in bracket, reported in column 4 are the values measured in this work. As discussed in the text, they are a little bit worse than the expected ones.

In this work we present the study of the performances of a large diameter ceramic scintillator (2" x 0.3" GYGAG:Ce) and two large volume new generation scintillator crystals (a $\text{SrI}_2:\text{Eu}$ 2"x2" and a CeBr_3 2"x3"). We discuss the dependence of the peak centroid and its FWHM produced using a 1 mm collimated ^{137}Cs source as a function of the position of the interaction point in the detector. Furthermore we compare the anode pulse measured for different gamma-ray energies and for different positions of the collimated source.

This study is focused on the possible use of these crystals for high-energy gamma ray spectroscopy, for which large volume, self-absorption and homogeneity in the light yield are very crucial features.

In section 2 we describe the measurements performed in the gamma spectroscopy laboratory of the University of Milan. The results obtained for $\text{SrI}_2:\text{Eu}$ are discussed in section 3 while those of CeBr_3 and GYGAG:Ce in sections 4 and 5, respectively. For each crystal, we present the results of the energy resolution of the scintillator (subsection A), the detector response as a function of the interaction point along the three axes by using a collimated beam of 662 keV gamma rays from a ^{137}Cs source (subsection B) and the study of the signal shape up to 9 MeV gamma rays (subsection C). Comparison of the properties of these three detectors is given in section 6, with a conclusion in section 7.

2. The characterization measurements

This study offers a comparison of three new scintillators, at volumes relevant to high energy gamma spectroscopy applications. The 2" x 2" $\text{SrI}_2:\text{Eu}$ crystal is among the largest yet produced, the 2" x 3" CeBr_3 crystal is one of the biggest available on the market and while the 2" x 0.3" GYGAG:Ce ceramic scintillator is smaller, its performance surpasses other available oxide scintillators. The $\text{SrI}_2:\text{Eu}$ (3% doped) was grown by RMD while the GYGAG:Ce was produced by Lawrence Livermore National Laboratory (LLNL) which own both samples. The CeBr_3 was bought from

Scionix and it is from the Institut de Physique Nucleaire Orsay. The measurements of the detector performances were carried out in the gamma spectroscopy laboratory of the University of Milan. Table 2 lists the crystals, the PMTs, the voltage dividers (VD) and the voltage applied to the PMTs used for this set of measurements. In the case of CeBr₃ two different VDs were used: the first one is a standard VD from HAMAMASTU (E1198-27), while the second one, LABRVD, was developed by the electronic workgroup of the University of Milan and especially designed to work with the LaBr₃:Ce scintillators [19, 36]. We used this VD (as the CeBr₃ signal is very similar to the LaBr₃:Ce one) to reduce the PMT induced non linearity in energy. The SrI₂ has been coupled to a larger PMT as no 2" PMT were available. This should affect the crystal performances only marginally.

Crystal	PMT	VD	HV
SrI ₂	R6233-100SEL	E1198-27	800 V
GYGAG	R6233-100SEL	E1198-27	850 V
CeBr ₃	R6231-100MOD	E1198-27	600 V
CeBr ₃	R6231-100MOD	LABRVD	800 V

Table 2. Readout configurations used for each scintillator. The scintillators are listed in column 1, the PMTs, Voltage Dividers and the applied Voltages are listed in columns 2, 3, and 4, respectively.

The scintillation response was measured using standard gamma ray sources (²²Na, ⁶⁰Co, ⁸⁸Y, ¹³³Ba, ¹³⁷Cs, ¹⁵²Eu) and an AmBe(Ni) composite source for gamma rays up to 9 MeV [37]. In the latter, a core of ⁹Be and alpha-unstable ²⁴¹Am is surrounded by a thick layer of paraffin; some metal discs of nickel are also placed inside the paraffin layer. When an alpha particle is emitted by the ²⁴¹Am, there is a high probability that it is captured by ⁹Be, leading to ⁹Be(α ,n)¹²C reaction. The neutrons are thermalized by multiple scattering in the paraffin layer, which serves both as moderator and as shielding.

The signals of the three detectors were sent to a spectroscopic amplifier (TENNELEC TC244) and to an ADC (ORTEC ASPEC MCA 926). The detector anode pulses were also digitized using a 12 bit LeCroy HDO 6054 oscilloscope. A set of pulses (~1000) at a fixed energy were averaged to produce the reference signals. The signal properties (rise time and fall time) of all detectors were compared and the signals of each scintillator were studied from 662 keV up to 9 MeV. The rise time and the fall time are the convolution between the detector signal and the PMT intrinsic response.

The energy spectra and the pulses were measured using a collimated beam of 662 keV gamma rays scanning the detector along the x, y, and z axes.

Figure 1: The setup used to study the detector response as a function of the interaction point. The source is on a platform that could be moved both along the x and the y axes with a micrometer screw.

A non-collimated ^{88}Y source, providing two calibration points, was placed nearby and used as a reference. In this way it was possible to study how the position of the centroid, the FWHM, and the area of the peak at 662 keV changes as a function of the position of the incident radiation. The set-up, shown in Fig. 1, was composed of the detector under test, a collimated source of 400 MBq of ^{137}Cs and a platform which could be moved both along the x and the y axes by adjusting a micrometer screw. The collimator of heavy metal [38] was 8 cm long with a hole diameter of 1 mm, so that 96% of the γ rays were collimated within a 1 mm wide beam spot. The detector was placed on a second platform that is maintained at a fixed position. The source was placed in front of the detector by hands and the two platforms were aligned by eye. Particular care was taken to place the gamma ray beam perpendicular to the detector. The distance between the detector surface and the collimator was about 1.5 cm.

2.1 Test limitations

As previously discussed the signals of the three detectors were sent to a spectroscopic amplifier (TENNELEC TC244) and to an ADC (ORTEC ASPEC MCA 926) or, alternatively, the detector anode pulses were digitized using a 12 bit LeCroy HDO 6054 oscilloscope. The aim of this work was, in fact, to test these detector's performances using a standard electronics chain. The energy resolution measured using such approach could not be the 'best achievable' as will be discussed for the different detectors, therefore it is important to remember that i) The $\text{SrI}_2:\text{Eu}$ is not tested using a digital readout which should be able to overcome effects of self-absorption and re-emission (see discussion in section 3) ii) CeBr_3 is tested with an "issue" substantially worsening its energy resolution as it was the retreated by scionix (see discussion in sec. 4), iii) $\text{GYGAG}:\text{Ce}$ readout does not offer sufficient quantum efficiency and the crystal is too thin for an efficient high energy gamma-ray detection.

3. The $\text{SrI}_2:\text{Eu}$ scintillator

3.A The energy resolution

The energy resolution of the $\text{SrI}_2:\text{Eu}$ crystal was studied using standard gamma sources (^{152}Eu , ^{137}Cs , ^{60}Co , ^{88}Y) and an $\text{AmBe}(\text{Ni})$ composite source. The detector was placed on the paraffin over the AmBe source. The spectra using the other sources was measured placing them in front of the crystal but not necessary on the crystal axis. The used detector-source distance was chosen to have a count rate of approximately one kHz. The signal of the detector was sent to the amplifier (TENNELEC TC 244). The shaping time was set to 12 μs .

Fig. 2 shows the energy spectrum obtained by irradiating the scintillator with a ^{137}Cs and a ^{60}Co source. The measured energy resolution is $\sim 4.0\%$ at 662 keV (FWHM ~ 27 keV). The possibility to have different sources allows to measure the trend of the energy resolution. Fig. 3 shows the energy resolution trend as a function of the gamma-ray energy. The continuous line, in Fig. 3 represents the expected trend ($R \propto 1 / \sqrt{E}$). Since the 4.4 MeV peak FWHM has an intrinsic width, as explained in section 2, it could not be used for the energy resolution trend. The 9 MeV peak FWHM (see figure 4) was estimated to be $100 \text{ keV} \pm 20 \text{ keV}$ (1.1%) which is consistent with the expected trend.

The energy spectra of the crystal were obtained via two methods: in the first case we used the TENNELEC amplifier, while in the second one we obtained the spectra from the digitized signals. The measured energy resolution are comparable with the two methods.

The energy resolution obtained with the same crystal and other large volume $\text{SrI}_2:\text{Eu}$ crystals at Lawrence Livermore Laboratory, using instead the digital readout method, was better than 3% [31,33] and this is consistent with

what we have found using the collimated gamma rays source (see subsection B). With the digital readout, an on-the-fly correction factor is applied to the scintillation pulses as a function of their effective decay time, thus accounting for self-absorption and allowing an accurate energy histogram (gamma spectrum) to be obtained [31, 33].

Figure 2. The energy spectrum of the SrI₂:Eu scintillator, acquired with a standard spectroscopic chain, using 12 μ s of shaping time. The sources were ⁶⁰Co and ¹³⁷Cs. The energy resolution is about 4% at 662 keV. The acquisition time was about 10 minutes.

Figure 3. The measured energy resolution as a function of the energy of the incident radiation. The continuous line indicates the expected trend ($E \propto 1/\sqrt{E}$).

Figure 4. The energy spectrum of the $\text{SrI}_2:\text{Eu}$, acquired with a standard spectroscopic amplifier and ADC. The sources used were ^{88}Y and $\text{AmBe}(\text{Ni})$. The acquisition time was about 6 hours.

3.B Detector response as a function of the interaction point

The detector response as a function of the interaction position was studied along the crystal axes. We chose the x and y axes, on the crystal face. The z axes was the cylindrical symmetry axis, starting from the PMT to the crystal frontal face, as shown in Fig. 5.

Figure 5: The coordinate system used for the measurements of the detectors' response as a function of the interaction point.

The collimated source of ^{137}Cs was placed on a platform and moved with a micrometric screw. The measured energy spectra were calibrated using the ^{88}Y source. The position of the centroid with respect to the ^{88}Y peaks, the FWHM and the area of the 662 keV gamma peak were studied. The acquisition rate was 400 Hz.

The position of the centroid shows a variation of $\sim 3\%$, moving the source along the x and the y axes. Based on previous studies, we believe this to be due to optical "light-trapping" resulting from the Eu^{2+} self-absorption and re-emission [27]. Along lateral surfaces we also observe small variations of the centroid position (those are included in the $\sim 3\%$, mention above), FWHM, owing to absorption phenomena.

Fig. 6 and Fig. 7 show the full energy peak and its FWHM as a function of the interaction position along the z axis. The energy resolution changes from 22 keV (3.2 %) up to 34 keV (5.1%).

The result of the self-absorption effect is that a digital readout method, described by Cherepy and co-authors is needed to obtain the best possible energy resolution with large volume crystals [31,33]. The digital readout method was explained in subsection A.

Figure 6. The 662 keV peaks from a collimated ^{137}Cs source on the $\text{SrI}_2:\text{Eu}$ crystal. The different peaks were acquired by moving the collimated beam along the z axis of the crystal, where $z = 0$ is at the PMT window. The spectra were calibrated using a non-collimated ^{88}Y sources that was placed nearby as a reference.

Figure 7. The FWHM of 662 keV peaks from a collimated ^{137}Cs source on the $\text{SrI}_2:\text{Eu}$ crystal. The different values were obtained by moving the collimated beam along the z axis of the crystal. The origin of the z axis is the PMT side, as shown in Fig. 5. The spectra were calibrated using a non-collimated ^{88}Y sources that was placed nearby as a reference.

3.C Signal shape

The signal shape of the $\text{SrI}_2:\text{Eu}$ was studied from 662 keV up to 9 MeV. The signals of the detector were digitized with a LeCroy waverunner oscilloscope (HDO 6054). The sampling frequency was 0.5 Gsamples/s and the sampling range was 20 μs .

A set of pulses (~ 1000) were average to produce the pulses of Fig. 8. At 9 MeV, the signals, whose energy is in a range of 1 MeV (from 8 MeV up to 9 MeV), were used to produce the average signal (see legend of Fig. 8). The rise time (10% - 90% of amplitude) was calculated for all the averaged signals and it was found equal to 24 ± 2 ns. No rise time variations were observed as a function of the energy of the incident gamma ray. The fall time (90% - 10% of amplitude) of the average signals was measured to be 7 μs . Fig. 8 shows the average signals of the $\text{SrI}_2:\text{Eu}$. The plot

shows that at different deposited energies no significant changes in pulse shape is observed, therefore only one curve is practically observable in figure 8.

Figure 8. The area normalized average pulses of the $\text{SrI}_2\text{:Eu}$ excited gamma rays from 662 keV up to 9 MeV. No significant changes in shape are present. The areas of the pulses are normalized to one. The different curves overlaps and therefore only one line is visible.

The signal shape was also studied as a function of the gamma-ray interaction point along the z axis ($z = 0.5$ cm is the measured point closest to the PMT). Four different positions along the z axis were selected, as shown in Fig. 9. The signals are different as a function of the interaction point, as it is emphasized in the inset of Fig. 9. The rise time remains unchanged while the fall time is longer (it changes from $6.7 \mu\text{s}$ to $7.3 \mu\text{s}$) when gamma rays enter near the PMT. The area of the pulses is normalized to one to underline the different shapes of Fig. 9.

Figure 9. The area normalized pulses of the $\text{SrI}_2\text{:Eu}$ for the 662 keV gamma ray from a collimated ^{137}Cs source. The area of the pulses is normalized to one.

4. The CeBr_3 scintillator

4.A The energy resolution

The signal of the CeBr₃ detector was sent to a spectroscopy amplifier (TC 244) and the shaping time was set to 1 μ s.

Fig. 10 shows the energy spectra acquired when irradiating the scintillator with ⁸⁸Y, ⁶⁰Co and AmBe(Ni) radioactive sources. The measured energy resolution is $4.4 \pm 0.1\%$ at 662 keV (FWHM ~ 29 keV). As for the SrI₂ the measured energy resolution follows the expected trend ($R \propto 1 / \sqrt{E}$). The measured FWHM for the 9 MeV gamma-rays is 120 ± 5 keV which is comparable with the expected value of 108 keV. The 9 MeV peak, in bottom panel of Fig. 6, is located at 8.6 MeV due to the non-linearity of the PMT tube ($\sim 4\%$ at 9 MeV). To reduce the non-linearity effect we coupled the PMT to an active voltage divider especially designed for the LaBr₃:Ce detectors, called LABRVD [19,36]. The energy resolution was unchanged but the measured non linearity reduced to 1%.

The measured energy resolution for this detector is bigger than the nominal value (3.6% at 662 keV) due to an issue in the crystal gluing process repaired after these tests. At present, after a repair procedure performed by the crystal manufacturer the detection properties of the scintillators are completely recovered. The measured value of the energy resolution does not affect the detector response as a function of the interaction point (subsection B) and the signal properties (subsection C).

Figure 10. The energy spectrum of the CeBr₃, acquired with a standard spectroscopic amplifier and ADC. The sources used were ⁶⁰Co and ⁸⁸Y and AmBe(Ni). Top panel: zoom of the low-energy part of the spectrum. Bottom panel: the whole energy spectrum. The acquisition time was about 16 hours.

4.B Detector response as a function of the interaction point

The detector response as a function of the interaction point was studied along the crystal axes, as done for the SrI₂:Eu (section 3, subsection B). In this case, the acquisition rate was 1000 Hz, due to the acquisition threshold that was very low.

The position of the centroid as well as the peak FWHM and area, does not show variations while moving the source along the x and the y axes. Fig. 11 shows the peak at 662 keV for different interaction positions along the z axis and in

this case a variation of the centroid position and width is visible. The energy resolution remains however constant around 28-29 keV at 662 keV (4.4%), as shown in Fig. 12.

Figure 11. The 662 keV peaks from a collimated ^{137}Cs source on the CeBr_3 crystal. The different peaks were acquired by moving the collimated beam along the z axis of the crystal. The spectra were calibrated using a non-collimated ^{88}Y sources that was placed nearby as a reference.

Figure 12. The FWHM of 662 keV peaks from a collimated ^{137}Cs source on the CeBr_3 crystal. The different values were obtained by moving the collimated beam along the z axis of the crystal. The origin of the z axis is the PMT side, as shown in Fig. 5.

4.C Signal shape

The anode signal line-shape was studied from 662 keV up to 9 MeV. The only difference with respect to the $\text{SrI}_2:\text{Eu}$ measurement was in the sampling frequency of the signal (2.5 Gsamples/s), owing to the fast CeBr_3 signal. A constant rise time of 18 ns (10% - 90% of amplitude) was extracted for all signals. The fall time of the signals is about 70 ns.

Fig. 13 shows the averaged signals for different energies. The small observed changes in the signal line-shape could be explained by a non-linearity effect in the PMT, owing to the fast signal and high light yield of CeBr_3 . A similar non-linearity effect was already observed for $\text{LaBr}_3:\text{Ce}$ [19].

Figure 13. The area normalized average pulses of the CeBr_3 for gamma rays from 662 keV up to 9 MeV. Changes in shape are present, as shown in the inset. These changes are related on the PMT non-ideal behavior owing to the high light yield and the fast signal (rise time ~ 20 ns and fall time ~ 70 ns) of CeBr_3 .

5. The GYGAG:Ce scintillator

5.A. The energy resolution

The GYGAG:Ce ceramic scintillator, differently from the previous two materials, is neither a single crystal nor hygroscopic. Therefore it was directly coupled to the PMT (HAMAMATSU R6233-100sel). The scintillator was covered by Teflon to optimize the light collection and the energy resolution.

The energy resolution of the GYGAG:Ce scintillator was studied using standard gamma-emitting sources (^{152}Eu , ^{137}Cs , ^{60}Co , ^{88}Y), as for the other two crystals. The ceramic was coupled to a HAMAMATSU R6233-100sel PMT. The shaping time of the amplifier was set to 6 μs . As for the previous two detectors the measured energy resolution values follow the expected trend ($R \propto 1 / \sqrt{E}$).

Fig. 14 shows the energy spectrum acquired irradiating the scintillator with a ^{137}Cs and a ^{60}Co source at the same time. The measured energy resolution is $5.2 \pm 0.1\%$ at 662 keV (FWHM ~ 34 keV). The used PMT is equipped with a photocathode optimized for the blue region of visible spectrum while for the GYGAG:Ce, typical emission wave-length is in the yellow (530 nm). Thus, the quantum efficiency of the used PMT, which is about 35% at 350 nm, is considerably reduced (around 12%) at 530 nm. If using a device with a quantum efficiency of $\sim 35\%$ in the yellow region, we expect an energy resolution of $\sim 3.2\%$ at 662 keV. Furthermore the energy resolution of the GYGAG:Ce could be also affected by the afterglow phenomenon. To limit and eventually eliminate this effect, it is necessary to store GYGAG:Ce (after handling in room lights) in the dark for several days before using, to obtain the best possible energy resolution. This same scintillator, measured in [34] offered 4.9% resolution at 662 keV. The measurements reported here were performed 48 hours after coupling the ceramic with the PMT and wrapping it with black tape for light tightness. The value of the energy resolution does not significantly affect the detector response as a function of the interaction point (subsection B) and the signal properties (subsection C). The energy resolution of the smaller ceramic samples was $< 4\%$ with a silicon photodiode, in optimized condition at LLNL [34].

High-energy gamma rays from the AmBe(Ni) source were measured also using the GYGAG:Ce detector. However, the peak at 9 MeV is just barely visible, as shown in Fig. 18, due to the low statistics accumulated due to the reduced thickness of the detector.

Figure 14. The energy spectrum of the GYGAG:Ce, acquired by standard spectroscopic amplifier and an ADC. The sources used were ^{60}Co and ^{137}Cs . The acquisition time was about 1 hour.

5.B. Detector response as a function of the interaction point

The detector response as a function of the interaction point was studied along the crystal axes, using for the GYGAG:Ce the same experimental procedure, already described for the other scintillators. Owing to the crystal thickness (0.3"), the response was studied only along the x and y axes. In this case, the acquisition rate was 800 Hz.

The position of the centroid does not show variations. Furthermore the FWHM and area are constant (the variation is smaller than 10%), as well. Fig. 15 shows the trend of the FWHM as a function of the interaction position along the x axis. The FWHM value is almost constant about 33 keV at 662 keV (< 5%).

Figure 15. The FWHM of 662 keV peaks from a collimated ^{137}Cs source. The different values were obtained by moving the collimated beam along the x axis of the crystal. The spectra were calibrated using a non-collimated ^{88}Y sources that was placed nearby as a reference.

5.C. Signal line-shape

In this section, the signal shape of the GYGAG:Ce was studied from 662 keV up to 9 MeV. The sampling frequency was 1.25 Gsamples/s. As at 4 MeV and at 9 MeV the statistic was not high, a range of 1 MeV (from 3.5 MeV up to 4.5 MeV and from 8 MeV up to 9 MeV, respectively) was used to select the signals.

The signal of the GYGAG:Ce has a rise time (10% - 90% of amplitude) of 26 ns. The PMT (HAMAMATSU R6233-100sel) intrinsic rise time that is 9.5 ns at 1000V, could affect this value. The fall time of the average signals is about 700 ns.

Fig. 16 shows the average signals for the different investigated gamma-ray energies. No significant changes in shape were observed neither changing gamma-ray energy nor the incident position of the 662 keV collimated source.

Figure 16. The area normalized average pulses of the GYGAG:Ce for the 662 keV gamma ray from a collimated ^{137}Cs source. No significant changes in shape are present.

6. Detector Comparison

The detectors that have been studied in this work, $\text{SrI}_2\text{:Eu}$, $\text{CeBr}_3\text{:Ce}$ and GYGAG:Ce scintillators show energy resolution of 4.0%, 4.4% and 5.2% at 662 keV, respectively. All the values of the energy resolution are better than that of NaI:Tl (6 % at 662 keV), the most widely used scintillator for gamma spectroscopy. The spectra measured using an ^{152}Eu source are compared in Fig. 17.

Figure 17. The ^{152}Eu energy spectra acquired using a standard spectroscopic amplifier for the three tested scintillators. The $\text{SrI}_2:\text{Eu}$ has the best energy resolution among these scintillators. The spectra are normalized on the area of the 344 keV peak of the ^{152}Eu source.

The energy spectra acquired irradiating the scintillators under test with an AmBe(Ni) source, which provides gamma rays up to 9 MeV, are compared in Fig. 18. The CeBr_3 spectrum shows clearly the 9 MeV peak and the corresponding first and second escape peaks. CeBr_3 has the largest volume (2" x 3") and has a density of 5.2 g/cm³. Because of the reduced statistics, the 9 MeV peak, its first escape and its second escape peaks are observed with SrI_2 , though not as clearly as for CeBr_3 . The thickness of the GYGAG:Ce is 0.3" and therefore the 9 MeV peak is barely visible. It is important to point out that the spectra of Fig. 18 are normalized to the area of the 898 keV peak of the ^{88}Y source.

Figure 18. The measured $\text{SrI}_2:\text{Eu}$, GYGAG:Ce and CeBr_3 acquired using a standard spectroscopic amplifier and an ADC. The used sources were AmBe(Ni) and ^{88}Y . The CeBr_3 spectrum shows also the two ^{60}Co peaks and it is the detector with highest statistics at 9 MeV owing to its large volume and to a long the acquisition time (6, 4, and 16 hours for the $\text{SrI}_2:\text{Eu}$, the CeBr_3 and the GYGAG:Ce, respectively). The spectra are normalized to the area of the 898 keV peak of the ^{88}Y source.

The signals of the three detectors for energies ranging between 662 keV up to 9 MeV were digitized and compared. Fig. 19 shows the comparison of the three signal shapes, at 662 keV, for the three scintillators. The CeBr_3 provides the fastest signal: it has a rise time of ~18 ns and a fall time of ~70 ns. Otherwise $\text{SrI}_2:\text{Eu}$ has the slowest signal among the tested detectors (fall time ~7 μs).

Figure 19. The area normalized pulses of the three detectors for 662 keV gamma rays. The CeBr_3 has the fastest signal among these scintillators (similar to that of $\text{LaBr}_3\text{:Ce}$), whereas $\text{SrI}_2\text{:Eu}$ has the slowest signal.

The detector response as a function of the interaction point is almost constant for the CeBr_3 and the GYGAG:Ce , in which no significant variation of the peak position or the FWHM are observed. While $\text{SrI}_2\text{:Eu}$ detector shows variations in the FWHM along the z axis imputable to the self-absorption effect.

$\text{SrI}_2\text{:Eu}$ has the best energy resolution among the studied detectors (4.0 % at 662keV), but this value depends on the position of the gamma-ray interaction point. Self-absorption affects the energy resolution of this detector. A FWHM value as good as 22 keV at 662 keV (3.3%), measured with the collimated source. This crystal has a slow decay time constant and so for this reason it would not be suited to high-rate experiments, nevertheless this scintillator can find a possible application as a second stage crystal in a phoswich detector, or for low count rate applications such as environmental monitoring, as it also has no intrinsic radioactivity.

CeBr_3 has an energy resolution (the measured value is 4.4% at 662 keV) that is a little worse than that of $\text{LaBr}_3\text{:Ce}$, but it is still considerably better than NaI:Tl . The absence of internal activity make this crystal ideal for low background measurements or in the cases in which an array with several detectors is used. The detector is characterized by a fast signal, similar to $\text{LaBr}_3\text{:Ce}$, therefore it could be used successfully in the case of high counting rate. The energy resolution is expected to be better (about 3%) at 662 keV in the case of co-doped crystal [20] but, at present co-doped CeBr_3 is still not commercially available. We observed non linearity effects at 9 MeV, as is also typically observed with $\text{LaBr}_3\text{:Ce}$ scintillators. No significant variation of the peak centroid the FWHM or of the area were observed as a function of the interaction point.

GYGAG:Ce provides high efficiency and good energy resolution of 5% at 662 keV with a PMT equipped with a non-optimal photocathode (blue sensitive, rather than green/yellow sensitive to match the 500-650 nm emission). No variations of the peak centroid, the FWHM or the area are observed as a function of the interaction point, along the x and the y axes.

7. Conclusion

In this work relatively large volume $\text{SrI}_2\text{:Eu}$, $\text{CeBr}_3\text{:Ce}$ and GYGAG:Ce scintillators were characterized by studying the pulse shape, the energy resolution their response to a collimated source and to 9 MeV γ -rays. All three scintillators offer improved energy resolution, compared to standard scintillators, such as NaI:Tl .

The detector sizes (6 in³ $\text{SrI}_2\text{:Eu}$, 9 in³ CeBr_3 and 1 in³ GYGAG:Ce scintillators) are large enough in volume to measure medium/high energy gamma rays and assess the homogeneity of their response. $\text{SrI}_2\text{:Eu}$, shows the best energy

resolution, however self-absorption impairs the performance when standard analog readout is used (as reported here). The $\text{SrI}_2\text{:Eu}$ is an excellent candidate for applications in which small volumes are used. The energy resolution for large volume $\text{SrI}_2\text{:Eu}$ can match that of small crystals (<3% at 662 keV) only if digital readout methods (capable to nullify self absorption effects) are applied, though the resolution obtained here with analog readout, R(662 keV), 4% is still better than NaI:Tl. It is free of intrinsic radioactivity and its relatively slow decay time is still fast enough to applications to measure low count rate sources.

CeBr_3 is 100% Ce doped $\text{LaBr}_3\text{:Ce}$, thus it is without internal radiation, but suffers from slightly worse energy resolution. Its fast decay time is useful in high count rate applications. It can be used to measure high energy γ -rays with good energy resolution and we have measured 1.1% at 9 MeV. As it is not doped very large volume crystals are not found to suffer from inhomogeneity.

The GYGAG:Ce seems to be an excellent detector without intrinsic radioactivity, while providing high efficiency and good energy resolution. Furthermore, the ceramic detectors offer the attractive possibility to be produced in any shape or dimension, and to be more stable to mechanical shock and to not degrade in humid conditions than halide single crystals. However GYGAG:Ce ceramics, are not yet commercially available.

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References

- [1] BrillanCescintillatorsperformancesummary.pdf. (<http://www.detectors.saint-go-bain.com/Brilliance380.aspx>).
- [2] E. V. D. Van Loef, P. Dorenbos, C. W. E. van Eijk K. Kramer and H. U. Gudel, “High-energy-resolution scintillator: Ce 3+ activated LaBr 3”, Appl. Phys. Lett., vol. 79, n. 10 pp. 1573 – 1575, Sept. 2001. .
- [3] E. V. D. Van Loef, P. Dorenbos, C. W. E. van Eijk K. Kramer and H. U. Gudel, “Scintillation properties of LaBr3:Ce3+ crystals: fast, efficient and high-energy-resolution scintillators”, Nucl. Instr. Meth A, vol. 486, n. 1-2, pp 254-258, Jun. 2002.
- [4] W. W. Moses, “Current trends in scintillator detectors and materials”, Nucl. Instr. Meth. A, vol. 487, n. 1-2, pp 123-128, Jul. 2002.
- [5] P. Dorenbos, J. T. M. de Hass and C. W. E. Eijk, “Gamma ray spectroscopy with a O19×19 mm3 LaBr3:0.5% Ce3+ scintillator”, IEEE Trans. on Nucl. Science, vol. 51, n. 3, pp. 1289 – 1296, Jul. 2004.
- [6] B. D. Milbrath, J. I. McIntyre, R. C. Runkle, and L. E. Smith, “Contamination Studies of LaCl3:Ce Scintillators”, IEEE Trans. on Nucl. Science, vol. 53, n. 5, pp. 3031 - 3034, Oct. 2006.
- [7] A. Iltis, M. R. Mayhugh, P. Menge, C. M. Rozsa, O. Selles and V. Solovyev, “Lanthanum halide scintillators: Properties and applications”, Nucl. Instr. Meth. A, vol. 563, n. 2, pp. 359 - 363, Jul. 2006.
- [8] M. Moszynski, M. Gierlik, M. Kapusta, A. Nassalski, T. Szczęśniak, Ch. Fontaine and P. Lavoute, “New Photonis XP20D0 photomultiplier for fast timing in nuclear medicine”, Nucl. Instr. Meth. A, vol. 567, n. 1, pp. 31–35, Nov. 2006.
- [9] M. Moszynski, A. Nassalski, A. Syntfeld-Kazuch, T. Szczęśniak, W. Czarnacki, D. Wolski, G. Pausch and J. Stein, “Temperature dependences of LaBr3(Ce), LaCl3(Ce) and NaI(Tl) scintillators”, Nucl. Instr. Meth. A, vol. 568, n. 2, pp. 739–751, Dec. 2006.
- [10] S. Normand, A. Iltis, F. Bernard, T. Domenech and P. Delacour, “Resistance to γ irradiation of LaBr3:Ce and LaCl3:Ce single crystals”, Nucl. Instr. Meth. A, vol. 572, n. 2, pp. 754–759, Mar. 2007.
- [11] A. Owens, A. J. J. Bos, S. Brandenburg, E.-J. Buis, C. Dathy, P. Dorenbos, C. W. E. van Eijk, S. Kraft, R. W. Ostendorf, V. Ouspenski, and F. Quarati, “Assessment of the radiation tolerance of LaBr3:Ce scintillators to solar proton events”, Nucl. Instr. Meth. A, vol. 572, n. 2, pp. 785–793, Mar. 2007.

- [12] F. Quarati, A. J. J. Bos, S. Brandenburg, C. Dathy, P. Dorenbos, S. Kraft, R. W. Ostendorf, V. Ouspenski and A. Owens, "X-ray and gamma-ray response of a 2"×2" LaBr₃:Ce scintillation detector", Nucl. Instr. Meth. A, vol. 574, n. 1, pp. 115–120, Apr. 2007.
- [13] P. Menge, G. Gautier, A. Iltis, C. Rozsa and V. Solov'yev, "Performance of large lanthanum bromide scintillators", Nucl. Instr. Meth. A, vol. 579, n. 1, pp. 6–10, Aug. 2007.
- [14] R. Nicolini, F. Camera, N. Blasi, S. Brambilla, R. Bassini, C. Boiano, A. Bracco, F.C.L. Crespi, O. Wieland, G. Benzoni, S. Leoni, B. Million, D. Montanari and A. Zalite, "Investigation of the properties of a 1"×1" LaBr₃:Ce scintillator", Nucl. Instr. Meth. A, vol. 582, n. 2, pp. 554–561, Nov. 2007.
- [15] F. C. L. Crespi, F. Camera, N. Blasi, A. Bracco, S. Brambilla, B. Million, R. Nicolini, L. Pellegri, S. Riboldi, M. Sassi, O. Wieland, F. Quarati and A. Owens, "Alpha–gamma discrimination by pulse shape in LaBr₃:Ce and LaCl₃:Ce", Nucl. Instr. Meth. A, vol. 602, n. 2, pp. 520–524, Apr. 2009.
- [16] M. Ciemala, D. Balabanski, M. Csatlós, J.M. Daugas, G. Georgiev, J. Gulyás, M. Kmiecik, A. Krasznahorkay, S. Lalkovski, A. Lefebvre-Schuhl, R. Lozeva, A. Maj and A. Vitez, "Measurements of high-energy γ -rays with LaBr₃:Ce detectors", Nucl. Instr. Meth. A, vol. 608, n. 1, pp. 76–79, Sep. 2009.
- [17] M. Nocente, M. Tardocchi, I. Chugunov, R. C. Pereira, T. Edlington, A. M. Fernandes, D. Gin, G. Grosso, V. Kiptily, A. Murari, A. Neto, E. Perelli Cippo, A. Pietropaolo, I. Proverbio, A. Shevelev, J. Sousa, B. Syme, G. Gorini and JET-EFDA Contributors, "Energy resolution of gamma-ray spectroscopy of JET plasmas with a LaBr₃ scintillator detector and digital data acquisition", Rev. Sci. Instrum., vol. 81, n. 10, pp. 10D321, Oct. 2010.
- [18] M. Nocente, M. Tardocchi, A. Olariu, S. Olariu, R. C. Pereira, I. N. Chugunov, A. Fernandes, D. B. Gin, G. Grosso, V. G. Kiptily, A. Neto, A. E. Shevelev, M. Silva, J. Sousa, G. Gorini, "High Resolution Gamma Ray Spectroscopy at MHz Counting Rates With LaBr₃ Scintillators for Fusion Plasma Applications", IEEE Trans. on Nucl. Science, vol. 60, n. 2, pp. 1408 - 1415, Apr. 2013.
- [19] A. Giaz, L. Pellegri, S. Riboldi, F. Camera, N. Blasi, C. Boiano, A. Bracco, S. Brambilla, S. Ceruti, S. Coelli, F.C.L. Crespi, M. Csatlós, S. Frega, J. Gulyás, A. Krasznahorkay, S. Lodetti, B. Million, A. Owens, F. Quarati, L. Stuhl and O. Wieland, "Characterization of large volume 3.5"×8" LaBr₃:Ce detectors", Nucl. Instr. Meth. A, vol. 729, n. 1, pp. 910–921, Nov. 2013.
- [20] F. Quarati, M. S. Alekhin, K. W. Krämer and P. Dorenbos, "Co-doping of CeBr₃ scintillator detectors for energy resolution enhancement", Nucl. Instr. Meth. A, vol. 735, n. 1, pp. 655–658, Jan. 2014.
- [21] F. Quarati, P. Dorenbos, J. van der Biezen, A. Owens, M. Selle, L. Parthier and P. Schotanus, "Scintillation and detection characteristics of high-sensitivity CeBr₃ gamma-ray spectrometers", Nucl. Instr. Meth. A, vol. 729, n. 1, pp. 596–604, Nov. 2013.
- [22] P. Guss, M. Reed, D. Yuan, A. Reed and S. Mukhopadhyay, "CeBr₃ as a room-temperature, high-resolution gamma-ray detector", Nucl. Instr. Meth. A, vol. 608, n. 2, pp. 297–304, Sep. 2009.
- [23] W. Drozdowski, P. Dorenbos, A. J. J. Bos, G. Bizarri, A. Owens and F. G. A. Quarati, "CeBr₃ Scintillator Development for Possible Use in Space Missions", IEEE Trans. on Nucl. Science, vol. 55, n. 3, pp. 1391 - 1396, Jun. 2008.
- [24] K. Shah, J. Glodo, W. Higgins, E. van Loef, W. W. Moses, S. E. Derenzo, and M. J. Weber, "CeBr₃ scintillators for gamma-ray spectroscopy", IEEE Trans. on Nucl. Science, vol. 52, n. 6, pp. 1391 - 1396, Feb. 2005.
- [25] N. Cherepy, S. A. Payne, B. W. Sturm, O. B. Drury, S. P. O'Neal, P. A. Thelin, K. S. Shah, R. Hawrami, M. Momayezi, B. Hurst, A. Burger, B. Wiggins, P. Bhattacharya, L. A. Boatner and J. O. Ramey, "Instrument Development and Gamma Spectroscopy With Strontium Iodide", IEEE Trans. on Nucl. Science, vol. 60, n. 2, pp. 955 - 958, Feb. 2013.
- [26] P. Belli, R. Bernabei, R. Cerulli, F. A. Danevich, E. Galenin, A. Gektin, A. Incicchitti, V. Isaienko, V. V. Kobaychev, M. Laubenstein, S. S. Nagorny, R. B. Podviyanuk, S. Tkachenko and V. I. Tretyak, "Radioactive contamination of SrI₂(Eu) crystal scintillator", Nucl. Instr. Meth. A, vol. 670, n. 1, pp. 10–17, Apr. 2012.
- [27] B. W. Sturm, N. J. Cherepy, O. B. Drury, P. A. Thelin, S. E. Fisher, S. A. Payne, A. Burger, L. A. Boatner, J. O. Ramey, K. S. Shah and R. Hawrami, "Effects of packaging SrI₂(Eu) scintillator crystals", Nucl. Instr. Meth. A, vol. 652, n. 1, pp. 242–246, Apr. 2011.
- [28] J. Glodo, E. V. Van Loef, N. J. Cherepy, S. A. Payne and K. S. Shah, "Concentration Effects in Eu Doped SrI₂", IEEE Trans. on Nucl. Science, vol. 57, n. 3, pp. 1228 - 1232, Jun. 2010.
- [29] N. Cherepy, S. A. Payne, B. W. Sturm, J. D. Kuntz, Z. M. Seeley, B. L. Rupert, R. D. Sanner, O. B. Drury, T. A. Hurst, S. E. Fisher, M. Groza, L. Matei, A. Burger, R. Hawrami, K. S. Shah and L. A. Boatner, "Comparative gamma spectroscopy with SrI₂(Eu), GYGAG(Ce) and Bi-loaded plastic scintillators", Nucl. Science Symp. Conf. Record (NSS/MIC), Knoxville, TN, 2010, pp. 1288 – 1291.
- [30] Z. Seeley, N. J. Cherepy and S. A. Payne, "Homogeneity of Gd-based garnet transparent ceramic scintillators for gamma spectroscopy", Jour. of Crys. Growth, vol. 379, n. 1, pp. 79–83, Sep. 2013.
- [31] N. Cherepy, Z. M. Seeley, S. A. Payne, P. R. Beck, O. B. Drury, S. P. O'Neal, K. M. Figueroa, S. Hunter, L. Ahle, P. A. Thelin, T. Stefanik and J. Kindem, "Development of transparent ceramic Ce-doped gadolinium garnet gamma spectrometers", Nucl. Science Symp. Conf. Record (NSS/MIC), Anaheim, CA, 2012, pp. 1692 - 1697.
- [32] L. A. Boatner, J.O. Ramey, J. A. Kolopus, R. Hawrami, W. M. Higgins, E. van Loef, J. Glodo, K. S. Shah, E. Rowe, P. Bhattacharya, E. Tupitsyn, M. Groza, A. Burger, N. J. Cherepy and S. A. Payne, "Bridgman growth of large SrI₂:Eu²⁺ single crystals: A high-performance scintillator for radiation detection applications", Jour. of Crys. Growth, vol. 379, n. 1, pp. 63–68, Sep. 2013.
- [33] P. R. Beck, N. J. Cherepy, S. A. Payne, E. L. Swanberg, K. E. Nelson, P. A. Thelin, S. E. Fisher, S. Hunter, B. M. Wihl, K. S. Shah, R. Hawrami, A. Burger, L. A. Boatner, M. Momayezi, K. T. Stevens, M. H. Randles and D. Solodovnikov, "Strontium iodide instrument development for gamma spectroscopy and radioisotope identification", SPIE Optical Engineering Applications, San Diego, CA, USA, 2014, pp. 92130N.
- [34] N. J. Cherepy, Z. M. Seeley, S. A. Payne, P. R. Beck, S. Hunter, L. Ahle, S. E. Fisher, C. Melcher, H. Wei, T. Stefanik, Y.-S. Chung and J. Kindem, "High energy resolution with transparent ceramic garnet scintillators", SPIE Optical Engineering Applications, San Diego, CA, USA, 2014, pp. 921302.

- 1 [35] http://www.crystals.saint-gobain.com/Scintillation_Materials.aspx
- 2 [36] S. Riboldi, F. Camera, N. Blasi, S. Brambilla, C. Boiano, F. C. L. Crespi, A. Giaz, B. Million, R. Nicolini, L. Pellegrini and O. Wieland, “Active
- 3 voltage divider for improved estimation of interacting radiation energy with photomultiplier tubes coupled to high light yield scintillators”, Nucl.
- 4 Science Symp. Conf. Record (NSS/MIC), Valencia, Spain, 2011, pp 776 - 778.
- 5 [37] J.G. Rogers, M. S Andreaco, C. Moisan, I. M Thorson, “A 7–9 MeV isotopic gamma-ray source for detector testing”, Nucl. Instr. Meth. A, vol.
- 6 413, n. 2-3, pp. 249–254, Aug. 1998.
- 7 [38] <http://www.wolfmet.com/documents>.