Scintillation Properties of Ø 1×1 Inch^3 LaBr$_3$: 5%Ce^{3+} Crystal

G. Bizarri, J. T. M. de Haas, P. Dorenbos, and C. W. E. van Eijk, *Member, IEEE*

***Abstract*—The scintillation properties of the first large Ø 1×1 inch^3 LaBr$_3$:5%Ce^{3+} scintillator were studied under γ-ray excitation. This crystal is a LaBr$_3$:5%Ce^{3+} standard product from Saint Gobain company. The sample was cut and polished by the manufacturer from a larger size crystal boule. It was then assembled in a cylindric aluminum case with a front glass window. The results are compared with those recorded with a 0.4×0.3\,$\times 0.5$ inch^3 unpacked crystal. The highest light yield of 21500 ± 2200 photoelectrons per MeV (60500 ± 6000 photons per MeV) of absorbed γ-ray energy was measured for the unpacked crystal—compared to 16800 ± 1700 photoelectrons per MeV for the Ø1×1 inch^3 sample. At room temperature the scintillation decays with a decay time of 16 ± 1 ns. The $0.4 \times 0.3 \times 0.5$ inch^3 unpacked crystal–coupled to photomultiplier tube (Hamamatsu R1791) and measured at 662 keV–exhibited an energy resolution of 2.8%, better than the one of the Ø1×1 inch^3 crystal (3.1%). Special attention was devoted to energy and temperature dependencies of scintillation properties: the results show a good proportionality for light output versus γ-ray energy. The light output deviates from the linear response of 5% between 20 keV and 2 MeV. Furthermore temperature dependency of gamma-ray excited decay time (80–700 K) does not indicate the presence of thermal quenching from the cerium 5d state. These characteristics suggest a broad range of applications for the LaBr$_3$:5%Ce^{3+} scintillator.**

***Index Terms*—Bromide compounds, γ detection, energy resolution, nonproportionality, scintillation crystal.**

I. INTRODUCTION

IN view of the attractive properties of LaCl$_3$:Ce^{3+} and LaBr$_3$:Ce^{3+} crystals, intensive research has been made to report their properties according to the dopant concentration and to the crystal size. The knowledge of LaCl$_3$:Ce^{3+} scintillation properties which were discovered one year earlier than those of LaBr$_3$:Ce^{3+}, are now well known [1], [2] whereas the scintillation characteristics of LaBr$_3$:Ce^{3+} are still incomplete. For LaBr$_3$:Ce^{3+} scintillator, the majority of experimental results published to date are from samples activated with a relatively low level of cerium concentration [3] and/or from small sized crystals smaller than 2.5 cm^3 [4], [5].

In 2001 we reported on the new scintillator LaBr$_3$:Ce^{3+} [6]. The typical size of these crystals was about $3 \times 3 \times 10$ mm^3 with cerium concentrations varying from 0.5 to 10%. Within three years after this initial report, the growth of LaBr$_3$:Ce^{3+} has been scaled up to sizes of Ø19×19 mm^3 for 0.5% cerium-doped samples [3].

In this article we report on the scintillation properties of the first large Ø1×1 inch^3 packed LaBr$_3$:5%Ce^{3+} crystal. The sample is a LaBr$_3$:Ce^{3+} standard product from Saint Gobain company. The sample was cut and polished by the manufacturer from a large size crystal boule. It was then assembled in an aluminum case with a front glass window. For comparison a $0.4 \times 0.3 \times 0.5$ inch^3 bare LaBr$_3$:5%Ce^{3+} crystal, also from Saint Gobain company, was used in this study. Pulse height spectra, γ-ray excited decay curves, and afterglow luminescence are presented. The good scintillation properties, which were observed initially on small sized and/or low concentration samples, are maintained on large LaBr$_3$:5%Ce^{3+} crystal. Finally, some aspects of LaBr$_3$:5%Ce^{3+} scintillation mechanism are also discussed.

II. EXPERIMENTAL

To improve the legibility of the article, the two samples are referred to as *sample A* and *sample B* for the Ø1×1 inch^3 packed crystal and the $0.4 \times 0.3 \times 0.5$ unpacked crystal, respectively.

Pulse height spectra were recorded with a Hamamatsu R1791 photomultiplier tube (Quartz version of Hamamatsu R878 PMT) connected to a homemade preamplifier, an Ortec 672 spectroscopic amplifier and an Ortec AD114 CAMAC Analog to Digital Converter. The PMT high voltage was fixed at -600 V. Sample A was optically coupled onto the window of the PMT with Viscasil 60 000 cSt from General Electric, whereas sample B was directly mounted on the PMT. To improve the collection of scintillation light, sample B was covered with layers of ultraviolet-reflecting Teflon tape (PFTE tape).

An Amersham variable X-ray source was used to excite the crystals at energies between 13.4 and 44.5 keV. This source contains ^{241}Am that produces characteristic K$_\alpha$ and K$_\beta$ X-rays from Rb, Mo, Ag, Ba, and Tb targets. ^{241}Am, ^{133}Ba, ^{137}Cs, ^{22}Na, and ^{60}Co gamma-ray sources were used for exciting with gamma-ray energies between 59.5 keV and 1.33 MeV. The high light yield of LaBr$_3$:Ce^{3+} combined with a very fast decay may lead to nonproportionality in the PMT gain and distorted pulse height spectra results. To cope with high peak currents we used a voltage divider as described in [3].

The light yield, expressed in photoelectrons per MeV of absorbed γ energy, was determined by comparing the peak position of the photopeak in the pulse height spectra (Shaping Time 2 μs) with that of the single photoelectron peak [7]. To obtain the absolute light yield of sample B, formula 1 of [8] was used

$$Y_{\rm ph} = Y_{pe}\frac{1 - R_{\rm eff}}{R_{\rm PTFE} QE_{\rm eff}} \tag{1}$$

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The authors are with the Faculty of Applied Sciences, Delft University of Technology, 2629 JB Delft, The Netherlands (e-mail: g.bizarri@tnw.tudelft.nl).

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TABLE I
LIGHT YIELDS AND ENERGY RESOLUTIONS VALUES DERIVED FROM PULSE HEIGHT SPECTRA OF $LaBr_3$:5%Ce^{3+}, SAMPLE A AND SAMPLE B, UNDER ^{137}Cs (662 keV) γ-RAY EXCITATION

$LaBr_3$:5%Ce^{3+}	Size (inch^3)	Y_{phe} (phe/MeV)	Y_{ph} (ph/MeV)	R (%)
Sample A	Ø1×1	16800	-	3.1
Sample B	$0.4 \times 0.3 \times 0.5$	21500	60500	2.8

where QE_{eff} and R_{eff} are respectively the effective quantum efficiency and effective reflectivity of the R1791 PMT photocathode. Both are averaged over the spectral profile of the scintillator emission spectrum. The calculated values are 28.7% and 20% for QE_{eff} and R_{eff}, respectively. The estimated value of PTFE package reflection (R_{PTFE}) is 0.98. The energy resolution R (FWHM over peak position) was deduced by fitting the photopeak for each pulse height spectrum.

The scintillation decays were recorded by using an experimental set-up that allows for temperature dependent measurements. The samples are fixed at the bottom of a parabolic-like stainless steel reflector. Both are mounted onto the cold finger of a Janis cryostat (80–700 K). The sample and the reflector face a PMT, which collects nearly all the scintillation light pulse. This PMT acts as the Start PMT (XP2020Q). A hole in the back of the reflector allows a few of the scintillation photons to reach a second PMT acting as the Stop PMT (XP2020Q). The electronic part of the set-up is identical to the conventional delayed-coincidence method described by Bollinger *et al.* [9]. LeCroy 934 Constant Fraction Discriminators (CFDs) and a LeCroy 4208 Time to Analog Converter (TAC) were used.

The afterglow spectra were recorded at room temperature using an X-ray tube with Cu anode, operating at 35 kV and 25 mA. The crystals were exposed to X-rays for 30 s. A Hamamatsu R943-02 PMT (HV − 2100 V) was used to measure the total emission intensity (number of counts per second as function of time).

III. SCINTILLATION PROPERTIES OF LARGE $LaBr_3$:5%Ce^{3+} CRYSTALS

A. Light Output and Energy Resolution

Pulse-height spectra under ^{137}Cs (662 keV) γ-ray excitation of $LaBr_3$:5%Ce^{3+}, sample A and B, have been recorded. Light yields and energy resolutions derived from these measurements are compiled in Table I.

Yields of 16 800 ± 1700 photoelectrons and 21 500 ± 2000 photoelectrons (60 500 ± 6000 photons) per MeV of absorbed gamma ray energy at 662 keV were measured for sample A and sample B, respectively. The 20% drop in light output between both samples can be associated with an imperfect packing of sample A. These results demonstrate that the light output of $LaBr_3$:5%Ce^{3+} is not strongly affected by the crystal size and the cerium concentration. The absolute light yield for sample B is close to the 55 300 ph/MeV found by Shah *et al.* for a four times smaller $LaBr_3$:5%Ce^{3+} sample [5]. And of the same order than those measured by van Loef *et al.* on small sized 0.5% cerium-doped crystals [4].

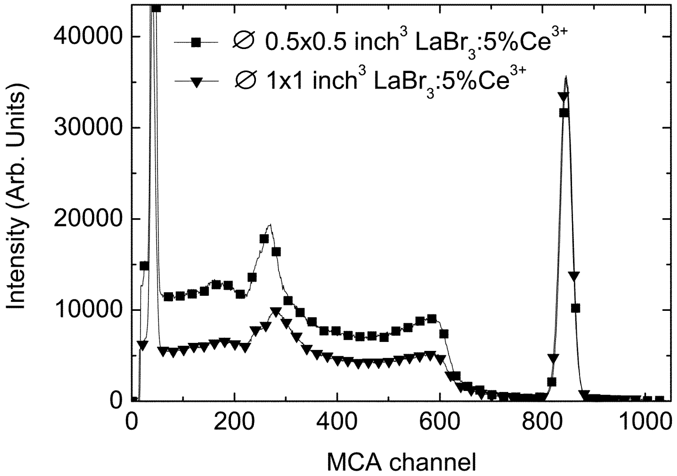

Fig. 1. Pulse height spectra of $LaBr_3$:5%Ce^{3+}, Ø1×1 inch^3 and Ø0.5×0.5 inch^3 crystals, under ^{137}Cs (662 keV) γ-ray excitation, recorded with a shaping time of 2 μs. The spectra are normalized with respect to the light output value at 662 keV energy.

Finally, the absolute light yield of sample A is very close to the expected fundamental limit of 63 000 ph/MeV for this compound. This theoretical value was estimated from formula 2 of [10] assuming $\beta = 2.5$

$$Y_{\mathrm{ph}} = \frac{E_\gamma}{\beta E_g} \tag{2}$$

with E_γ the gamma-ray energy and E_g the band gap energy. This result shows that losses during the scintillation process are very small. This point was also noticed for other chloride and bromide compounds; those showing the highest values, K_2LaCl_5:Ce [11], $LaCl_3$:Ce [12], $RbGd_2Br_7$:Ce [13] also fall very close to the fundamental limit.

The energy resolution R for the 662 keV full energy peak are 3.1% and 2.8% for sample A and B, respectively. These values are the best for inorganic scintillation detectors reported to date [14]–[16].

The comparison between the energy resolution for sample A and for a smaller packed crystal (Ø0.5×0.5 inch^3 $LaBr_3$:5%Ce^{3+}) is presented in Fig. 1. The energy resolution value of both samples is identical; the energy resolution is not influenced by the crystal size.

B. Time Profile

Scintillation decay time spectra of sample A at room temperature under ^{137}Cs γ-ray and optical excitation are shown in Fig. 2. The gamma-ray excited decay curve corresponds to a single exponential characterized by a lifetime of 16 ± 1 ns. The rise time is less than 1 ns. These two characteristics were also observed for small crystal sizes with the same dopant concentration [5]. The fast decay time and the short rise time support the hypothesis that the main scintillation mechanism is due to *sequential capturing of first the hole and then the electron* by Ce^{3+} ions. This hypothesis is consistent with the value of Ce^{3+} intrinsic lifetime in $LaBr_3$ measured under optical excitation around 15 ns (Fig. 2).

Dopant concentration dependency of gamma-ray excited decay time can also be observed in Fig. 2. The rise time and

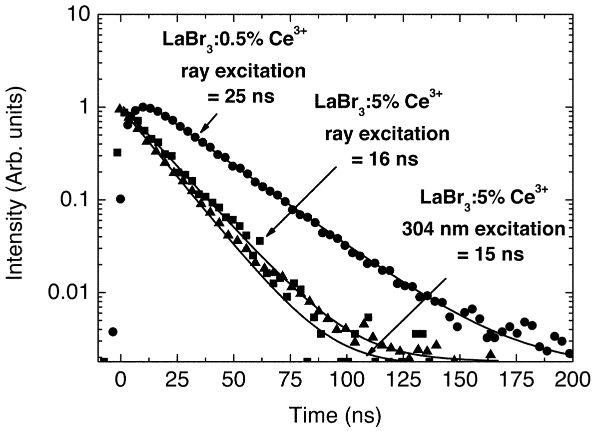

Fig. 2. Scintillation decay curves at room temperature under ^{137}Cs γ-ray of LaBr$_3$:5%Ce^{3+}, and LaBr$_3$:0.5%Ce^{3+}. For comparison the LaBr$_3$:5%Ce^{3+} UV excited decay curve is also plotted.

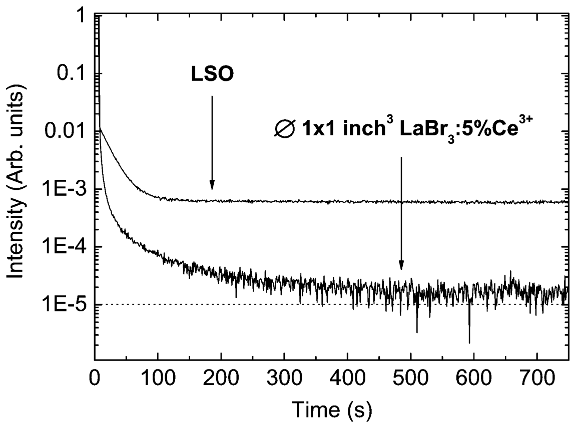

Fig. 3. Afterglow of Ø1 × 1 inch^3 LaBr$_3$:5%Ce^{3+} and LSO:0.2%Ce crystals at room temperature. The crystals were exposed for 30 s to an X-ray beam (λ_{Cu}, 35 kV and 25 mA).

the decay time significantly decrease as the cerium ion concentration increases from 0.5% to 5%. The rise time decreases from few nanoseconds to less than 1 ns as the decay time is shortened from 25 ns to 16 ns. Such observation was recently done by Glodo *et al.* [17] on small crystal sizes.

C. Afterglow

A comparison between afterglow behavior in LaBr$_3$:Ce and Lu$_2$SiO$_5$:Ce (LSO) is presented in Fig. 3. For LSO:Ce, after 200 s, the residual intensity induced by the X-ray exposure is about 0.2% of the initial intensity. Furthermore this intensity is still decreasing after several minutes, which demonstrates a strong afterglow in the lutetium oxyorthosilicate scintillator [18]. In the case of LaBr$_3$:Ce, this residual intensity is less than 0.01% after 200 s. After 500 s the intensity remains constant, corresponding to the background of the measurement. The background is similar for both spectra (dashed line in Fig. 3).

IV. ENERGY AND TEMPERATURE DEPENDENCE

A. Non-Proportionality Response

The proportionality response of sample A at room temperature has been evaluated. Light yield nonproportionality in a function of energy is one of the important reasons for degraded energy resolution of scintillators [19]. The data points, shown in Fig. 4, were normalized with respect to the light output value at 662 keV energy. The light yield of LaBr$_3$:5%Ce^{3+} in Fig. 4 shows a very good proportionality at energies between 20 keV and 2 MeV (decrease less than 5%). At lower energies (from 20 to 6 keV), there is a decrease in efficiency by about 15%. This behavior is about the same as the proportionality response curve measured for a smaller crystal with a lower cerium ion concentration (Ø19 × 19 mm^3 LaBr$_3$:0.5%Ce^{3+}) [3]. We conclude that cerium concentration and crystal size do not seem to be relevant parameters in nonproportionality response of LaBr$_3$:Ce^{3+} material.

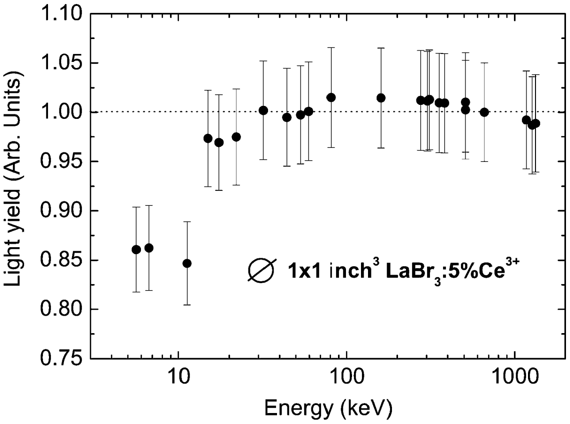

Fig. 4. Proportionality in the light yield as a function of γ-ray energy measured at room temperature for Ø1 × 1 inch^3 LaBr$_3$:5%Ce^{3+} crystal.

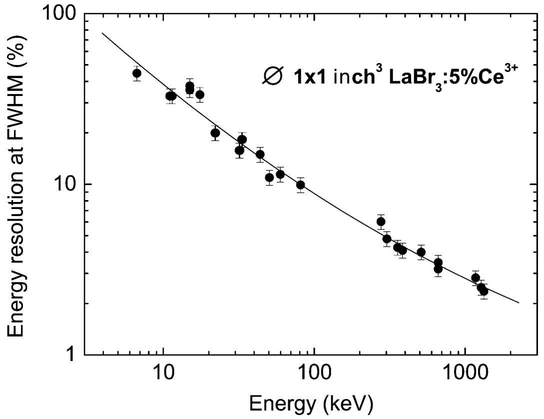

Fig. 5. Energy resolution as a function of γ-ray energy measured at room temperature for Ø1 × 1 inch^3 LaBr$_3$:5%Ce^{3+} crystal. The solid curve is drawn to guide the eye.

B. Energy Resolution

The energy resolution R has been determined from peaks in the pulse height spectra. The data are plotted as a function of energy on a double logarithmic scale in Fig. 5. These measurements were obtained from sample A. The data between 6 keV and 2 MeV display an energy resolution that decreases with the square root of the gamma ray energy. The energy resolution is 3.1% for an excitation energy of 662 keV. The best energy resolution, 2.5%, is found for an excitation energy of 1.17 MeV. These values are similar to the ones which have been reported in the literature for low cerium ion concentration LaBr$_3$:Ce^{3+} crystals [3] and/or small sized scintillators [4]. These results confirm our previous conclusions; the upgrade from the small sample to a large one does not lead to much deterioration of

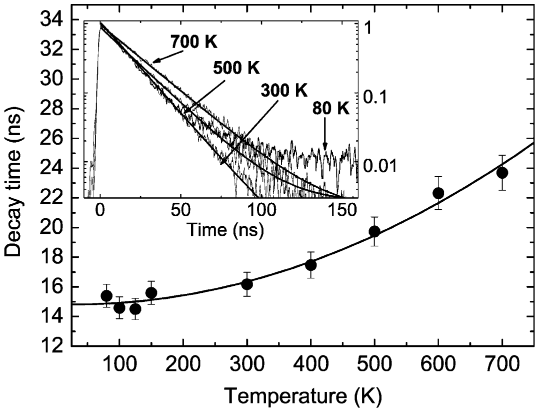

Fig. 6. Temperature dependent decay time of LaBr$_3$:5%Ce^{3+} fast component under γ-ray excitation. The inset presents four decay curves measured at 80, 300, 500, and 700 K. The solid curve is drawn to guide the eye.

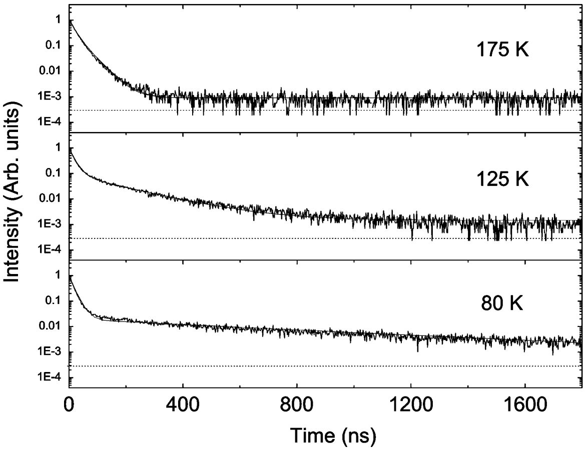

Fig. 7. Temperature-dependent decay curves of the slow component in decay curves of LaBr$_3$:5%Ce^{3+} measured under γ-ray excitation. The dashed curve shows background level for each measurement.

LaBr$_3$:Ce^{3+} scintillator performance. And the cerium ion concentration, from 0.5 to 5%, does not have any influence on the energy resolution of LaBr$_3$:Ce^{3+} crystals.

C. Decay Time

The temperature dependence from 80 K to 700 K of fluorescence lifetime τ under gamma-ray excitation (^{137}Cs source) was studied on sample A. Above 200 K, the decay curves exhibit a fast, single exponential decay. This component is fast. Below 200 K, a long decay component extending into the microsecond region is also present. Temperature dependency of the fast decay component is displayed in Fig. 6. Fig. 7 shows the long decay component measured for three different temperatures at 80, 125, and 175 K. For the fast component, the lifetime increases from 15 ns to 24 ns respectively from 80 K to 700 K. This behavior seems to be a common trend for cerium-doped compounds [20]. Lyu *et al.* have observed an increase of cerium emission decay time in several fluoride and oxide compounds under laser excitation in cerium 5d states. In all their observations, the increase is followed by a rapid decrease of τ attributed to the onset of thermally induced nonradiative transitions. However the increase of cerium lifetime in LaBr$_3$:5%Ce^{3+} is more pronounced (>50%) compared to those measured by Lyu *et al.* (5–30% depending on the compound).

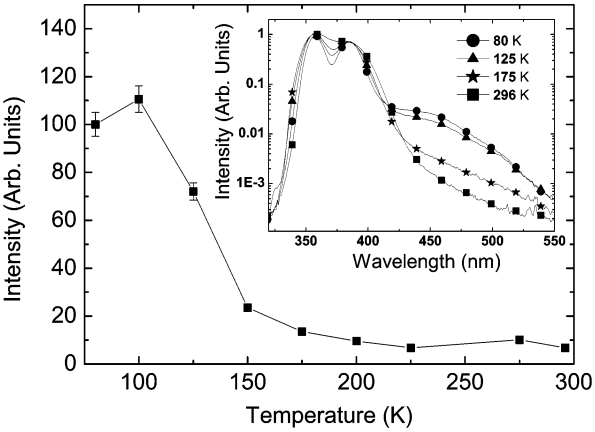

Fig. 8. Relative emission intensity of LaBr$_3$:5%Ce^{3+} recorded at 450 nm under X-ray excitation. The inset presents X-ray excited emission spectra at 80, 125, 175, and 300 K.

For the long component, the lifetime decreases with the increase of temperature from 80 K to 200 K (Fig. 7). The long component decay times are 800 (50%), 165 (45%), and 40 (50%) ns at 80, 125, and 175 K, respectively. Above 200 K the slow component disappears. A decay time on the order of a few microseconds that shortens with increased temperature has been observed for several cerium-doped compounds. It has been attributed to an *energy transfer from STE to cerium ions* [15]. In this model the life time of the long cerium emission is governed by the life time of STE emission. The value of about 1 μs found at 80 K is consistent with the expected decay time of STE emission in this sample.

The decrease of STE luminescence with increase of temperature supports this model. The inset of Fig. 8 shows the X-ray excited emission as function of temperature. The Ce^{3+} emission doublet at 356 and 385 nm is seen together with a band around 450 nm corresponding to the STE luminescence [4]. Fig. 8 presents the relative emission intensity measured at 450 nm as function of temperature. This result clearly shows a correlation between the occurrence of the STE emission and the occurence of the slow cerium decay component. Both are measurable only between 80 and 200 K. Further investigations are in progress to validate this mechanism.

V. SUMMARY AND CONCLUSIONS

We investigated cerium-doped lanthanum bromide scintillation crystals for γ detection. Our research concentrated on the scintillation properties of the first large Ø1 × 1 inch^3 sized LaBr$_3$:5%Ce^{3+} crystal.

This study confirms and completes earlier observations on small LaBr$_3$:Ce^{3+} crystals and high doped samples. Ø1 × 1 inch^3 LaBr$_3$:5%Ce^{3+} is a fast scintillator ($\tau \simeq 16$ ns at room temperature, without long components) with a high absolute light output (around 60 000 ph/MeV). The sample has an excellent energy resolution of about 3% for γ-rays from a ^{137}Cs source and a very good proportionality response as function of energy (less than 5% from 20 keV to 2 MeV). Our studies indicate that scintillation properties are maintained as the crystal volume increases up to Ø1×1 inch^3.

Finally, we have highlighted two different processes in the scintillation mechanism of LaBr$_3$:5%Ce^{3+} crystals. The scintillation is mainly due to a fast energy transfer process (Fig. 6). This mechanism is likely linked to the sequential capture of electron/hole pairs by cerium ions. In addition to this fast process, a slow energy transfer mechanism occurs at low temperature (Fig. 7). This process is likely correlated to the transfer from *STEs* to cerium ions. The mechanism was assumed to be present in low concentration LaBr$_3$ crystals, but has never been observed in high concentration samples. The nature of these mechanisms and the one of the fast decay time temperature dependency are a matter for continued investigations.

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