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The scintillation mechanism in $\text{LaCl}_3:\text{Ce}^{3+}$

E V D van Loef, P Dorenbos¹ and C W E van Eijk

Radiation Technology Group, Interfaculty Reactor Institute, Delft University of Technology,
Mekelweg 15, 2629 JB Delft, The Netherlands

E-mail: dorenbos@iri.tudelft.nl

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Abstract

The scintillation properties of LaCl_3 crystals, doped with Ce^{3+} concentrations of 0.57, 1.0, 2.0, 4.0, and 10%, are studied under x-ray and γ -quanta excitation at various temperatures. Under x-ray excitation, characteristic doublet Ce^{3+} emission is observed with a maximum at 3.7 eV. Also self-trapped exciton (STE) emission is observed with a maximum near 3.1 eV. The contribution of STE luminescence to the total light yield decreases with increasing Ce concentration. For $\text{LaCl}_3:0.57\% \text{Ce}^{3+}$, the contribution is 33%, whereas for $\text{LaCl}_3:10\% \text{Ce}^{3+}$ it is 4%. The total light yield of the Ce^{3+} -doped samples ranges from 38 000 photons per megaelectronvolt (ph MeV^{-1}) of absorbed γ -ray energy to 47 000 (ph MeV^{-1}) for $\text{LaCl}_3:0.57\% \text{Ce}^{3+}$ and $\text{LaCl}_3:10\% \text{Ce}^{3+}$, respectively. A model is proposed to explain the energy transfer from the host lattice to the Ce^{3+} ions. At these Ce concentrations, energy transfer by STE diffusion is held to be dominant.

1. Introduction

$\text{LaCl}_3:\text{Ce}^{3+}$ scintillators show a high light output, fast response, and excellent energy resolution. The scintillation properties have already been published by us in various papers [1, 2], but a detailed investigation into the scintillation mechanism has not yet been commenced. In this work, we review the scintillation properties of $\text{LaCl}_3:\text{Ce}^{3+}$ and present new data. We have used spectroscopic and magnetic methods in order to get information on the radiation-induced luminescence of $\text{LaCl}_3:\text{Ce}^{3+}$ and its temperature dependence. It is shown that self-trapped exciton (STE) diffusion is the dominant scintillation mechanism in $\text{LaCl}_3:\text{Ce}^{3+}$.

¹ Author to whom any correspondence should be addressed.

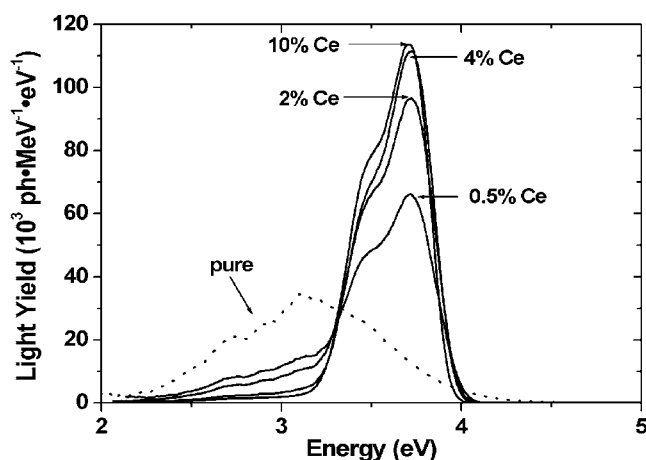


Figure 1. X-ray-excited optical luminescence spectra of pure LaCl_3 and $\text{LaCl}_3:0.57$, 2, 4, and 10% Ce^{3+} at room temperature. The spectra are corrected for the wavelength dependence of the photodetector quantum efficiency and the transmission as well as monochromator transmission.

2. Experimental details

Single crystals of LaCl_3 with 0.57, 1, 2, 4, and 10% Ce^{3+} concentrations were grown by the vertical Bridgman technique using a moving furnace and a static vertical ampoule. The preparation has already been described in [1]. LaCl_3 has the UCl_3 -type structure [3], space group $P6_3/m$ (No 176). The lattice is not layered and does not cleave easily. The melting point of LaCl_3 is 859 °C. The Ce^{3+} concentration in the crystals was determined by induction-coupled plasma spectroscopy (ICPS); to achieve this, a small part of the sample was dissolved and sprayed into an argon plasma, and the Ce^{3+} emission detected. Details on experimental techniques can be found in [1].

3. Results

X-ray-excited optical luminescence spectra of $\text{LaCl}_3:0.57$, 2, 4, and 10% Ce^{3+} at room temperature are shown in figure 1 [2]. Also shown is the spectrum of pure LaCl_3 (dotted trace). The spectra are corrected for the wavelength dependence of the photodetector quantum efficiency as well as monochromator transmission. Each spectrum is normalized such that its integral is equal to the value for the absolute light yield expressed in photons per megaelectronvolt (ph MeV^{-1}), as found from pulse height spectra recorded with a shaping time of 10 μs . In addition to intense Ce^{3+} luminescence peaking at 3.7 eV, a broad (FWHM = 0.8 eV) emission band is observed for the pure chloride with a maximum near 3.1 eV. We have attributed this to luminescence from STEs [4–6]. With increasing Ce^{3+} concentration, the intensity of the STE luminescence decreases and that of the Ce^{3+} luminescence increases. For $\text{LaCl}_3:10\%$ Ce^{3+} , the contribution of Ce^{3+} emission to the total light is about 35 times higher than that of the STE, whereas for $\text{LaCl}_3:0.57\%$ Ce^{3+} , it is only two times higher. Assuming that the peak position and the width of the STE luminescence band remain equal for all Ce^{3+} concentrations, the light yields of both the STE and the Ce^{3+} luminescence were estimated. The results are shown in table 1.

The temperature dependence of the Ce^{3+} and STE luminescence intensity in $\text{LaCl}_3:\text{Ce}^{3+}$ were measured for pure LaCl_3 and $\text{LaCl}_3:0.57$, 2, 4, and 10% Ce^{3+} . The results on

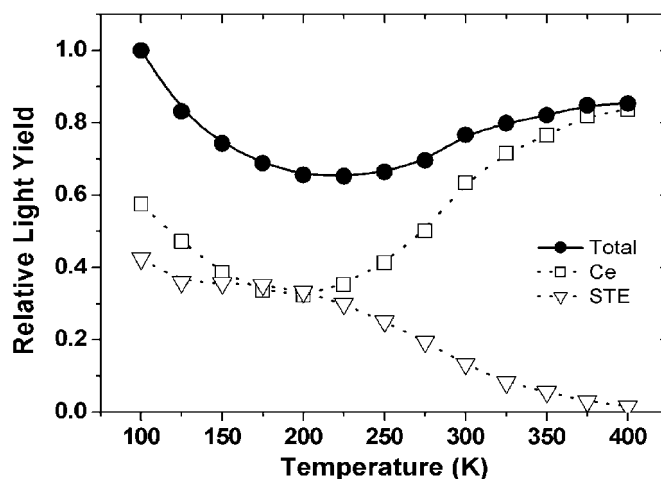


Figure 2. Temperature dependences of the light yields of Ce^{3+} , the STE, and the total luminescence of $\text{LaCl}_3:2\% \text{Ce}^{3+}$, derived from x-ray-excited optical luminescence spectra.

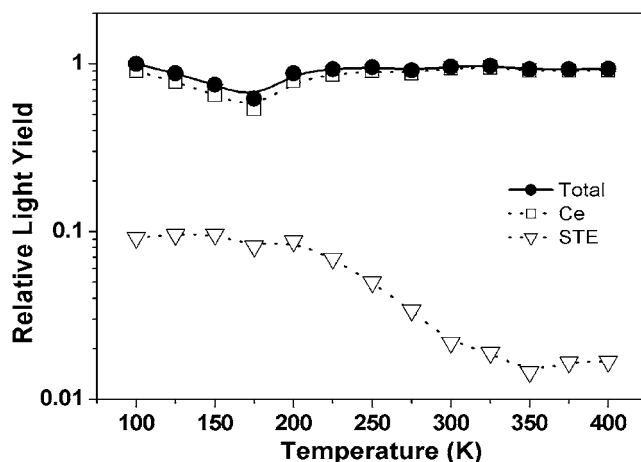


Figure 3. Temperature dependences of the light yields of Ce^{3+} , the STE, and the total luminescence of $\text{LaCl}_3:10\% \text{Ce}^{3+}$, derived from x-ray-excited optical luminescence spectra. Note the logarithmic scale of the ordinate.

$\text{LaCl}_3:0.57\% \text{Ce}^{3+}$ [1] and $\text{LaCl}_3:4\% \text{Ce}^{3+}$ [2] have already been published. Here, we show the temperature dependences of the light yields of Ce^{3+} , the STE, and the total luminescence for $\text{LaCl}_3:2\% \text{Ce}^{3+}$ and $\text{LaCl}_3:10\% \text{Ce}^{3+}$ in figures 2 and 3, respectively. Between 100 and 225 K, the light yield of $\text{LaCl}_3:2\% \text{Ce}^{3+}$ is due to both Ce^{3+} and STE luminescence; see figure 2. Both decrease with increasing temperature. From 225 up to 400 K, the Ce^{3+} luminescence intensity is enhanced at the expense of STE luminescence intensity while the total light yield increases as well. The anti-correlation between the Ce^{3+} and STE luminescence intensity has been observed by Guillot-Noël *et al* in $\text{LaCl}_3:0.57\% \text{Ce}^{3+}$ [1] and by van't Spijker *et al* in $\text{K}_2\text{LaCl}_5:0.23\% \text{Ce}^{3+}$ [5]. For $\text{LaCl}_3:0.57, 2,$ and $4\% \text{Ce}^{3+}$, it is observed between 230 and 400 K, whereas in the case of $\text{K}_2\text{LaCl}_5:0.23\% \text{Ce}^{3+}$, it is observed between 100 and 300 K. As for $\text{LaCl}_3:2\% \text{Ce}^{3+}$, we do observe a change in the Ce^{3+} and STE luminescence yield for

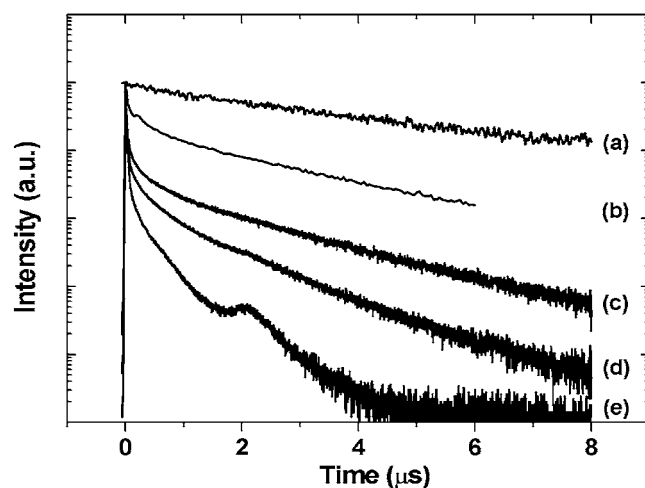


Figure 4. Scintillation decay time spectra at room temperature under ^{137}Cs gamma-ray excitation of (a) pure LaCl_3 , (b) $\text{LaCl}_3:0.57\% \text{Ce}^{3+}$, (c) $\text{LaCl}_3:2\% \text{Ce}^{3+}$, (d) $\text{LaCl}_3:4\% \text{Ce}^{3+}$, and (e) $\text{LaCl}_3:10\% \text{Ce}^{3+}$.

Table 1. Light yields of $\text{LaCl}_3:\text{Ce}^{3+}$ derived from x-ray-excited optical luminescence spectra. The error in the total, Ce^{3+} , and STE luminescence yields is about 10%.

$[\text{Ce}^{3+}]$ (at.%)	Ce^{3+} light yield (ph MeV^{-1})	STE light yield (ph MeV^{-1})	Total light yield (ph MeV^{-1})
—	—	32 000	32 000
0.57	25 300	12 700	38 000
2	38 200	8 800	47 000
4	44 000	3 000	47 000
10	45 300	1 700	47 000

$\text{LaCl}_3:10\% \text{Ce}^{3+}$ at temperatures in the 200–400 K range; see figure 3. However, at these temperatures the luminescence intensity is mainly due to Ce^{3+} emission. The contribution of STE luminescence to the total light yield is less than 10%. In order to emphasize the anti-correlation between the Ce^{3+} and STE luminescence intensity, we used a logarithmic scale for the ordinate. Between 100 and 175 K, the STE luminescence intensity remains more or less constant, whereas the Ce^{3+} luminescence and total light yield decrease about 40%. Between 200 and 400 K, the Ce^{3+} luminescence and total light yield level at 95% with respect to that at 100 K. In the same temperature interval, the STE luminescence intensity decreases by about 80%.

The scintillation decay time spectra of pure LaCl_3 , $\text{LaCl}_3:0.57$, 2, 4, and 10% Ce^{3+} at room temperature under ^{137}Cs γ -ray excitation are shown in figure 4. The irregularity in curve (e) at $t = 2 \mu\text{s}$ is due to after-pulses in the photomultiplier. For pure LaCl_3 the scintillation pulse decays exponentially with a decay time of about $3.5 \mu\text{s}$ [2]. We assume that it represents the lifetime of the STE at room temperature. For LaCl_3 doped with Ce^{3+} , this component is also present and can be observed in the scintillation pulse at times larger than, say, $1 \mu\text{s}$ after the start of the scintillation pulse. The lifetime of this component becomes increasingly short when the Ce concentration is increased: for $\text{LaCl}_3:0.57$, 2, 4, and 10% Ce^{3+} it is 2.5, 1.8, 1.1, and $0.8 \mu\text{s}$, respectively [1, 2]. The origin of this decay component is related to STE luminescence.

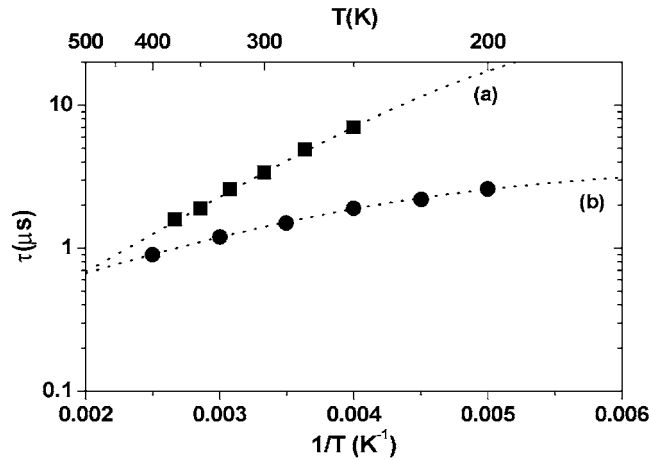


Figure 5. The decay time of the STE, τ , in (a) pure LaCl_3 and (b) $\text{LaCl}_3:1\% \text{Ce}^{3+}$ as a function of temperature.

Table 2. The decay time of the STE, τ_{STE} , and the calculated transfer rate, k_t , as functions of Ce concentration.

[Ce] (%)	τ_{STE} (μs)	k_t (10^5 s^{-1})
—	3.5 ± 0.3	—
0.57	2.5 ± 0.3	1.1 ± 0.1
2	1.8 ± 0.2	2.7 ± 0.3
4	1.1 ± 0.1	6.2 ± 0.6
10	0.8 ± 0.1	9.6 ± 0.9

However, the slow scintillation component contains much more light than expected on the basis of STE intensity alone, as is observed in the x-ray-excited optical luminescence spectra (see table 1). This implies that a large fraction of the Ce^{3+} luminescence is emitted in this slow component as well. For $\text{K}_2\text{LaCl}_5:\text{Ce}^{3+}$ [5] a similar correlation between the lifetime of the STE luminescence and Ce concentration was found. This is caused by energy transfer from the STE to Ce^{3+} . From the decrease of the decay time from $3.5 \mu\text{s}$ in pure LaCl_3 to $2.5 \mu\text{s}$ in $\text{LaCl}_3:0.57\% \text{Ce}^{3+}$, a transfer rate $k_t \approx 1.1 \times 10^5 \text{ s}^{-1}$ for the 0.57% doped sample is obtained. The values of the transfer rate for the other Ce concentrations are compiled in table 2.

Additionally, a fast decay component is present in the scintillation decay of $\text{LaCl}_3:\text{Ce}^{3+}$ that can be observed in the scintillation pulse at times shorter than, say, $0.5 \mu\text{s}$ after the start of the scintillation pulse. The relative contribution of this component to the total light yield becomes increasingly large when the Ce concentration is increased: for $\text{LaCl}_3:0.57, 2, 4,$ and $10\% \text{Ce}^{3+}$ it is 8, 10, 18, and 41%, respectively [1, 2].

In figure 5 the decay time τ of the STE in (a) pure LaCl_3 and (b) $\text{LaCl}_3:1\% \text{Ce}^{3+}$ is presented as a function of $1/T$ (K^{-1}). The dotted curves show model calculations according to the quenching process

$$\frac{1}{\tau} = \frac{1}{\tau_R} + \nu_q \exp\left(-\frac{\Delta E_q}{kT}\right) + \nu_t \exp\left(-\frac{\Delta E_t}{kT}\right) \quad (1)$$

where the first term of (1) is the probability of radiative decay, the second term of (1) the probability of non-radiative decay by thermal quenching, and the third term of (1) the

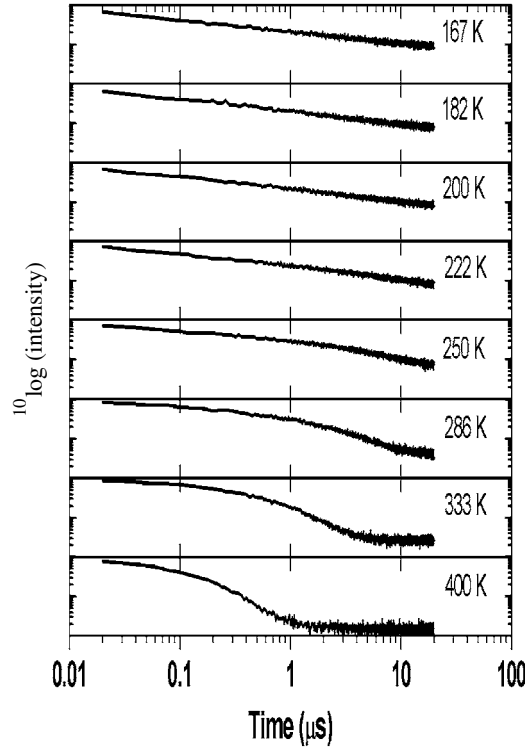


Figure 6. Decay curves of $\text{LaCl}_3:1\% \text{Ce}^{3+}$ as a function of temperature.

Table 3. The lifetime of radiative decay, τ_R , the activation energy and frequency factor for thermal quenching, ΔE_q and ν_q , respectively, and the activation energy and frequency factor for energy transfer, ΔE_t and ν_t , respectively, of the STE in pure LaCl_3 and $\text{LaCl}_3:1\% \text{Ce}^{3+}$.

Compound	Radiative decay	Thermal quenching		Energy transfer	
	τ_R (μs)	ΔE_q (meV)	ν_q (10^6 s^{-1})	ΔE_t (meV)	ν_t (10^6 s^{-1})
LaCl_3	~ 50	106 ± 6	17 ± 1	—	—
$\text{LaCl}_3:1\% \text{Ce}^{3+}$	~ 50	106 ± 6	9 ± 1	23 ± 2	1.3 ± 0.1

probability of energy transfer of the STE to Ce^{3+} centres. In this formula, τ_R is the radiative decay rate of the STE, ΔE_q and ν_q are the activation energy and frequency factor for thermal quenching, respectively, ΔE_t and ν_t are the activation energy and frequency factor for energy transfer from the STE to Ce^{3+} centres, respectively, and k is the Boltzmann constant. From a fit to equation (1), values for τ_R , ΔE_q , ν_q , ΔE_t , and ν_t were obtained. They are summarized in table 3. For $\text{LaCl}_3:1\% \text{Ce}^{3+}$, the values for τ_R and ΔE_q were taken from the fit for pure LaCl_3 and were assumed to be constant. The activation energies for thermal quenching and energy transfer are $\Delta E_q = 106 \pm 6$ meV and $\Delta E_t = 23 \pm 2$ meV, respectively. For comparison, the activation energies for STE diffusion in NaCl [7] and NaI(K) , NaI(Tl) [8] were estimated to be 150 and 70 meV, respectively. The value for the radiative decay rate of the STE ($\tau_R \approx 50 \mu\text{s}$) could not be determined with high accuracy due to the limited temperature range.

Figure 6 shows the decay curves of $\text{LaCl}_3:1\% \text{Ce}^{3+}$ as a function of temperature monitoring Ce^{3+} emission at 3.7 eV. At 167 K, the decay curve of $\text{LaCl}_3:1\% \text{Ce}^{3+}$ shows an almost perfect

power-law dependence between Ce^{3+} luminescence intensity and time, according to

$$I(t) \propto t^{-s} \quad (2)$$

with $s \approx 0.3$. If the temperature is raised, the decay curve of $\text{LaCl}_3:1\% \text{Ce}^{3+}$ gradually turns into an exponential decay curve.

4. Discussion

In the past we proposed several scintillation mechanisms for $\text{LaCl}_3:\text{Ce}$ [1, 9], of which the one involving *STE diffusion* seems to be the most dominant, especially at low Ce concentrations [2]. Recently, two triplet systems have been found at 10 K in the x-ray luminescence EPR of pure and Ce^{3+} -doped LaCl_3 [10]. Stable self-trapped holes (V_K centres) were not detected at low temperatures ($T < 100$ K) in the EPR spectra of pure and Ce^{3+} , probably because the V_K centre immediately ($t < 10^{-9}$ s) captures an electron from the conduction band to form a STE.

The presence of STEs in the EPR of $\text{LaCl}_3:\text{Ce}^{3+}$ and the absence of stable V_K centres is a strong indication that STE diffusion is, indeed, an important scintillation mechanism in $\text{LaCl}_3:\text{Ce}^{3+}$. But also the anti-correlation between STE and Ce^{3+} luminescence intensity, the lifetime of STE luminescence decreasing with increasing Ce concentration, and the evolution of the decay profile of $\text{LaCl}_3:\text{Ce}^{3+}$ with temperature from a power-law dependence to an exponential one support this assumption.

Let us review the data presented thus far. The anti-correlation between STE and Ce^{3+} luminescence intensity as a function of Ce concentration is attributed to a process of *direct capture of electron–hole pairs* and the immediate trapping of some of the STEs by Ce^{3+} . In all samples, the fast decay component is due to the former process: when the Ce concentration is increased, the contribution of the fast decay component to the total light yield is expected to increase as well, because the probability of direct capture of electron–hole pairs by Ce^{3+} increases. Indeed, from $\text{LaCl}_3:0.57\% \text{Ce}^{3+}$ to $\text{LaCl}_3:10\% \text{Ce}^{3+}$, the contribution of this component to the total light yield increases from 8 to 41% (see figure 4). In this case, the decay of excited Ce^{3+} ions is given by

$$n_{\text{Ce}}(t) = n_{\text{Ce}}^0 \exp\left(-\frac{t}{\tau_{\text{Ce}}}\right) \quad (3)$$

where n_{Ce}^0 is the number of excited Ce^{3+} ions at time $t = 0$, and $1/\tau_{\text{Ce}}$ is the radiative decay rate of the Ce^{3+} ions. The decrease of the relative contribution of the STE luminescence to the total light yield with Ce concentration can be ascribed to both direct capture of electron–hole pairs and the immediate trapping of some of the STEs by Ce^{3+} . Indeed, for $\text{LaCl}_3:2\% \text{Ce}^{3+}$ (see figure 2) the contribution of STE luminescence to the total light yield is never to exceed 50%, whereas for $\text{LaCl}_3:10\% \text{Ce}^{3+}$ it is only 10% at best (see figure 3).

The anti-correlation between the STE and Ce^{3+} luminescence intensity as a function of temperature is due to the mobility of the STE. When the temperature is raised, the mobility of the STE increases and the energy is transferred to a Ce^{3+} centre the moment the STE encounters a Ce^{3+} ion. The probability increases with the mobility of the STE and the Ce^{3+} luminescence intensity increases whereas the STE luminescence intensity decreases. It can be observed for both $\text{LaCl}_3:2\% \text{Ce}^{3+}$ (see figure 2) and $\text{LaCl}_3:10\% \text{Ce}^{3+}$ (see figure 3) at temperatures $T > 200$ K. The shortening of the decay time of the STE (see figure 4) is consistent with this model. If the Ce concentration is increased, the diffusion path length for the STE to a Ce^{3+} centre is shortened, as well as the decay time of the STE; see equation (1).

For $\text{LaCl}_3:\text{Ce}^{3+}$ it is assumed that the STE transfers its energy to Ce^{3+} before it decays non-radiatively. This assumption seems reasonable since the activation energy for energy transfer

($\Delta E_t = 23 \pm 2$ meV) is significantly lower than the activation energy for thermal quenching ($\Delta E_q = 106 \pm 6$ meV). In the high-temperature limit, the diffusion of the STE through the lattice can be described by a hopping-like motion from one lattice site to a nearest-neighbour site [11]. The rate of energy transfer from the STE to Ce^{3+} in an isotropic medium is given by [12]

$$\frac{1}{\tau_t} = 4\pi DRN_{\text{Ce}} \quad (4)$$

where D , R , and N_{Ce} are the diffusion constant of the STE, the radial distance from a Ce^{3+} centre within which the STE transfers its energy, and the Ce concentration, respectively. Then, the decay $n_{\text{STE}}(t, x)$ of the STE and $n_{\text{Ce}}(t, x)$ of the excited Ce^{3+} ions are given by

$$n_{\text{STE}}(t, x) = n_{\text{STE}}^0(x) \exp\left\{-\left(\frac{1}{\tau_{Rq}} + \frac{1}{\tau_t}\right)t\right\} \quad (5)$$

$$n_{\text{Ce}}(t, x) = \frac{n_{\text{STE}}^0(x) \frac{1}{\tau_t}}{\frac{1}{\tau_{\text{Ce}}} - \left(\frac{1}{\tau_{Rq}} + \frac{1}{\tau_t}\right)} \left[\exp\left\{-\left(\frac{1}{\tau_{Rq}} + \frac{1}{\tau_t}\right)t\right\} - \exp\left(-\frac{t}{\tau_{\text{Ce}}}\right) \right] + n_{\text{Ce}}^0(x) \exp\left(-\frac{t}{\tau_{\text{Ce}}}\right) \quad (6)$$

where $n_{\text{STE}}^0(x)$ is the number of STEs initially created after absorption of a gamma quantum as a function of Ce concentration at time $t = 0$, and $n_{\text{Ce}}^0(x)$ is the number of excited Ce^{3+} ions as a function of Ce concentration at time $t = 0$. $1/\tau_{Rq}$ is the sum of the first two terms on the right side of equation (1). The total scintillation light yield as a function of time and Ce concentration will be given by the sum of (5) and (6). The first term of (6) is due to the process of STE diffusion which is dependent on both STE and Ce concentration, whereas the second term of (6) is due to direct capture of electron–hole pairs by Ce^{3+} which is governed by the Ce concentration only. If the Ce concentration is increased, $n_{\text{Ce}}^0(x)$ will increase and the second term of (6) will become more important. As a consequence, the relative contribution of the fast decay component to the total light yield increases (see figure 4).

Equations (5) and (6) predict an exponential decay of both STE and Ce^{3+} luminescence and, indeed, at temperatures above 250 K (see figure 6) an exponential scintillation decay is observed. In the past, a diffusion-limited energy transfer has been suggested for NaCl:Li [7] to account for the observed exponential square-root dependence of the luminescence decay on time:

$$I(t) = I_0 \exp(-at - 2b\sqrt{t}) \quad (7)$$

where I_0 , a , and b are constants. However, the decay curves in figure 6 at a temperature, say, lower than 250 K are clearly non-exponential and are not well fitted to equation (7). In a review by Jonscher and de Polignac [13] it was shown that often the luminescence does not decay exponentially but follows a power-law dependence on time with fractional exponent s :

$$I(t) \propto t^{-s} \quad (8)$$

with $0 < s < 2$ [14]. The physical processes underlying the delayed luminescence according to the power law of equation (8) may be very complex and may involve several stages, but in general a power-law dependence between luminescence intensity and time is expected when there is some sort of tunnelling or diffusive motion through or across energy barriers with a random distribution in width or height. At low temperatures, i.e. $T < 250$ K, the energy transfer from the STE to Ce^{3+} is probably governed by one of these models, whereas at higher temperatures the motion of the STE through the lattice is no longer effected by the random distribution in energy barrier properties, and the scintillation decay is described best by an exponential decay, according to equation (6).

5. Conclusions

For low Ce concentrations, V_K centres are created, which immediately recombine with an electron to form a STE. Thermally activated STE diffusion to Ce^{3+} is then the dominant scintillation mechanism. It results in a relatively slow decay component. If the Ce concentration is increased, the probability that a STE transfers its energy to Ce^{3+} increases as well. It is also possible that a V_K centre is trapped near Ce^{3+} before a STE can be formed. Recombination with the electron yields Ce^{3+} emission. However, this process must then be very fast, since there was no evidence of stable V_K or F (electron) centres in the EPR of $\text{LaCl}_3:\text{Ce}^{3+}$. As such, it does not contribute to the elongation of the scintillation decay. In principle, Ce^{3+} could also capture a free electron and hole from the conduction and valence band, respectively, leading to fast Ce^{3+} luminescence. However, the contribution of this component to the scintillation decay is relatively small.

Acknowledgment

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