Thermoluminescence and Scintillation Properties of Rare Earth Oxyorthosilicate Scintillators

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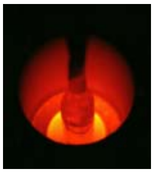
Abstract In recent years the scintillation properties of several cerium-doped rare earth oxyorthosilicate scintillators, Ln_2SiO_5:Ce where Ln = Y, La - Lu, have been reported and, in some cases, extensively studied. In addition, binary and ternary compounds such as $(Lu,Y)_2SiO_5$:Ce, $(Lu,Gd)_2SiO_5$ and $(Lu,Y,Gd)_2SiO_5$:Ce have been reported. All of these crystals have either monoclinic P or C structures with characteristic SiO_4 tetrahedra and trivalent cations occupying two unique crystallographic positions. The trivalent cerium activator ions are assumed to occupy the cation lattice sites and possibly interstitial positions as well. The excited 5d state of Ce^{3+} is split into 3 observable levels with luminescence emission occurring only from the lowest 5d level to the 4f ground state (~ 3 eV) with a Stokes shift of ~ 0.5 eV. The band gap is about 6 eV, and the index of refraction is close to 1.8, with some variation according to crystallographic axes. Despite these similarities, important differences remain among the crystals including scintillation efficiency, decay time, rise time, and afterglow. In this paper, we report thermoluminescence measurements between 10K and 350K that allow the determination of trapping levels that may influence scintillation properties.

Experimental procedures



We used the Czochralski technique to grow cerium-doped single crystal boules of **LSO, LYSO, LGSO and LYGSO**. The boules were grown from inductively heated iridium crucibles in a nitrogen atmosphere. The Lu_2O_3, Y_2O_3, Gd_2O_3, SiO_2, and CeO_2 starting materials were at least 99.99% pure.

For measurements of the **scintillation light output**, the crystal was stored in the dark for at least 24 hours and then placed directly onto a PMT Hamamatsu R877 without any optical coupling materials. The crystal was covered with a loose fitting Teflon cap to enhance the light collection efficiency. The crystal was excited with 662 keV gamma rays from a 10 µCi ^{137}Cs. Light output was calculated relative to a BGO crystal measured in the same geometry.

Scintillation decay time was measured using the time-correlated single photon technique originated by Bollinger and Thomas.

For **thermoluminescence glow curve measurements** the crystal was irradiated with x-rays in a vacuum chamber (~15min) at 10K, and then heated at a constant rate of β=0.16 K/s. A Hamamatsu R2059 PMT was used to measure the luminescence emitted by the crystal as a function of temperature.

The same setup was used to measure the **temperature dependence of the scintillation light output**, with the exception that the crystal was irradiated with gamma rays from a 10 µCi ^{137}Cs source and the energy spectrum was measured with a Canberra System100 multichannel analyzer.

Results

TABLE I. COMPARISON OF SCINTILLATION PROPERTIES OF RARE EARTH OXYORTOSILICATES

Sample	*Light output [%BGO]*	*Decay time [ns]*
LSO	464	40.4
LYSO (70% Y)	395	50.1
LGSO 50%Gd)	319	39.3, 415
LYGSO (15%Y,15% Gd)	232	37.6, 261

The light output was calculated relative to BGO and corrected in accordance with the quantum efficiency of the PMT used in experiments (QE = 19% for BGO, 24% for LSO). Decay times were obtained from fitting to experimental points.

Fig. 1 Scintillation time profiles

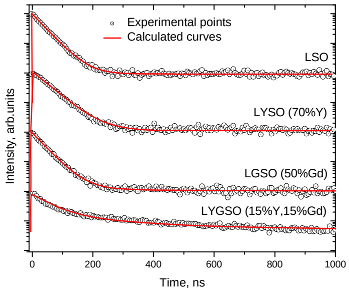


Fig. 2 Thermoluminescence glow curves

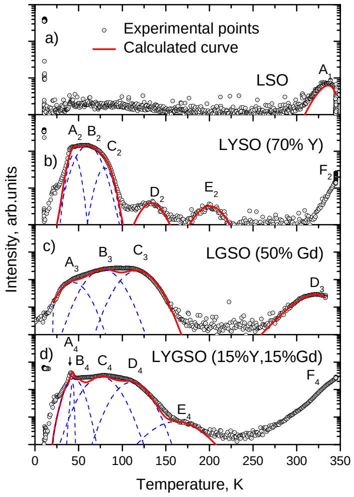
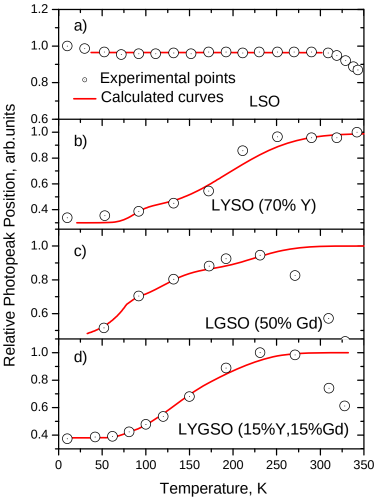


Fig. 3 Thermoluminescence glow curves



Discussion

Thermoluminescence is a useful tool for studying the energy transfer or, specifically, the role of charge carriers in the scintillation process, which can be carried out on crystals with a variety of compositions. It is generally accepted that deep traps, revealed by high-temperature thermoluminescence (measured at temperatures higher than 300K), can reduce the scintillation light yield and that shallow traps are usually responsible for slow components in scintillation decays.

In our model we assumed: (i) that broad bands in thermoluminescence glow curves measured for LYSO, LGSO and LYGSO crystals are caused by three to five trap distributions in place of single traps, and (ii) the traps within the distribution do not interfere with each other. Consequently, the glow curves can be described as a linear combination of traps of different activation energies and concentrations, the last determined by a distribution function. For our purpose we chose a Gaussian function. Thus in general the traps are described by three parameters: *E*, *s* and *w*. *E* is the thermal activation energy at the center of Gaussian distribution of traps. The frequency factor *s* is the same for all traps in the distribution. The FWHM is *w*. The linear combination is achieved by integration of all the equations (1-4) over the appropriate energy range.

(1) Randall and Wilkins equation:

$$\mathrm{I}(\mathrm{T}-\Delta\mathrm{T}) = ns\exp\left(-\frac{E}{kT}\right)\exp\left[-\left(\frac{s}{\beta}\right)\int_0^T \exp\left(-\frac{E}{kT'}\right)dT'\right]$$

(2) The carrier's lifetime in the trap level :

$$\tau_t = \frac{1}{s}\exp\left(\frac{E}{kT}\right)$$

(3) The scintillation time profile:

$$I(t) = \frac{n}{\tau_{Ce}-\tau_t}\left[\exp\left(-\frac{t}{\tau_{Ce}}\right)-\exp\left(-\frac{t}{\tau_t}\right)\right]$$

(4) The thermal dependence of scintillation efficiency:

$$I_\int(T) \approx n\frac{\tau_{Ce}}{\tau_{Ce}-\tau_t}\left\{1+\frac{\tau_t}{\tau_{Ce}}\left[\exp\left(-\frac{t_{col}}{\tau_t}\right)-1\right]\right\}$$

Where τ_{Ce} radiative decay of the cerium emission, *n* is the trap concentration, t_{col} is the collection time, *k* is the Boltzmann constant.

Using data solely from the thermoluminescence glow curves generates too many possible values for trap parameters. In order to arrive at a unique solution we used the thermoluminescence data coupled with temperature dependent scintillation efficiency data. Simultaneously analyzing results obtained from the two different experiments. The set of parameters *E*, *s*, and *w* that give the best match of the calculated curves and the experimental points of the thermoluminescence glow curve and temperature dependence of scintillation efficiency data are assumed to be a good approximation.
Simulated glow curves were calculated in accordance with the model described above for LYSO, LGSO and LYGSO as shown in the Fig. 2 and 3 (solid lines) Based on the same trap parameters the scintillation time profiles presented in Fig 1 were calculated. Agreement between theoretical and experimental points is very good.

TABLE II. TRAP PARAMETERS OBTAIN AS A RESULT OF SIMULATION

Sample	*Trap*	*E [eV]*	*Lns [s^{-1}]*	*w*
LSO	A_1	0.92	27.54	0
LYSO (70% Y)	A_2	0.1	24.64	0.012
	B_2	0.11	17.5	0.02
	C_2	0.15	18.42	0.012
	D_2	0.36	27.51	0.03
	E_2	0.54	27.58	0.035
LGSO 50%Gd)	A_3	0.08	20.03	0.025
	B_3	0.23	32.93	0.045
	C_3	0.29	26.05	0.038
	D_3	0.51	13.66	0
LYGSO (15%Y,15% Gd)	A_4	0.1	27.53	0.001
	B_4	0.08	20.03	0.015
	C_4	0.18	25.33	0.035
	D_4	0.27	28.81	0.035
	E_4	0.49	32.24	0.069