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Thermal quenching and electron traps in LSO

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Abstract

It is demonstrated by comparison of thermoluminescence and scintillation light outputs of LSO as functions of radiation time that a previously suggested thermal quenching correction is inappropriate. Approximate solutions of rate equations are employed to infer absolute trap concentrations and to explore the effects of thermal quenching on the shapes of thermoluminescence glow curves.

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1. Introduction

LSO ($\text{Ce:Lu}_2\text{SiO}_5$) is a fast scintillator material with high stopping power and high light output. The efficiency of scintillator response, η , is conventionally expressed as the product of three partial efficiencies:

$$\eta = \beta S Q, \quad (1)$$

where β is the conversion efficiency (the ratio of actually produced electron–hole pairs to the maximum possible), S is the transfer efficiency and Q is the quantum efficiency of luminescence. Bartram and Lempicki [1] have demonstrated theoretically that efficient conversion ($\beta \cong 1$) is expected for wide-band gap insulators. Energy storage by deep electron traps was probed by comparison of thermoluminescence and scintilla-

tion light outputs, facilitated by a common apparatus and detection scheme for both measurements [2], to assess the extent to which electron trapping limits transfer efficiency ($S < 1$). This method exploits a common activator with identical emission spectra for both thermoluminescence and scintillation. However, application of the method to LSO is complicated by thermal quenching just above room temperature [3,4].

2. Experimental results

The single crystal samples investigated, LSO-L8 and LSO-S8, were provided by C. Melcher. A sample LSO-H that had been characterized previously [5] served as a calibration standard. Light output and sample temperature for sample LSO-L8 are plotted as functions of time for an irradiation time of 16 min in Fig. 1. The ratio of integrated thermoluminescence light output and integrated scintillation light output (G/S) is plotted

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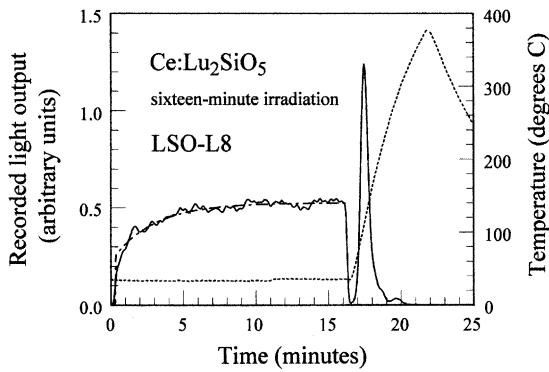


Fig. 1. Recorded (—) and simulated (---) light output and sample temperature (· · ·) for LSO-L8.

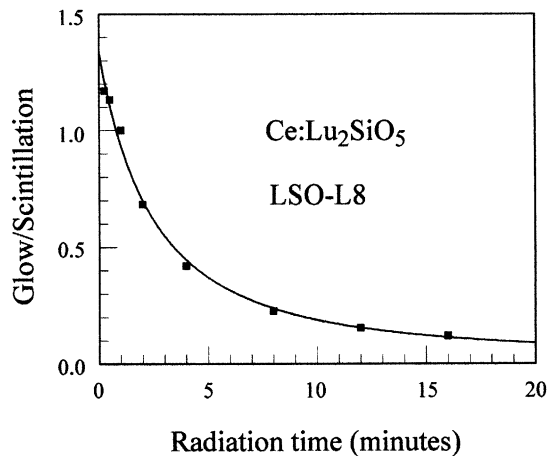


Fig. 2. Recorded G/S vs. radiation time for LSO-L8.

in Fig. 2 and the combined integrated thermoluminescence light output and integrated scintillation light output divided by irradiation time $[(G+S)/t]$ is plotted in Fig. 3, both as functions of irradiation time, for sample LSO-L8.

3. Thermal quenching correction

The quantum efficiency of luminescence Q was evaluated by Dorenbos et al. [3] for LSO in the form

$$Q = \frac{1}{1 + 34\,000 \exp(-3700/T)}, \quad (2)$$

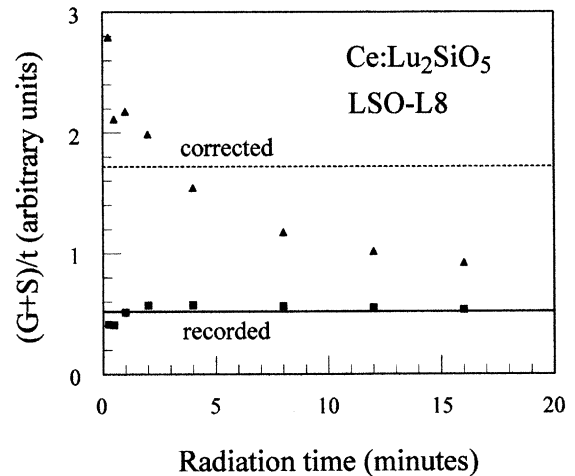


Fig. 3. Recorded and corrected $(G+S)/t$ vs. radiation time for LSO-L8.

where T is the temperature on the Kelvin scale. These investigators subsequently corrected recorded thermoluminescence glow curves for the effects of thermal quenching by dividing them by Q . However, the latter application depends on the tacit assumption that the mechanism of thermal quenching involves a competing process of radiationless recombination at cerium activators such as a multi-phonon radiationless transition to the cerium ground state. In a subsequent investigation of thermal quenching in cerium-doped crystals, Yen et al. [4] demonstrated convincingly by photoconductivity measurements that the thermal quenching mechanism in LSO is thermal ionization from the lowest 5d excited state to the conduction band, rather than a multi-phonon radiationless transition to the 4f ground state. The ionized electron may continue to contribute to thermoluminescence by a multiple re-trapping mechanism, thus obviating the quenching correction. In order to test its validity, the quenching correction is applied to $(G+S)/t$ in Fig. 3. Since $(G+S)/t$ should be constant when all electrons are accounted for, a criterion satisfied by the recorded data but not by the corrected data, it is concluded that the quenching correction is inappropriate in the present context.

4. Rate equations

Following the production of thermalized electron–hole pairs by incident gamma rays [1], the fundamental processes involved in subsequent stages of scintillation and thermoluminescence can be described by a set of rate equations [2,6,7], modified to accommodate thermal ionization [4].

$$\frac{dn_c}{dt} = f - n_c n_h A_r - n_c(N - n)A + np + n_a q, \quad (3a)$$

$$\frac{dn}{dt} = n_c(N - n)A - np, \quad (3b)$$

$$\frac{dn_h}{dt} = f - n_c n_h A_r + n_a q, \quad (3c)$$

$$\frac{dn_a}{dt} = n_c n_h A_r - n_a \tau_r^{-1} - n_a q, \quad (3d)$$

where f is the rate of production of electron–hole pairs per unit volume, N is the concentration of deep electron traps, n_c and n are the respective concentrations of conduction electrons and trapped electrons, n_h is the concentration of trapped holes (Ce^{4+}) and n_a is the concentration of excited activators (Ce^{3+*}). A single species of deep electron trap is assumed for simplicity. The coefficient A_r governs the rate of recombination of conduction electrons with trapped holes, A governs the rate of trapping of conduction electrons at deep electron traps, p is the rate of thermal ionization of deep electron traps, and q and τ_r are, respectively, the rate of thermal ionization and the radiative lifetime of excited activators. The trapping of valence-band holes on Ce^{3+} ions is assumed to be instantaneous. Rates p and q are assumed to be thermally activated.

Eqs. (3) can be simplified by neglecting dn_c/dt and dn_a/dt and by adopting the approximations $n_h \cong n_{h0} + n$ and $A \cong A_r$, where n_{h0} is the initial concentration of trapped holes (Ce^{4+}). It is convenient to employ reduced variables defined by

$$\begin{aligned} \tilde{n} &\equiv n/N, \quad \gamma \equiv f/N, \quad \alpha \equiv NA/n_{h0}A_r \cong N/n_{h0}, \\ \tilde{I} &\equiv I/N. \end{aligned} \quad (4)$$

We are then left with the single differential equation:

$$\frac{d\tilde{n}}{dt} = \frac{(\gamma + \tilde{n}p)(1 - \tilde{n})}{\alpha^{-1}Q[1 + \alpha\tilde{n}] + (1 - \tilde{n})} - \tilde{n}p, \quad (5)$$

$$Q = \frac{\tau_r^{-1}}{\tau_r^{-1} + q} \quad (6)$$

and the reduced light output per unit volume, \tilde{I} , is given by

$$\tilde{I} = Q \left\{ \frac{(\gamma + \tilde{n}p)[1 + \alpha\tilde{n}]}{Q[1 + \alpha\tilde{n}] + \alpha(1 - \tilde{n})} \right\}. \quad (7)$$

5. Scintillation phase

The rate f of electron–hole pair production is maintained at a constant value during the scintillation phase, and the thermal ionization rates are neglected. Eq. (5) is then reduced to a linear equation with the solution

$$\tilde{n} = 1 - \exp(-\beta t), \quad (8a)$$

$$\beta \equiv \frac{\alpha\gamma}{1 + \alpha}. \quad (8b)$$

It then follows from Eq. (7) that the light output is given by

$$\tilde{I} = \gamma - \beta \exp(-\beta t). \quad (9)$$

The ratio G/S predicted by the present model is

$$\frac{G}{S} = \frac{\tilde{n}}{\gamma t - \tilde{n}} = \frac{1 - \exp(-\beta t)}{\gamma t - 1 + \exp(-\beta t)}. \quad (10)$$

This quantity is plotted in Fig. 2 with parameters α and γ adjusted for a least-squares fit to recorded data for LSO-L8. The dash-dot curve in Fig. 1 is proportional to Eq. (9) with the same parameter values. Optimized parameters are listed in Table 1 together with values of N and n_{h0} derived from them and from the rate of electron–hole pair production, $f \cong 6 \times 10^{16} \text{cm}^{-3} \text{min}^{-1}$. Also listed in Table 1 are both measured and estimated values of efficiency η , defined as the prompt scintillation light output divided by the theoretical number of electron–hole pairs/MeV, 69,444 [1], and the radiative fraction of recombination events $\varepsilon \geq \eta(1 + \alpha)$. The inequality accommodates scintillation delayed by shallow traps [5,6],

Table 1
Optimized parameters, concentrations and efficiencies

Sample	α	γ (min ⁻¹)	N (cm ⁻³)	n_{h0} (cm ⁻³)	η (%)	ε (%)
LSO-L8	1.34	0.61	1×10^{17}	7×10^{16}	≤ 7 –12	16–28
LSO-S8	0.128	25.1	2×10^{15}	2×10^{16}	≤ 22 –37	25–42
LSO-H	0.086	1.64	4×10^{16}	4×10^{17}	36 ^a	39

^a measured.

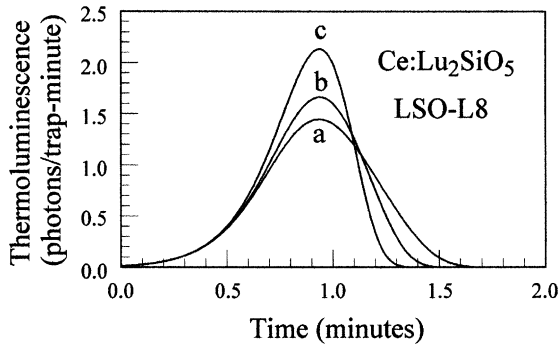


Fig. 4. Simulated glow curves: (a) thermal ionization and re-trapping, (b) re-trapping only ($Q=1$), (c) first-order kinetics ($Q=1$, $\alpha=0$).

Table 2
Assumed parameter values for Fig. 4

s_d (min ⁻¹)	E_d (K)	ramp (K/min)
4.3×10^{14}	11 400	60

indistinguishable from prompt scintillation in this experiment.

6. Thermoluminescence phase

There is no electron–hole pair production during the thermoluminescence phase, but the thermal ionization rates increase with temperature. Thermoluminescence is governed by Eqs. (5)–(7) with $\gamma = 0$. A linear temperature ramp is assumed over the range of interest and the equations are solved by numerical integration with initial condi-

tion $\tilde{n}(0) = 1$. Simulated glow curves for sample LSO-L8 are plotted in Fig. 4 for three cases with the typical parameter values listed in Table 2.

7. Discussion

A thermal quenching correction [3] is shown to be inappropriate for the present experiment. Thermal ionization [4] and re-trapping modify simulated glow curves. Electron traps reduce the efficiency of prompt scintillation. Traps may be associated with oxygen vacancies [8]. The saturation level of scintillation is not correlated with trap concentration but energy transfer to radiationless recombination centers [9] may further reduce light output.

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