LPE Growth of Composite Thermoluminescent Detectors Based on the Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce Single Crystalline Films and YAG:Ce Crystals

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Abstract: This work is dedicated to the development of new types of composite thermoluminescent (TL) detectors for simultaneous registration of the different components of ionization radiation based on the single crystalline films (SCFs) of Ce$^{3+}$-doped Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce ($x = 0–1.5$) garnet and Y$_3$Al$_5$O$_{12}$:Ce (YAG:Ce) substrates using the liquid phase epitaxy (LPE) growth method. For this purpose, the TL properties of the mentioned epitaxial structures were examined in Risø TL/OSL-DA-20 reader under excitation by $\alpha$- and $\beta$-particles from $^{242}$Am and $^{90}$Sr-$^{90}$Y sources. We have shown that the cation engineering of SCF content can result in more significant separation of the TL glow curves of SCFs and substrates under $\alpha$- and $\beta$-particle excitations in comparison with the prototype of such composite detectors based on the Lu$^3$Al$_5$O$_{12}$:Ce (LuAG:Ce)/YAG:Ce epitaxial structure. Specifically, the difference between the TL glow curves of Lu$_{1.5}$Gd$_{1.5}$Al$_5$O$_{12}$:Ce SCFs and YAG:Ce substrates increases up to 120 K in comparison with a respective value of 80 degrees in the prototype based on the Lu$^3$Al$_5$O$_{12}$:Ce/YAG:Ce epitaxial structure. Therefore, the LPE-grown epitaxial structures containing Lu$_{1.5}$Gd$_{1.5}$Al$_5$O$_{12}$:Ce SCFs and Ce$^{3+}$-doped YAG:Ce substrate can be successfully applied for simultaneous registration of $\alpha$- and $\beta$-particles in mixed fluxes of ionization radiation.

Keywords: liquid phase epitaxy; single crystalline films; mixed garnets; thermoluminescence; TL detector cations

1. Introduction

The advancement of composite scintillators (CSs) and thermoluminescent (TL) detectors for the registration of the components of ionization fluxes is now an actual subject of luminescent materials engineering. The basis for such novel engineering are the latest decisions in creating bulk single crystal (SC) and single crystalline film (SCF) scintillators [1,2] as well as the technologies of their production using the Czochralski method [3,4] and the liquid phase epitaxy (LPE) growth technique, respectively [5–11]. Namely, in our previous work, we have shown the possibility of simultaneous registration of $\alpha$-particles and $\gamma$-quanta using the separation of the scintillation decay kinetics of the film (SCF) and crystal (SC) parts of composite scintillators, based on the epitaxial structures of different garnet compounds [12–16].

The application of composite scintillators presupposes the active in situ mode of the registration of incoming ionization fluxes. Meanwhile, such a mode of registration is not always possible to use...
in the case of low doses of radiation and a long-time exposition of radiation. Namely, particles and quanta with different energies cannot be registered using the separation between scintillation decay kinetics curves. There are also restrictions in application of the above-mentioned materials regarding the analysis of liquid and gaseous radioactive materials or the registration of high-dose sources.

The problems described above stimulated us to developing new approaches in the production of composite ionizing radiation detectors. One solution is to develop TL detectors for the simultaneous registration of different components of mixed ionization fluxes using the differences in the thermoluminescence (TL) glow curves. Particularly, such differences can be described by $\Delta T = T_F - T_S$ parameter, e.g., the difference between the position of the main TL peaks, recording from the film ($T_F$) and substrate ($T_S$) parts of the composite detector (Figure 1a).

Figure 1. (a) principal scheme of the composite thermoluminescent (TL) detector, based on the Lu$_{3-x}$Gd$_x$AG:Ce SCF/Y$_3$Al$_5$O$_{12}$:Ce (YAG:Ce) single crystal (SC) epitaxial structure with single crystalline film (SCF) and SC thickness in the 12-15 µm and 0.9-1.0 mm ranges, respectively, and photomultiplier (PMT); (b) the set of TL detectors based on the liquid phase epitaxy (LPE)-grown epitaxial structures containing Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs at $x = 0–1.5$ and YAG:Ce substrates. The irregular shapes of the samples result from cutting the fragment of these samples with a 3.75 mm $\times$ 3. 5 mm $\times$ 5.25 mm size for TL measurements using a Risø TL/OSL-DA-20 reader.

The possibility of the creation of such types of composite TL detectors for the simultaneous registration of $\alpha$- and $\beta$-particle excitation was shown firstly by us in [17]. This work is a continuation of the research in this direction and dedicated to the development of new types of composite TL detectors for the simultaneous registration of the different components of mixed ionization fluxes based on the SCFs of Ce$^{3+}$-doped Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs (at $x = 0–1.5$) garnet and Y$_3$Al$_5$O$_{12}$:Ce (YAG:Ce) SC substrates using the LPE growth method.

Recently, the SCFs of Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce garnet with the Gd content, $x$, from 0 up to 2.5 has been successfully crystallized by the LPE method onto undoped YAG substrates from the melt-solutions based on PbO-B$_2$O$_3$ flux, and their optical properties have been investigated as well [12]. In the present work, we used the LPE-grown epitaxial structures, containing Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs with different Gd concentrations in the $x = 0–1.5$ range, and Ce$^{3+}$-doped YAG:Ce substrates for the simultaneous registration of $\alpha$- and $\beta$-particles in the mixed ionization fluxes. For this purpose, the TL properties of the epitaxial structures were examined under excitation by $\alpha$- and $\beta$-particles from $^{242}$Am and $^{90}$Sr-$^{90}$Y sources, respectively. We expected that the cation engineering of SCF content would result in more significant separation of the TL glow curves of SCFs and substrates in comparison with the prototype of such composite detectors based on the LuAG:Ce SCF/YAG:Ce SC epitaxial structure [17].

2. Growth of Composite Detectors

Four sets of composite detectors, based on the epitaxial structures containing Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs with Gd content $x = 0$, 0.5, 1, and 1.5 and YAG:Ce substrates with the (110) orientation and a thickness of 0.5 mm, were crystallized using the LPE method from the melt-solution based on PbO-B$_2$O$_3$ flux (Figure 1). The conditions of the growth of these structures are presented in Table 1.
**Table 1.** Conditions of the LPE growth of Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs at $x = 0-1.5$ onto YAG:Ce substrates. H—SCF thickness; f and T—velocity and temperature of the SCFs growth; a—SCF lattice constants, m—SCF/substrate misfit, LY—relative photoelectron light yield (LY) of SCFs under $\alpha$-particle excitation in comparison with the LY of YAG:Ce SCF standard samples with a LY of 2.65 ph/KeV.

| Nominal Content of SCFs in Melt-Solution | Substrate | $m$, % | $h$, $\mu$m | $T$, $^\circ$C | $f$, $\mu$m/min | LY, % |
|-----------------------------------------|-----------|--------|-------------|--------------|---------------|------|
| YAG:Ce SC                               | YAG:Ce    | 0.83   | 37          | 973          | 0.43          | 98   |
| Lu$_{2.5}$Gd$_{0.5}$AG:Ce SCF           | YAG:Ce    | 0.49   | 51          | 970          | 0.73          | 63   |
| Lu$_2$Gd$_{1.5}$AG:Ce SCF               | YAG:Ce    | 16     | 970         | 0.53         | 54            |
| Lu$_{1.5}$Gd$_{1.5}$AG:Ce SCF           | YAG:Ce    | 0.04   | 20          | 975          | 0.4           | 45   |

Due to the fact that the segregation coefficient of Gd$^{3+}$ ions in the LPE growth of LuAG-based SCF onto YAG substrates is equal to 0.95–1.05 [18], the content of SCFs was close to a nominal content of garnets in the melt-solution. The concentration of Ce$^{3+}$ ions in SCFs and YAG:Ce substrates was in the 0.25–0.5 at % range.

XRD measurements were used for the characterization of the structural quality of Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCF samples with different Gd content and determination of the SCF lattice constants and SCF/substrate misfit m (Figure 2).

![XRD patterns](image1)

**Figure 2.** (a) XRD patterns of (880) planes of Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs at $x = 0, 0.5,$ and 1.5, grown onto YAG:Ce substrates; (b) the dependence of the misfit between the lattice constants of Lu$_3$–$x$ Gd$_x$Al$_5$O$_{12}$:Ce SCF and YAG substrate on Gd content $x$.

From the respective XRD patterns of the Lu$_3$–$x$Gd$_x$Al$_5$O$_{12}$:Ce SCFs at $x$ value ranging from 0 to 1.5 we also calculated the lattice constant of the different garnet compositions and estimated the misfit between the lattice constants of SCF and YAG substrate $\Delta a = (a_{SCF} - a_{sub}/a_{sub}) \times 100\%$ (Table 1). We found that the lattice constant of Lu$_3$–$x$Gd$_x$Al$_5$O$_{12}$:Ce SCF and the misfit value $m$ steeply depends on the Gd content in accordance with Vegard’s law. Namely, the lattice constant changed from 11.902 Å for LuAG SCFs to 11.998 Å for Lu$_{1.5}$Gd$_{1.5}$Al$_5$O$_{12}$:Ce SCFs. The value of misfit $m$ varies from $-0.83\%$ for LuAG SCF to $0.04\%$ for Lu$_{1.5}$Gd$_{1.5}$Al$_5$O$_{12}$:Ce SCF (Figure 1b), i.e., the last sample was grown practically without SCF/substrate misfit.

The full width at half maximum (FWHM) of the XRD peaks of the garnet samples under study was also determined from the respective patterns. As can be seen from this figure, the structural quality of Lu$_{2.5}$Gd$_{0.5}$AG:Ce and Lu$_{1.5}$Gd$_{1.5}$AG:Ce SCF samples, proportional to the FWHM of the respective XRD peaks (0.15 and 0.14 degree, respectively), is very close to that of LuAG:Ce SC (0.12 degree).
3. Experimental Technique

For the characterization of the luminescent properties of the Lu$_{3-x}$GdxAl$_5$O$_{12}$:Ce SCF/YAG:Ce SC structures, we used absorption spectra, cathodoluminescence (CL), and thermoluminescent (TL) spectra. The absorption spectra were measured using a Jasco 760 UV-Vis spectrometer (Jasco Int. Co. Ltd., Tokyo, Japan) in the 200–1100 nm range. The CL spectra were measured at room temperature (RT) using a SEM JEOL JSM-820 electron microscope (JEOL, Tokyo, Japan) additionally equipped with a spectrometer Stellar Net with a TE-cooled Charge Coupled Device (CCD) detector working in the 200–925 nm range (StellarNet Inc, Tampa, FL, USA).

The TL glow curves were measured under the excitation by α- and β-particles from $^{241}$Am and $^{90}$Sr + $^{90}$Y sources. For measuring the TL in a Risø TL/OSL-DA-20 reader (Risø DTU, Roskilde, Denmark) we used the triangular fragment of samples with 3.75 mm × 3.75 mm × 5.25mm dimensions.

It is worth to note here that the mechanism of TL in the SCF samples under study was connected with the electron liberation from deeper electron traps and their subsequent recombination with the holes localized around Ce$^{3+}$ ions [17,18]. Therefore, for the registration of the TL glow curves, a “green” Schott BG 39 filter was used. The transmittance range of this filter, extending from 350 to 700 nm, was well matched with the emission range of the Ce$^{3+}$ luminescence in the SCF samples.

4. Optical Properties of Lu$_{3-x}$GdxAl$_5$O$_{12}$:Ce SCFs/YAG:Ce Epitaxial Structures

4.1. Absorption Spectra

The absorption spectra of Lu$_{3-x}$GdxAl$_5$O$_{12}$:Ce SCF/YAG:Ce SC epitaxial structures in comparison with the absorption spectrum of the YAG:Ce substrate are shown in Figure 3. It is worth noting that the absorption spectra of the epitaxial structures represent the mixes of the spectra of the YAG:Ce substrate and the respective SCF sample with a total thickness in the 32–100 µm range (see Table 1).

![Figure 3. Absorption spectra of LuAG:Ce SCF/YAG:Ce SC, Lu$_{2.5}$Gd$_{0.5}$AG:CeSCF/YAG:Ce, LuGdAG:Ce SCF/YAG:Ce SC, and Lu$_{1.5}$Gd$_{1.5}$AG:Ce SCF/YAG:Ce composite detectors in comparison with YAG:Ce substrate.](image-url)

The absorption bands peaked within 453–461 nm and at the 340–342 nm range in the spectra of all the composite detector samples and YAG:Ce substrates were related to the 4f(2$^F_{5/2}$)→5d$^{1,2}$ transitions of Ce$^{3+}$ ions E$_1$ and E$_2$ bands, respectively. The ∆E values, proportional to the crystal field strength on the dodecahedral position of the garnet lattice [18,19], were equal approximately to 1 eV for these samples.

The bands that peaked around 263 nm correspond to the absorption of Pb$^{2+}$ flux-related impurity in Lu$_{3-x}$GdxAl$_5$O$_{12}$:Ce SCFs and caused by the $^1S_0→^3P_1$ transitions of these ions [18]. The intensity of this band increased with raising the Gd content in SCFs. Such behavior was connected with
increasing the lattice constant of the garnet and respective dimension of the dodecahedral sites for
the localization of the Pb\(^{2+}\) ions with large ionic radii (1.29 Å) in comparison with Lu\(^{3+}\) (0.997 Å) and
Gd\(^{3+}\) (1.078 Å) cations. Meanwhile, the concentration of this impurity even in the highly Gd-doped
SCFs was still below the detection limit (100 ppm) of a SEM JEOL JSM-820 electron microscope used
for the microanalyses of the samples’ content and only the intensity of Pb\(^{2+}\)-related absorption bands
(Figure 3) could be used for the estimation of the concentration of these ions.

The large Pb\(^{2+}\) ion content in Gd-doped samples could also be contributed to the decrease of the
scintillation LY of these SCFs (see Table 1). A negative influence of Pb\(^{2+}\) on the LY of SCF scintillators
was recently observed for many types of SCF scintillators [6,9,18]. Such influence can also be connected
with the possible creation of Pb\(^{2+}\)-Ce\(^{4+}\) pairs with local charge and local compensation at relatively large
concentration of lead ions in SCF samples. Indeed, the absorption spectrum of Lu\(_{1.5}\)Gd\(_{1.5}\)Al\(_5\)O\(_{12}\):Ce
SCF (Figure 2, curve 5) shows the presence of an additional broad band peak at approximately
255 nm. Most probably, this band was related to the O\(^{2-}\)→Ce\(^{4+}\) charge transfer transitions (CTT).
The confirmation of this conclusion was a close position of these bands in the mentioned SCF sample
to the similar O\(^{2-}\)→Ce\(^{4+}\) CTT band in Ca\(^{2+}\)- and Mg\(^{2+}\)-doped LuAG:Ce SCs [20,21].

4.2. Cathodoluminescence Spectra

The normalized CL spectra of Lu\(_{3-x}\)Gd\(_x\)Al\(_5\)O\(_{12}\):Ce SCFs at x = 0, 0.5, and 1.5 in comparison with
the CL spectrum of YAG:Ce substrate are shown in Figure 4. The maximum of Ce\(^{3+}\) emission bands
was shifted (see Figure 4b) to the red range with increasing Gd content in the SCF samples due to
increasing crystal field strength in the dodecahedral position of the garnet lattice, see also [18,19].

Figure 4. CL spectra (a) of LuAG:Ce, Lu\(_{2.5}\)Gd\(_{0.5}\)AG:Ce, LuGdAG:Ce, Lu\(_{1.5}\)Gd\(_{1.5}\)AG:Ce SCFs in
comparison with YAG:Ce substrate and (b) shift of emission band.

4.3. Thermoluminescence Spectra

The first attempt to create a composite TL detector based on the LuAG:Ce SCF/YAG:Ce epitaxial
structures was described in our previous work [17]. The observed difference (about 80 degrees) between
the position of the main TSL peaks of the SC and SCF parts of the composite detector under α- and
β-particle excitation enabled the simultaneous registration of these particles in the mixed ionization
fluxes. Meanwhile, the optimization of the TSL properties of such a type of composite detector is also
possible and was considered in this work.

In the first stage, we analyzed the thermoluminescent curves for substrate. Figure 5 shows the
similar TSL properties under excitation by α- and β-particles, but the integral intensity of the TSL
peaks is strongly determined by the absorbed dose of radiation (curves 1 and 2, respectively). The TSL
glow curves of Lu\(_{3-x}\)Gd\(_x\)Al\(_5\)O\(_{12}\):Ce SCFs/YAG:Ce SC epitaxial structures with different Gd content x
under excitation by α- and β-particles from \(^{241}\)Am and \(^{90}\)Sr + \(^{90}\)Y sources, respectively, are shown in
Figure 6.
As mentioned above, the $\alpha$-particles from an $^{241}$Am (5.5 MeV) source with a typical passway in the LuAG of 12–15 $\mu$m were fully stopped in the SCF parts of a composite detector with a thickness of several tens of microns, when $\beta$-particles from a $^{90}$Sr + $^{90}$Y source with an average energy of 1.1 MeV could penetrate in the whole substrates. Therefore, due to the high thickness of the SCF samples in the 16–51 $\mu$m range (Table 1), the TSL glow curves under $\alpha$-particle excitation correspond exclusively to SCFs.
As can be seen from Figure 6a, after α-particle irradiation, the main peak of the LuAG:Ce SCF/YAG:Ce SC structure was observed at 600 K. This result is consistent with the results of our previous work [17]. In accordance with the results of the work in [18], increasing the Gd content in the Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs led to the shift of position of the main TSL peaks to the low temperature range, to 390 K in the Lu$_{1.5}$Gd$_{1.5}$Al$_5$O$_{12}$:Ce SCF sample (Table 2 and Figure 6d). Meanwhile, the position of the main TL peaks in the Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCF/YAG:Ce SC structure after β-particle irradiation did not change notably (Table 1) due to the fact that the β-particles were mainly absorbed by the YAG:Ce substrate. Therefore, the difference ΔT between the main TL peaks of SCF and substrate parts of a composite detector, corresponding to the registration of α- and β-particles, increased steeply with an increasing Gd content x in the Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs. Namely, ΔT value is equal to 215 degrees for Lu$_{1.5}$Gd$_{1.5}$Al$_5$O$_{12}$:Ce SCFs (Figures 6d and 7, curve 2).

Table 2. Position of the main TL peaks in Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCF/YAG:Ce SC composite structures after irradiation by α- and β-particles. * the main TL peaks.

| SC and SCF Content                    | α-Particles ($^{241}$Am) | β-Particles ($^{90}$Sr + $^{90}$Y) |
|---------------------------------------|--------------------------|------------------------------------|
| LuAG:Ce SCF/YAG:Ce SC                 | 450, 600 *               | 445, 500, 615 *                     |
| Lu$_{2.5}$Gd$_{0.5}$AG:Ce SCF/YAG:Ce SC | 385, 445 *               | 435, 495, 605 *                     |
| Lu$_{2}$Gd$_{1}$AG:Ce SCF/YAG:Ce SC   | 385, 445 *, 593          | 435, 490, 600 *                     |
| Lu$_{1.5}$Gd$_{1.5}$AG:Ce SCF/YAG:Ce SC | 390 *, 510, 615         | 435, 490, 605 *                     |

Figure 7. Dependence of position of the main TL peaks in Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCF/YAG:Ce SC epitaxial structures under α-particle ($^{241}$Am) excitation (1) and the difference between the main TSL peaks of SCFs and SC parts of Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCF/YAG:Ce SC composite detectors under α-particle ($^{241}$Am) and β-particle ($^{90}$Sr + $^{90}$Y) excitation (2).

It is important to note that due to the large feeding in the case of the TL peak positions in the low temperature range, of the most optimal was the location of the TL peaks of Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs in the range above 150 K. For this reason, the creation of a composite detector based on the Lu$_{2}$Gd$_{1}$Al$_5$O$_{12}$:Ce SCF/YAG:Ce SC epitaxial structure was the optimal option. For such a type of composite detector the ΔT value is equal to 155 degrees, which is completely enough for the simultaneous registration of TL signals coming from the SCF and SC parts of a composite detector in the case of the registration of α- and β-particles, respectively.

5. Conclusions

The creation of novel types of composite TL detectors based on the Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCF/YAG:Ce SC epitaxial structures for simultaneous registration of the mixed ionization fluxes, containing α- and β-particles, is reported in this work. The detectors, consisting of Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCFs with Gd content in the x = 0–1.5 range, were fabricated using the liquid phase epitaxy (LPE) growth method onto YAG:Ce substrates. The registration of α- and β-particles by the SCF and SC
components of composite detectors was performed using the differences between TL glow curves of these components of composite detectors.

The results of testing of Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCF/YAG:Ce SC epitaxial structures are encouraging. In the Lu$_{3-x}$Gd$_x$Al$_5$O$_{12}$:Ce SCF/YAG:Ce SC detector, the difference between the positions of the main TL peaks of the glow curves of the SCF and SC components after α- and β-particle irradiation, ΔT, increases steeply from 15 to 215 degrees with an increasing Gd content in the 0–1.5 range. Therefore, such a ΔT value is significantly larger than the 80 degrees obtained recently for an LuAG:Ce SCF/YAG:Ce SC epitaxial structure. Meanwhile, due to feeding influence, the optimal garnet combination for the creation of a composite detector is the Lu$_2$GdAl$_5$O$_{12}$:Ce SCF/YAG:Ce SC epitaxial structure with the ΔT value above 150 degrees.

The obtained results confirm the assumption that the cation engineering of SCF content enables the significant improvement of the TL properties of composite detectors. Namely, the application of such an approach leads to an increase in the difference between the TL glow curves of Lu$_{2.5}$Gd$_{1-0.5}$Al$_5$O$_{12}$:Ce SCFs and YAG:Ce substrates up to 150–215 degrees in comparison with a respective value of 80 degrees for the prototype of a composite detector based on LuAG:Ce/YAG:Ce epitaxial structures [17].

**Author Contributions:** S.W.-L. collected and analyzed the SCF structural and optical properties and participated in writing and preparation of the paper; V.G. performed SCF growth experiments and wrote the growth part of the paper; A.F. measured the XRD patterns of the SCF samples; T.Z. performed and analyzed the absorption and cathodoluminescence measurements; P.B. and A.M. performed the TSL measurements of the SCF samples; and Y.Z. analyzed all of the experimental materials and wrote the introduction, third part, and conclusion of the paper. All authors have read and agreed to the published version of the manuscript.

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