# Effect of Electron Traps on Scintillation of Praseodymium Activated Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>

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Abstract—In this paper we present the studies performed on a set of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr (LuAG:Pr) crystals with praseodymium concentration between 1.5 and 10%, grown by the micro-pulling-down ( $\mu$ PD) technique. The research comprises the measurements of X-ray excited emission spectra and <sup>137</sup>Cs gamma-ray pulse height spectra in a range from 78 to 600 K, and thermoluminescence glow curves. Based on experimental data we discuss the dependence of scintillation properties of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr on praseodymium content and temperature. The main attention is focused on a distinct increase of scintillation yield with temperature, which we attribute to existence of shallow electron traps and their temperature-dependent contribution to scintillation of LuAG:Pr. An active role of traps is demonstrated by a novel experiment combining X-ray and laser excitation.

Index Terms—LuAG:Pr, light yield, scintillation mechanism, trap.

#### I. INTRODUCTION

IVERSE rare earth activated wide band gap oxide crystals have been acquiring an increasing interest in recent years as potential detectors of ionizing radiation in nuclear and high-energy physics, space exploration, nuclear medicine, and industry. Among many materials studied, praseodymium activated lutetium aluminum garnet, Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr (LuAG:Pr), seems to be one of the most promising ones because of its high density of 6.7 g/cm<sup>3</sup>, very good energy resolution of 4.6%, and fast scintillation decay time of 20 ns [1]. Currently efforts are being made to optimize LuAG:Pr, mainly by improving the growth technology [2]-[10]. The scintillation mechanism of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr has also attracted some attention. Yoshikawa et al. [11] have suggested that there are two types of energy transfer from the host to the  $Pr^{3+}$  ions: a fast process due to a prompt migration of excitons to  $Pr^{3+}$  and a slow process due to trapping and detrapping of electrons and/or holes, most probably at so-called antisite defects. An alternative view has been presented by Drozdowski et al. [1]. They have attributed

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the fast 20 ns scintillation component to capture of valence band holes  $h_V$  at  $Pr^{3+}$  ions  $(Pr^{3+}+h_V\rightarrow Pr^{4+})$ , followed by capture of conduction band electron  $e_C$  and creation of excited states of  $Pr^{3+}(Pr^{4+}+e_C\rightarrow (Pr^{3+})^*)$ . An excitonic transfer has been considered as responsible for presence of slow components in scintillation time profiles of LuAG:Pr.

In the current work we have investigated a series of new  $Lu_3Al_5O_{12}$ :Pr crystals with praseodymium concentration between 1.5 and 10%. Beside a characterization by means of pulse height and X-ray excited emission spectra, we have focused on temperature-dependent studies in order to improve our knowledge on the scintillation mechanism of LuAG:Pr. The acquired data are interpreted quantitatively within the framework of a simple model based on the aforementioned consecutive capture of charge carriers at  $Pr^{3+}$  ions, including a possibility of trapping of electrons. A good agreement among the results from different experimental techniques and model predictions supports an important role of shallow electron traps in scintillation of  $Lu_3Al_5O_{12}$ :Pr.

### **II. MATERIALS AND EXPERIMENT**

Single crystals of  $(Lu_{1-x}Pr_x)_3Al_5O_{12}$  (x = 0.015, 0.021, 0.03, 0.05, 0.1) were grown at Pisa University by the micropulling-down ( $\mu$ PD) method. Lu<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub> and Pr<sub>6</sub>O<sub>11</sub> powders of 99.999% purity were the starting materials. After the desired quantities were carefully weighed and mixed, the growth procedures described by Alshourbagy *et al.* [12] were applied. Transparent, crack-free, yellow-green crystal fibers with no visible inclusions were obtained (Fig. 1). Each fiber was then cut into several samples and polished with alumina and diamond powders. The dimensions of the prepared samples are listed in Table I. The real concentrations of  $Pr^{3+}$  ions are not known. However, they can be expected to be much closer to the nominal ones than in case of the Czochralski method, in which the so-called segregation coefficient is below 0.1 [4], because of the 100% solidification in the  $\mu$ PD technology [12], [13].

Room temperature (RT) pulse height spectra were collected under 662 keV gamma excitation from a <sup>137</sup>Cs source. The samples were placed with their fiber axis horizontally on the quartz window of a Hamamatsu R1791 photomultiplier tube (PMT) and covered with several layers of Teflon tape in a configuration of a reflective "umbrella". The output signal from the PMT, supplied with a high voltage of 500 V, was processed by a home-made preamplifier, an Ortec 672 spectroscopy amplifier, an Ortec AD114 analog-to-digital converter, and a multichannel analyzer. From the channel position of the 662 keV photopeak in a pulse height spectrum and the mean of the single photoelectron response peak, the corresponding scintillation yield ex-

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Fig. 1. A LuAG:2.1%Pr single crystal grown by the  $\mu$ PD technique.

TABLE I DIMENSIONS OF THE STUDIED SAMPLES

Nominal Pr <sup>3+</sup> concentration (%)	Length (mm)	Diameter (mm)
1.5	6.6	2.8
2.1	5.5	2.7
3.0	6.7	2.6
5.0	5.8	2.7
10.0	6.5	3.0

pressed as the number of photoelectrons from the PMT photocathode detected per unit of energy deposited in the crystal was obtained. To provide compatibility with our previous measurements on LuAG:Pr [1], the shaping time was set at 3  $\mu$ s.

Pulse height spectra were also studied as a function of temperature with a technique described by Bizarri *et al.* [14]. The crystals were kept in clean vacuum inside a Janis cryostat and excited by a <sup>137</sup>Cs source. In these measurements two shaping times, 3 and 10  $\mu$ s, were used.

A typical setup consisting of a Philips PW2253 X-ray tube with a Cu anode, operated at 55 kV and 35 mA, an Acton Research Corporation VM504 monochromator, a Hamamatsu R943-02 photomultiplier, a Janis cryostat, and a LakeShore 331 programmable temperature controller, was employed to record X-ray excited emission spectra at temperatures between 78 and 600 K.

A novel method developed by Poolton et al. [15] was utilized to study the role of shallow electron traps in scintillation of LuAG:Pr. In this experiment the luminescence of the crystal during separate or simultaneous X-ray and infrared laser excitation was monitored. Both sources, a Philips PW2253 X-ray tube and a 980 nm laser diode, were operated independently. The former provided ionizing excitation, whereas the latter was releasing electrons from shallow traps back to the conduction band. The detection was carried out at temperatures between 10 and 300 K with the Mobile Luminescence End-Station (MoLES) [16]. To limit the area of interest to the fast  $Pr^{3+}4f5d \rightarrow 4f^2$  emission, a Schott UG2 colour glass filter with a transmission maximum at 340 nm (FWHM  $\sim 70$  nm) was placed in front of the PMT. This setup was also used to measure thermoluminescence glow curves at a heating rate of 0.15 K/s.

## **III. RESULTS AND DISCUSSION**

An example of a pulse height spectrum of  $Lu_3Al_5O_{12}$ :Pr is shown in Fig. 2. The values of photoelectron yield and energy resolution (at 662 keV) obtained for all crystals are summarized



Fig. 2. A <sup>137</sup>Cs pulse height spectrum of LuAG:1.5%Pr. The data symbols come from the experiment; the solid curve is a Gaussian fit.

TABLE II PHOTOELECTRON YIELD Y and Energy Resolution R of LuAG:PR as Functions of Praseodymium Concentration

Nominal Pr <sup>3+</sup> concentration (%)	Y (phe/MeV)	<i>R</i> (%) at 662 keV
1.5	$4600 \pm 230$	$7.0 \pm 0.4$
2.1	$3970 \pm 200$	$8.4 \pm 0.4$
3.0	$3330 \pm 170$	$14.1 \pm 0.7$
5.0	$2930 \pm 150$	$13.0 \pm 0.7$
10.0	$2880 \pm 150$	$12.0 \pm 0.6$

in Table II and Fig. 3. The yield clearly deteriorates with increasing concentration of Pr<sup>3+</sup> ions. The best resolution is displayed by the two samples with the lowest concentration. These results can be used for a rough estimation of the real  $Pr^{3+}$  content in the investigated crystals. According to Ogino *et al.* [4] the yield of LuAG:Pr first increases with concentration, peaks at 0.2–0.3% Pr, and then decreases. Based on experimental data and "2R" model calculations reported for Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr samples with a measured concentration of 0.23%Pr [1], a highquality, polished, 2.8 mm high crystal of LuAG:0.23%Pr is anticipated to display a yield of  $6270 \pm 320$  phe/MeV. Including such assumption as an extra point in Fig. 3, we get a decrease of yield as a function of Pr<sup>3+</sup> content, which agrees with the conclusions of Ogino et al. [4]. Since we deal with transparent, crack-free, polished samples, we do not expect significant losses related to imperfect material quality. Hence it seems that the real concentrations of  $Pr^{3+}$  ions in the studied  $\mu PD$ -grown crystals



Fig. 3. Room temperature photoelectron yield of LuAG:Pr as a function of praseodymium concentration. A value expected for a high-quality, polished, 2.8 mm high LuAG:0.23%Pr crystal is also included.



Fig. 4. Room temperature X-ray excited emission spectra of LuAG:Pr with three different praseodymium concentrations, normalized to the same intensity in the  $4f5d \rightarrow 4f^2$  band around 310 nm.

are indeed quite close to the nominal ones, which is a real advantage over the Czochralski method.

Fig. 4. compares room temperature X-ray excited emission spectra of three Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr samples with different Pr<sup>3+</sup> contents. Two spectral regions can be distinguished: the fast  $Pr^{3+}4f5d \rightarrow 4f^2$  luminescence between 290 and 450 nm, and the slow  $Pr^{3+}4f^2 \rightarrow 4f^2$  luminescence above 450 nm. The emission lines of the latter have been identified and are listed in Table III. The three spectra turn out to be quite similar, nevertheless two subtle features are worth noting. In the  $4f5d \rightarrow 4f^2$ range of the emission there is a shift of the leading edge on the high-energy side of the 310 nm band towards longer wavelengths, accompanied by a relative increase of the 365 nm band intensity. This effect is attributed to increasing self-absorption with  $Pr^{3+}$  concentration. In the  $4f^2 \rightarrow 4f^2$  region of the emission a strong concentration quenching of the 610 nm<sup>1</sup>D<sub>2</sub>  $\rightarrow$ <sup>3</sup>H<sub>4</sub>

TABLE III  $4f^2 \rightarrow 4f^2 \text{ Transitions Identified in Radioluminescence of LuAG:Pr}$ 

V	Vavelength (nm)	Transition
4	75	$^{3}P_{1} \rightarrow ^{3}H_{4}$
43	88	$^{3}P_{0} \rightarrow ^{3}H_{4}$
50	02	$^{3}P_{2} \rightarrow ^{3}H_{5}$
5.	33	$^{3}P_{1} \rightarrow ^{3}H_{5}$
5	65	$^{3}P_{2} \rightarrow ^{3}H_{6}$
59	97	$^{3}P_{1} \rightarrow ^{3}H_{6}$
6	10	$^{1}\text{D}_{2} \rightarrow {}^{3}\text{H}_{4}$
6	17	$^{3}P_{0} \rightarrow ^{3}H_{6}$
62	21	${}^{3}P_{1} \rightarrow {}^{3}F_{2}$
6.	32	${}^{3}P_{2} \rightarrow {}^{3}F_{3}$
6.	39	${}^{3}P_{0} \rightarrow {}^{3}F_{2}$
6	53	${}^{3}P_{2} \rightarrow {}^{3}F_{4}$
6	86	${}^{3}P_{1} \rightarrow {}^{3}F_{3}$
70	05	$^{1}\text{D}_{2} \rightarrow {}^{3}\text{H}_{5}$
7	16	${}^{3}\mathrm{P}_{1} \rightarrow {}^{3}\mathrm{F}_{4}$
74	47	$^{3}P_{0} \rightarrow ^{3}F_{4}$
82	28	$^{1}\text{D}_{2} \rightarrow {}^{3}\text{H}_{6}$



Fig. 5. X-ray excited emission spectra of LuAG:3%Pr at 78, 300, and 600 K.

line takes place, which is caused by cross-relaxation processes [17]. Anyway, the influence of praseodymium concentration between 1.5%Pr and 10%Pr on radioluminescence spectra of LuAG:Pr is not large. The two remaining crystals, containing 2.1%Pr and 5%Pr, account for intermediate cases and have not been included in Fig. 4 for clarity.

Fig. 5 presents X-ray excited emission spectra of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:3%Pr, recorded at 78, 300, and 600 K. They indicate clearly that both total intensity and relative contributions from the  $Pr^{3+}4f5d \rightarrow 4f^2$  and  $4f^2 \rightarrow 4f^2$  transitions vary with temperature. A more detailed analysis, embracing a set of 18 spectra taken between 78 and 600 K, is displayed in Fig. 6. The intensity integrated in the entire 280-880 nm range is regarded as a total radioluminescence yield, whereas the contributions from the  $4f5d \rightarrow 4f^2$  and  $4f^2 \rightarrow 4f^2$  luminescence are determined by integrals in the 280-450 nm and 450-880 nm regions, respectively. At 78 K both types of the  $Pr^{3+}$  emission contribute to the total yield almost equally. The contribution from the fast  $4f5d \rightarrow 4f^2$  luminescence



Fig. 6. Intensity of radioluminescence of LuAG:3%Pr, integrated between 280 and 880 nm, and contributions from the  $4f5d \rightarrow 4f^2$  (280–450 nm) and  $4f^2 \rightarrow 4f^2$  (450–880 nm) emissions, as functions of temperature. The total yield is normalized to 1 in maximum.

significantly goes up with temperature to 300 K, increasing its intensity by a factor of 2. Simultaneously the intensity of the slow  $4f^2 \rightarrow 4f^2$  luminescence remains nearly constant up to 175 K, whereby it starts decreasing. The total radioluminescence yield compiles these features, resulting in an increase by 20% between 78 and 250 K, and an almost constant value up to 325 K. Above 325 K both types of the Pr<sup>3+</sup> emissions decrease their intensities with temperature, causing a distinct drop of the total yield. Similar tendencies have been observed for other praseodymium concentrations. We note that the measurements have been carried out starting at 600 K and terminating at 78 K to avoid a possible contribution to the emission from thermal release of charge carriers.

The scintillation yield of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr has been studied as a function of temperature by means of <sup>137</sup>Cs pulse height spectra with a shaping time of 3 or 10  $\mu$ s. Since these values of shaping time limit the detection mostly to the fast  $4f5d \rightarrow 4f^2$  luminescence, the scintillation yield may reveal somewhat different temperature-dependent features than the radioluminescence yield. Fig. 7 shows the results obtained for LuAG:1.5% and LuAG:3%Pr with a shaping time of 10  $\mu$ s. The increase of the scintillation yield between 200 and 400 K by a factor of 2 clearly corresponds to the increase of the  $4f5d \rightarrow 4f^2$  emission intensity in Fig. 6. In case of the latter, however, such increase appears between 100 and 300 K. This shift is supposed to be related to the use of shaping time in scintillation measurements and will be discussed later on. Above 400 K the curves recorded with the two alternative techniques resemble each other, i.e., the yield goes down rapidly, with a loss of 50% at 600 K due to a thermal quenching of the  $4f5d \rightarrow 4f^2$  emission.

In order to understand the mechanism behind the increase of scintillation or radioluminescence yield in a specific temperature range, we employ the so-called single-trap model, which has been successfully applied recently for YAlO<sub>3</sub> :Ce, LuAlO<sub>3</sub>:Ce, and BaF<sub>2</sub>:Ce [18]–[21]. Adapting this model to



Fig. 7. Scintillation yield of LuAG:1.5% and LuAG:3%Pr as a function of temperature, normalized to 1 in maxima. The data symbols represent the values of yield derived from pulse height spectra recorded with a shaping time of  $10 \mu s$ ; the solid curves are fits based on the single-trap model.

 $Lu_3Al_5O_{12}$ :Pr, we assume that the prompt consecutive capture of charge carriers, followed by their radiative recombination at  $Pr^{3+}$  ions, constitutes the main route for the host-to-ion energy transfer in this material. This route is responsible for

 TABLE IV

 PARAMETERS DERIVED FROM THE SINGLE-TRAP MODEL FITS

Sample	$\tau_{sh}$ (µs)	а	<i>E</i> (eV)	ln s	T <sub>max</sub> (K) at 0.15 K/s
LuAG:1.5%Pr	10	$0.520 \pm 0.016$	$0.150 \pm 0.037$	17.1 ± 1.6	85
LuAG:3%Pr	10	$0.445 \pm 0.039$	$0.145 \pm 0.068$	$16.5 \pm 2.8$	85
LuAG:10%Pr	3	$0.433 \pm 0.004$	$0.130 \pm 0.005$	$17.0 \pm 0.2$	75
LuAG:10%Pr	10	$0.435 \pm 0.005$	$0.119 \pm 0.007$	$15.6 \pm 0.3$	73
LuAG:0.23%Pr [1]	3	$0.402 \pm 0.010$	$0.163 \pm 0.014$	$17.8 \pm 0.5$	89

the direct 20 ns scintillation component and the presence of the  $4f5d \rightarrow 4f^2$  (possibly also  $4f^2 \rightarrow 4f^2$ ) transitions in the X-ray excited spectra. Besides this there is a delayed, trap-mediated route for the energy transfer due to participation of some unintended charge carrier traps. Since the  $Pr^{3+}$  ions are likely to capture valence band holes promptly and efficiently [22], we suppose that these trapping centers are electron traps. The trap lifetime  $\tau$ , i.e., the mean time spent by a captured electron in such trap, is described by the classic Arrhenius formula:

$$\tau = \frac{1}{s} e^{\frac{E}{k_B T}} \tag{1}$$

where E is the trap depth, *s*—the frequency factor, T—the temperature, and  $k_B$ —the Boltzmann constant. At sufficiently low temperature the trap lifetime is longer than the shaping time in pulse height measurements or even the recording time of a radioluminescence spectrum. In both cases the trapped electrons do not contribute to the luminescence. The higher the temperature, the shorter the trap lifetime, hence the number of electrons that are trapped decreases with temperature. Consequently a corresponding increase of yield is observed. At elevated temperatures the effect of traps becomes negligible and a maximal available yield is displayed, unless there is some thermal quenching of the  $Pr^{3+}$  emission.

The single-trap model can also be described quantitatively by a set of kinetic equations [18]. The solution of these equations expresses the dependence of the scintillation yield Y on the trap lifetime  $\tau$ :

$$Y = Y_0 \left\{ a + b \frac{\tau_{\rm rad}}{\tau_{\rm rad} - \tau} \left[ 1 + \frac{\tau}{\tau_{\rm rad}} \left( e^{-\frac{2.35\tau_{\rm sh}}{\tau}} - 1 \right) \right] \right\}$$
(2)

where  $Y_0$  is the yield of a trap-free material, a and b are the relative contributions from the direct and trap-mediated scintillation components, respectively (a + b = 1),  $\tau_{rad}$  is the radiative lifetime of the  $Pr^{3+}$  ions, and  $\tau_{sh}$ —the shaping time. In accordance with (2) the fraction a of the electrons released by ionizing radiation directly recombines with holes at  $Pr^{3+}$  ions, while the fraction b = 1 - a is captured by the traps and undergoes the trap-mediated route. Thus the  $aY_0$  level of the scintillation yield is preserved at any temperature, whereas the contribution from  $bY_0$  strongly depends on the trap lifetime and, following (1), on temperature. At low temperatures the yield is decreased to  $aY_0$ , because the trap-mediated component  $bY_0$  is completely stored in the traps due to their very long lifetime. At high temperatures the short lifetime makes the traps ineffective and the entire yield of  $(a + b)Y_0 = Y_0$  is observed.

The solid curves in Fig. 7 result from fitting (2) throughout the data between 78 and 400 K, wherein  $\tau_{\rm rad} = 20$  ns [1],  $\tau_{\rm sh} = 10 \ \mu s$ , and  $Y_0 = 1$  are given constants, whilst a, E, and s are fit parameters. The values derived from fitting are summarized in Table IV. They predict the existence of shallow electron traps characterized by a depth of E = 0.145 - 0.150 eV and a frequency factor of  $s = (1.5-2.7) \cdot 10^7$  Hz. The relative contribution of the direct component is estimated as a = 0.45 - 0.52. Consequently b = 1 - a = 0.48 - 0.55 of electrons are captured by the traps, the role of which becomes negligible only above 400 K. Therefore the room temperature scintillation yield of LuAG:1.5% and LuAG:3%Pr is still affected by the traps, reaching about 85% of  $Y_0$  in a measurement with a shaping time of 10  $\mu$ s.

Based on (2) a clear dependence of the scintillation yield on the shaping time is expected in the temperature range, in which the traps play an important role. To verify this, we have recorded pulse height spectra of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:10%Pr as a function of temperature, using two values of that parameter, i.e., 3 and 10  $\mu$ s. The results illustrated in Fig. 8 indicate that, in agreement with expectations, the 3  $\mu$ s curve is shifted to higher temperatures compared to the 10  $\mu$ s one. The room temperature yield of LuAG:10%Pr is thereby decreased by 20% and 15% against Y<sub>0</sub> when measured with a shaping time of 3 and 10  $\mu$ s, respectively. The predicted trap depth of E = 0.119 - 0.130 eV is somewhat shallower than in LuAG:1.5% and LuAG:3%Pr, but the relative contribution of the direct component, a = 0.43, is similar. Probably the nature of the traps is the same in these three materials.

Fig. 9 presents another example of temperature-dependent variations of scintillation yield. The data points have been taken from our previous study on Czochralski-grown LuAG:0.23%Pr crystals [1]. The single-trap model provides the following parameters: E = 0.163 eV,  $s = 5.4 \cdot 10^7 \text{ Hz}$ , and a = 0.40. Since they are close to the ones obtained for the  $\mu$ PD-grown samples, it seems that the existence of ~0.15 eV electron traps is a genuine feature of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr. The trap depth decreases slightly with increasing Pr<sup>3+</sup> concentration, but the main effect of the traps remains the same: the room temperature scintillation yield of LuAG:Pr is about 20% lower than it could be achieved if these traps were absent.

Thermoluminescence (TL) is a powerful technique for investigation of possible presence and distribution of traps in any material. A resultant glow curve, i.e., luminescence as a function of temperature, can be fitted, yielding the values of the trap parameters E and s. Usually the fitting procedures utilize the well-known Randall-Wilkins formula [23]:

$$I = n_0 s e^{-\frac{E}{k_B T}} e^{\left(-\frac{s}{\beta} \int\limits_{T_0}^T e^{-\frac{E}{k_B T}} dT\right)}$$
(3)

where I is the TL intensity,  $n_0$ —the initial concentration of filled traps,  $\beta$ —the heating rate, and  $T_0$ —temperature, at which



Fig. 8. Scintillation yield of LuAG:10% as a function of temperature, normalized to 1 in maximum. The empty and filled data symbols represent the values of yield derived from pulse height spectra recorded with a shaping time of 3 and  $10 \mu s$ , respectively; the dashed and solid curve are fits based on the single-trap model. Error bars are not shown for clarity of the figure.



Fig. 9. Scintillation yield of LuAG:0.23% as a function of temperature, normalized to 1 in maximum. The data symbols represent the values of yield derived from pulse height spectra recorded with a shaping time of 3  $\mu$ s [1]; the solid curve is a fit based on the single-trap model.

the heating starts. This equation, however, holds only in case of a "one trap—one recombination center" system obeying 1storder kinetics. A glow curve of  $Lu_3Al_5O_{12}$ :3%Pr is shown in Fig. 10. At least three peaks can be distinguished around 80, 120, and 230 K, but none of them resembles a characteristic asymmetric 1st-order glow peak. Therefore instead of fitting the whole curve we proceed the other way round, employing (3) to simulate the peaks related to the predicted traps. As listed in Table IV, in TL measurements at a heating rate of 0.15 K/s the peaks would appear at 73-89 K. In particular, the 0.145 eV trap predicted for LuAG:3%Pr would peak at 85 K. Indeed there is thermoluminescence in this area, what confirms our calculations based on the single-trap model. The much broader recorded glow peak compared to the simulated one may be attributed to a large deviation from the 1st-order kinetics or with existence of a statistical distribution of trap depths instead of a single discrete energy. A similar feature has been reported by Wojtowicz *et al.* for YAlO<sub>3</sub>:Ce [19]. We note that the traps peaking at 120 and 230 K are too deep to produce any temperature-dependent variations of yield within our experimental limits, i.e., between 78 and 600 K. Nevertheless, these traps are likely to decrease the yield of the material at temperatures to far above 600 K on the same principle as the ~0.15 eV trap does below 150 K, unless they saturate rapidly. The scintillation yield of a completely trap-free LuAG:Pr could be thus significantly enhanced. 326



Fig. 10. A glow curve of LuAG:3%, measured at a heating rate of 0.15 K/s, following a 5 min X-ray irradiation. The data symbols come from the experiment; the solid curve is a simulated glow peak based on the parameters from Table IV.

A thermal release of a trapped carrier is not the only way of emptying a trap. An adequate amount of energy can also be delivered optically, which can be accomplished experimentally by laser excitation at an appropriate wavelength. Using this idea we can obtain an alternative evidence for the responsibility of traps for the decrease of radioluminescence or scintillation yield. Suppose the scintillating crystal is kept at low temperature. According to (2) its yield is then equal to  $aY_0$ . By switching the laser on we deliver enough energy to release any trapped electron quickly and thereby increase the yield to  $Y_0$ . The results of such experiment, employing an X-ray tube and an infrared laser as excitation sources, are illustrated in Figs. 11 and 12. To explain the experimental scheme, we first trace the solid curve in Fig. 11, measured at 10 K, together with the sequence specified in Table V:

- 0–10 s: there is no excitation, hence no signal is detected;
- 10–20 s: the laser is turned on and a weak signal attributed to optically-stimulated luminescence (OSL) is observed; evidently some of the traps must have already been filled;
- 20–50 s: the X-ray shutter is opened and the luminescence intensity goes up, saturating between 45 and 50 ns;
- 50–80 s: the laser is turned off and the intensity drops by 20%, because electrons are being trapped again;
- 80–150 s: the X-ray shutter is closed and there appears afterglow on a scale of several seconds, which indicates the presence of traps responsible for TL peaks above 10 K;
- 150–280 s: the laser is turned on again and a strong OSL signal emerges.

Contrary to the case of 10 K, the dashed curve in Fig. 11, recorded at 300 K, reveals no influence of presence or absence of the laser excitation. The signal between 20 and 80 s thus determines the level of radioluminescence yield unaffected by the shallow electron traps. This level can also be attained at 10 K, but only upon turning the laser on (20–50 s). Apparently the infrared light is capable of "switching off" the traps by



Fig. 11. Intensity of luminescence of LuAG:3%, recorded with a PMT during X-ray (X), infrared (IR), simultaneous X + IR, or no excitation according to the sequence in Table V, at 10 and 300 K.



Fig. 12. A 3D representation of evolution of the curves, the two marginal of which are shown and explained in Fig. 11, as a function of temperature.

delivering sufficient amounts of energy. During exclusive X-ray irradiation (50–80 s) the low temperature yield is decreased by about 20% due to the trapping of electrons. The OSL signal between 150 and 280 s provides direct evidence that those trapped electrons can be released by optical excitation.

A complete set of curves like in Fig. 11, measured one by one between 10 and 300 K with a 5 K interval, is displayed in Fig. 12. Looking along the temperature axis in the 50–80 s interval one observes a similar curve to the one in Fig. 6. The reduction of luminescence intensity caused by the trapping of electrons decreases with increasing temperature and vanishes above 250 K. The magnitude of the reduction correlates with the OSL intensity between 150 and 280 s. At any temperature, however, the traps can be "switched off" by turning the laser on,

TABLE V IRRADIATION SEQUENCE IN THE EXPERIMENT WITH SIMULTANEOUS X-RAY AND INFRARED EXCITATION

Time (s)	X-ray tube	Infrared laser
0-10	OFF	OFF
10-20	OFF	ON
20-50	ON	ON
50-80	ON	OFF
80-150	OFF	OFF
150-280	OFF	ON

as indicated by the constant intensity in the 45-50 s interval. We note that both the value of 20%, by which the yield is decreased at low temperature, and the range of 10-250 K, in which the traps affect the yield, are consistent with the data in Fig. 6, coming from our previous experiments at X-ray excitation.

## IV. CONCLUSION

The results presented in this paper suggest that the current state of the Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr scintillator development still leaves some room for improvement. Nowadays, when the crystals can be easily grown by the micro-pulling-down method and the optimal praseodymium concentration for the best scintillation performance has been established, efforts should be made to remove or at least reduce the contribution from traps, which are responsible for the temperature-dependent decrease of radioluminescence and scintillation yield. From the different kinds of trapping centers detected via thermoluminescence only the shallow electron traps could have been characterized quantitatively with our experimental techniques. These shallow traps account for a 20% reduction of the room temperature scintillation yield of LuAG:Pr in a pulse height measurement with a shaping time of 3  $\mu$ s. The deeper traps are also expected to decrease the yield at room and even much higher temperatures, but for the moment it is not possible to provide any numbers. Anyway, a successful growth of a trap-free Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Pr, characterized by a substantially increased scintillation yield, in addition to the already recognized splendid energy resolution and the fast 20 ns scintillation decay, would make this material one of the best oxide scintillators known today.

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