Characterization of the Lanthanum Chloride Scintillation Detector

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Abstract. This paper reports about the investigations on the performance of the new earth-rare halide Scintillator, the LaCl₃:10%Ce, which has been discovered with attractive scintillation properties: fast, efficient, and high-energy resolution. The combination of good brightness and linear response has naturally resulted in a very good energy resolution, whereas the fast decay time promoted counting applications at very high rate and very fast timing resolution. Energy resolutions of (2.99±0.02) %, (8.66±0.02) %, and (2.05±0.01) % have been achieved by exciting the detector with 662 keV ¹³⁷Cs, 122 keV ¹⁵²Eu, and 1332.5 keV ⁶⁰Co sources respectively, at room temperature. The variations of the resolution as a function of both shaping time and high voltage have been studied and found to be relatively linear. The timing resolution was also studied. The value of 1.07 ns FWHM has been recorded with LaCl₃:10%Ce and BaF₂ detectors operating in coincidence mode, using 1333.5 keV γ-ray ⁶⁰Co peak. Compared to similar studies, this timing resolution was worse than expected even if the exciting radioactive sources used in the two experiments were different. The efficiency values of (0.24±0.01)%, (0.01±0.01)% and (0.09±0.01)% were obtained with laboratory ¹³⁷Cs, ¹⁵²Eu, ⁶⁰Co isotopes respectively. These detector efficiency values were very low, due to the smaller detector sizes used in these earlier crystals and to the large detector-source-distance used in this experiment. The Lanthanum Chloride Scintillator detector is hygroscopic and relatively expensive compared to established scintillators. In addition, it carries internal contamination in ¹³⁸La and ²²⁷Ac for larger thicknesses. Nevertheless, the energy resolution figures achieved, twice as good as that of NaI(Tl), are in good agreement with those reported elsewhere. They credit the scintillation detectors as good challengers of semiconductor-based detectors, allowing their application fields to be extended.

KEYWORDS: Scintillator, Energy Resolution, LaCl₃:Ce, Gamma ray spectrometry

1. Introduction

It was in November 1895 that W.C. Roentgen discovered the X-ray [1]. The use of this new and penetrating radiation found more applications with the development of the luminescent materials, which efficiently absorb X-rays (Phosphors). The discovery of the scintillator material was undertaken in three successive main phases [2]. The first and the second phases included experiences with the earliest scintillators CaWO₄ used by Becquerel, Crookes and Rutherford; discovery of scintillation in naphthalene and development, by Hofstadter, of the thallium-activated NaI. The third phase, corresponding to the two past decades, witnessed the renaissance in research and development of scintillator materials.

This renewed interest to scintillator materials was motivated by two major applications in nuclear physics and medical imaging: the construction of the electromagnetic calorimeter of the CMS at Large Hadron Collider (LHC) and the increasing demand in Positron Emission Tomography (PET). Scintillation detectors are also widely used in the detection and spectroscopy of ionizing radiations. Common applications of scintillation spectrometers include non-destructive testing, nuclear non-proliferation, environmental monitoring, and X-ray diffraction. In these different applications, detector requirements vary considerably accordingly. In γ-ray counting and spectroscopy for example, the light yield, scintillation decay, and energy resolution are considered to be critical properties [3].

Today, Research and Development to introduce new scintillator materials has focused on better understanding of and improving the properties of two particular families of scintillator materials: (1) lanthanum halides doped-cerium [4] (LnX₃:Ce³⁺, with Ln = La, Gd, Lu and X = F, Cl, Br, I) and (2) PbWO₄ [5] or Lu₂SiO₅ (LSO) doped with Cerium. In the lanthanum halides based on the application of cerium Ce³⁺ as a luminescence centre, the allowed 5d-4f transition makes possible the response
times in the range of 10 ns to 100 ns. In addition to the ability of incorporating the Ce$^{3+}$ ions, the scintillators with high density and Z and a relatively small gap between the valence and the conduction bands, with potential to realize $> 50,000$ photons per MeV [6] were considered.

The Cerium-doped lanthanum chloride (LaCl$_3$:Ce) scintillator was discovered by Delft and Bern Universities [7]. Since 2003, it is commercially made available by Saint-Gobain Crystals with a trade name of BrilLance® 350 (B350) crystals. The new scintillation detector has been extensively studied and found to show excellent luminescence and scintillation properties, superior to that for NaI(Tl) [8].

This paper investigates this new scintillation detector and addresses the essential question of how the energy resolution of an inorganic scintillator can be improved and, consequently, the scintillation applications can be extended. In view of its attractive properties, study and characterization of the LaCl$_3$:(10%)Ce detector have been carried out for its applicability to gamma-ray spectroscopy.

2. Experimental and method

2.1. Experimental set-up

The experimental set-up is shown figure 1. In table 1 properties of the investigated LaCl$_3$:Ce detector are compared to other scintillators.

![Figure 1: BaF$_2$ scintillation detector (head to head) with LaCl$_3$:Ce detector and spectroscopy chains.](image)

<table>
<thead>
<tr>
<th>Scintillators</th>
<th>Light Yield (Photons/keV)</th>
<th>Decay Time (ns)</th>
<th>FOM$^*$ (t/LY)$^{1/2}$</th>
<th>Wavelength of maximum emission $\lambda_m$ (nm)</th>
<th>Refractive index at $\lambda_m$</th>
<th>Density (g/cm$^3$)</th>
<th>Thickness (cm) for 50% attenuation (662 keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaI(Tl)</td>
<td>38</td>
<td>250</td>
<td>2.6</td>
<td>415</td>
<td>1.85</td>
<td>3.67</td>
<td>2.5</td>
</tr>
<tr>
<td>B350</td>
<td>49</td>
<td>28</td>
<td>0.8</td>
<td>350</td>
<td>1.9</td>
<td>3.79</td>
<td>2.4</td>
</tr>
<tr>
<td>B380</td>
<td>63</td>
<td>16</td>
<td>0.5</td>
<td>380</td>
<td>1.9</td>
<td>5.29</td>
<td>1.8</td>
</tr>
<tr>
<td>BaF$_2$</td>
<td>1.8</td>
<td>6.7</td>
<td>0.6</td>
<td>~ 210</td>
<td>1.54</td>
<td>4.88</td>
<td>1.9</td>
</tr>
</tbody>
</table>

$^*$Figure Of Merit  LY = Light Yield

2.2. Method and Equipment

2.2.1. Energy resolution measurements

Gamma-ray energy resolution of the LaCl$_3$:Ce scintillation detector was measured using the experimental set-up on fig. 1. The detector was supplied with the Hamamatsu R6231 Mod PMT. The detector sample was irradiated with 661.54 keV $\gamma$-ray ($^{137}$Cs source at a detector-source-distance of approx. 10 cm) and the resulting signal was processed with the ORTEC 113 Preamplifier, shaped with
the spectroscopic amplifier ORTEC 572 and the pulse displayed through the MCA. Measurements were performed at room temperature, with the high voltage set at 750V and the shaping time at 6μs. The influence of both the high voltage and the shaping time on the resolution (FWHM) was studied through a series of other experiments.

2.2.2. Coincidence Timing resolution measurements

Coincidence timing resolution for LaCl$_3$:Ce detector was measured using the experimental set-up in figure 2.

**Figure 2:** Experimental set-up for Coincidence Timing Measurements

Two BaF$_2$ detectors, one in the START channel and the other in the STOP channel, were operated at -2400V in the first configuration set-up. Upon irradiation of each crystal with 1333.52 keV γ-rays $^{60}$Co source, the signal from the Anode of each PMT was processed by an ORTEC-935 Quad Constant Fraction Discriminator (CFD). The delay in the STOP signal was set at 32 ns by using the ORTEC-424A Delay Output. The resulting timing signals start and stop a Time to Amplitude Converter (ORTEC-TAC/SCA Model 567) whose the output was digitized and processed by the MCA through the Analog Digital Converter (ADC) card to produce a timing spectrum. The BaF$_2$ detector in the STOP channel was then replaced with the LaCl$_3$:Ce detector in the second configuration set-up.

3. Results

3.1. Energy resolution

Figure 3 shows the spectra recorded with BaF$_2$ and LaCl$_3$:Ce detectors in set-up configuration of figure 1.

**Figure 3:** showing the two spectra recorded with the two detectors irradiated with $^{152}$Eu, $^{60}$Co and $^{137}$Cs sources during calibration procedure
Energy resolutions and uncertainties for both lanthanum chloride and Barium detectors are summarized in table 2.

### Table 2: Energy resolution for $^{137}$Cs, $^{60}$Co and $^{152}$Eu sources

<table>
<thead>
<tr>
<th>Source</th>
<th>Energy (keV)</th>
<th>FWHM (keV)</th>
<th>Resolution (%)</th>
<th>Log E</th>
<th>Log R</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Cs</td>
<td>661.54</td>
<td>19.74</td>
<td>2.99±0.02</td>
<td>2.82</td>
<td>0.47</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>1173.2</td>
<td>13.39</td>
<td>2.32±0.01</td>
<td>3.06</td>
<td>0.37</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>1332.5</td>
<td>13.57</td>
<td>2.05±0.01</td>
<td>3.12</td>
<td>0.31</td>
</tr>
<tr>
<td></td>
<td>121.8</td>
<td>10.62</td>
<td>8.66±0.02</td>
<td>2.08</td>
<td>0.93</td>
</tr>
<tr>
<td></td>
<td>344.3</td>
<td>18.19</td>
<td>5.23±0.01</td>
<td>2.53</td>
<td>0.73</td>
</tr>
</tbody>
</table>

The response of the lanthanum chloride detector to variations of both the high voltage and the shaping time are displayed figures 4 and 5, respectively.

**Figures 4 and 5:** Response of the lanthanum chloride detector against variations of HV and shaping times with $^{137}$Cs source
3.2. Coincidence Timing Resolution

In figure 6 are shown spectra and results achieved following the set-up configuration of figure 2.

**Figure 6:** Coincidence timing resolution for (a) BaF$_2$- BaF$_2$ detectors and (b) LaCl$_3$:Ce in STOP channel and BaF$_2$ in START channel

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4. Discussions and Conclusions

Experimental results and calculations provided energy resolution values achieved with LaCl$_3$:Ce detector and shown in table 2. Apart from the $^{152}$Eu spectrum, which showed relatively high energy resolution, LaCl$_3$:Ce detector displayed excellent energy resolution figures. The figure of $(2.99\pm0.02)\%$ achieved in these experiments is three times better than that obtained with the BaF$_2$ detector. It is in good agreement with different values of energy resolution reported elsewhere for 662 keV $^{137}$Cs source [10,11,7]. Resolution results with LaCl$_3$:Ce are also good using $^{60}$Co spectrum for 1173.2 keV peak. By achieving these results, inorganic Scintillators realize their best performance ever reported in the literature [7,10,11] and seriously are challenging semiconductor detectors such as CdTe and CdZnTe [12,10].
Furthermore, using a high voltage of 735 V, the resolution value of 3.6 % (662 keV) has been achieved by the manufacturer upon delivery of the detector. In order to study the relationship between the FWHM and HV, this figure (735V) was compared with the results achieved in our experiments. Detector performance was also studied through a set of measurements, including the relationship between FWHM and shaping times as illustrated in figures 4 and 5. Finally, what really explain the best performance (in terms of energy resolution) achieved with the LaCl₃:Ce detector?

It results from the graph plotted in figures 4 and 5 that resolution (FWHM) can be affected inversely by changing both high voltage and shaping time values. Using a value of 750V, the optimum operating point for this detector was reached even if the best resolution figure is achieved with higher HV values (850 V in these experiments). This figure is in agreement with the one (735V) achieved by the manufacturer during the calibration procedure. On the other hand, the best value of energy resolution (between 2.5% and 3%) is achieved with an optimum shaping time of 3μs. This dependence between these two quantities (HV and shaping times) and the energy resolution suggests the advantage of selecting optimum operating parameters of the detector during experiments.

With regard to the energy resolution performance, for a typical scintillation detector, there are many factors that can contribute to achieving a good energy resolution. From the development viewpoint, the light output and the number of electron-holes (N_{phe}) pairs produced, and the detector size, are the most important factors to be considered. In particular for the LaCl₃:10%Ce, resolution is also dependent on the dopant concentration [6, 11]. In halide scintillators, in particular with chloride compounds, a higher doping concentration (10% Ce) allowed to achieve a high light yield (~ 49000 photons/MeV). In addition, the fundamental limit on the light yield is governed by the band gap and the value of β [13] (conversion efficiency for creating e-h pair). The light yield being proportional to Nphe, a good energy resolution requires a high light output. Those difficulties have been overcome by the incorporation of the Ce³⁺ ion as a luminescence centre in the lattice material and also by the quality of the host material. Those two technological achievements allow a relatively small band gap between the valence and the conduction bands. However, the energy resolution is maximal when the value of the combined effect of intrinsic and transfer resolutions or the contribution of the non-proportionality effect is small [14].

Figure 6 provides a comparison for the coincidence timing resolution measurements performed with 1333.2 keV ^{60}Co peak for both Barium Fluoride and lanthanum Chloride detectors. Timing resolution value of 435 ps (FWHM) was achieved in BaF₂- BaF₂ detectors configuration. In BaF₂-LaCl₃:Ce detectors set-up, the value of 1.07 ns FWHM was measured. In similar study [15], where detectors were irradiated with a 662 keV source, values of 273 ps HWHM and 264 ps FWHM were found for both BaF₂ and LaCl₃:Ce detectors respectively. These values are up to 4 times better than those achieved in the present experiment for the same detectors and configurations. Compared to BaF₂ detector, which considered to be the benchmark for fast timing measurements (with ~ 0.6 ns), LaCl₃:Ce detector is hence 4 times less faster. However, it should be noticed that both experiments were carried out using different radioisotope sources with different energies and different acquisition times.

The efficiency results obtained are as close as possible to those displayed by conventional scintillation detectors. Absolute efficiency values are low. This is due the small detector thickness (2.4 cm). On the other hand, the intrinsic efficiency values obtained in these experiments are good and in agreement with those of most established scintillators. Nevertheless, scintillation detectors are known to be more efficient in detecting γ-rays than semiconductor-based detectors.

The performance expected from the LaCl₃:Ce detector being achieved, what now the future of inorganic scintillators looks like? With 70000 photons per MeV and a resolution of 2.8% and a decay time of 25 ns, the Cerium-activated Lanthanum Bromide detector (LaBr₃:Ce³⁺), is the best scintillator for the moment. However, the demand for better ones is always growing. The improvements to be addressed include the density of the host material, the light output and the related energy resolution. A record of high phonon yield approaching 95000 Photons/MeV has been already achieved with the LuI₃:Ce³⁺ scintillator [6]. The decay time of 24 ns was reached and the density value of 5.6 gcm⁻³.
realized is also higher than that of LaBr$_3$:Ce$^{3+}$. As mentioned above, with smaller band gap scintillator materials, high light output can be achieved. Therefore, the area of ultimate interest could be found among compounds presenting forbidden band gap energy $E_g$ of about 3 eV. In this regards, potential candidates can be found among iodides, selenides and tellurides. With such materials, an ultimate energy resolution below 2% for 662 keV $\gamma$-rays can be predicted with Ce$^{3+}$ scintillators. That, of course inaugurates future fields of research.

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5. References