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# Application of scintillators based on single-crystalline Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce<sup>3+</sup> films for radiation monitoring in biology and medicine

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**Abstract.** Possibility of producing screens of X-ray detectors using liquid phase epitaxy on  $Y_3Al_5O_{12}$  substrates covered by  $Lu_3Al_5O_{12}$ :Ce single crystalline films (SCF) is studied. Optical, luminescent and luminous characteristics of these SCF doped with isoelectronic La, Y, Sc impurities are analyzed. The possibility of crystallization of  $Lu_3Al_5O_{12}$ : Ce SCF on  $Y_3Al_5O_{12}$  substrates by means of matching the lattice parameters of these garnets, when  $Lu^{3+}$  ions in  $Al^{3+}$  octa-sites, is proved. With the aim of matching the emission spectra of SCF based on  $Lu_3Al_5O_{12}$ : Ce, with the spectral sensitivity range of radiation detectors – CCD cameras – we investigated SCF containing  $Gd^{3+}$ ,  $Tb^{3+}$ , and  $Eu^{3+}$  impurities. The maximum light yield, exceeding that of analogs based on  $Y_3AL_5O_{12}$ :Ce SCF by the factor of 1.1-1.5, is shown to be intrinsic for  $Lu_3Al_5O_{12}$ :Ce3<sup>+</sup>,  $Lu_3Al_5O_{12}$ :Ce, Y, La and  $Lu_3Al_5O_{12}$ :Ce, Tb SCF. An increase of the effective atomic number  $Z_{eff}$  and density  $\rho$  up to the values of 60.6 and 7.35 g/cm³ respectively, enables the efficiency of X-ray absorption in comparison with  $Y_3Al_5O_{12}$ :Ce3<sup>+</sup> SCF by the factor of 2.5–8 higher and to reach spatial resolution not less than 0.75–1.0 μm at the SCF thickness of 1.0–2.0 μm.

**Keywords:** x-ray detector, single-crystalline film, spatial resolution, light yield, liquid phase epitaxy, impurities, garnets, Ce-dopant.

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## 1. Introduction

Sources of synchrotron radiation allow to perform visualization of X-ray images of micron and submicron dimensions. This, first of all, finds its application in microtomography, holography as well as in nondestructive testing of industrial samples. The main part of a detector of X-ray images is the screen made of SCF deposited by liquid phase epitaxy (LPE) on the surface of nonluminescent substrates. This screen is intended to transform X-ray images with high resolution into visible light images, which, using microscopic optics, are projected on a recording device based on charge-coupled device (CCD) cameras.

An example of the X-ray image detector of resolution  $R = 1.3-1.5 \, \mu \text{m}$  with the screen on the basis of  $Y_3Al_5O_{12}$ :Ce SCF of thickness  $h = 5 \, \mu \text{m}$  has been described by A.Koch et al. [1–3]. The further increase of R can be achieved only by reducing SCF thickness that, in turn, requires making the screens with maximum possible ab-

sorption coefficient of X-rays  $\eta_{\rm abs} \sim rZ_{\rm eff}^4$ , where  $Z_{\rm eff}$  is the effective atomic number of SCF, and the conversion efficiency  $\eta$  is not less than 2-5%, and a good overlap of emission spectrum with the sensitivity range of CCD cameras take place.

At the same time, LPE technology substantially restricts the class of scintillators suitable for applications in X-ray screens. In particular, high resolution of detectors can be reached only under conditions of a good structural quality of SCF with 1–5 mm thickness. At present only compounds with the garnet structure may be considered as optimal from the viewpoint of the possibility to obtain perfect single crystal luminescent covers by taking into account the development in creating high resolution screens for electron beam tubes [4] and  $\alpha$ -,  $\beta$ - scintillators [5] on the basis homoepitaxial SCF  $Y_3Al_5O_{12}$ :Ce<sup>3+</sup>/ $Y_3Al_5O_{12}$  structures.

Among known garnet compounds the largest values of  $Z_{\rm eff}$  = 58.9 and  $\rho$  = 6.67 g/cm<sup>3</sup> are characteristic to the Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce garnet which can be related to the number

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of efficient high response scintillators. ( $\lambda_{\rm max} = 510$  nm and  $\tau = 46$  ns) with  $\eta = 2.6\%$  [6]. This paper is devoted to analysis of the possibility of producing X-ray screens based on the Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce SCF grown by LPE on Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> substrates with relatively lower prices, as well as to investigation of peculiarities of their optical, luminescent and scintillation characteristics with the aim of choice of optimal compositions for practical applications.

# 2. Peculiarities of manufacturing of SCF by LPE

For reducing the mismatch of the lattice parameters of Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> substrate and SCF based on Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>  $\Delta a = 0.093 \text{ Å down to the value} \le 0.018 \text{ Å (the condition)}$ of epitaxial growth) one can use the options of substitution of Lu<sup>3+</sup> ions in dodecahedral sites (c-) and Al<sup>3+</sup> ions in octahedral (a-) ones of the garnet lattice by the larger isoelectronic impurities (II) which do not have the optical transitions in the range of Ce<sup>3+</sup> ion emission. In this paper, we have studied substitution of Lu<sup>3+</sup> ions by the La<sup>3+</sup> and  $Gd^{3+}$  ones located exclusively in c-cites, as well as substituting Al<sup>3+</sup> ions by Sc<sup>3+</sup> ones which at concentration of x < 0.3 per formula unit (f.u.) occupy a- and cpositions, respectively [7]. At higher scandium concentrations predominantly Al<sup>3+</sup> a-sites are replaced, according to the nearly linear law [8]. It should be noted that doping by Sc has also been used also by the authors of [3] when making Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Tb or Eu SCF, however they had assumed that Sc ions substitute only the a-sites of  $Al^{3+}$ .

The growth of SCF on the basis of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> from the melt solution (MS) based on the PbO:B<sub>2</sub>O<sub>3</sub> (10÷12:1 mol/mol) fluxing agent and the Lu<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub> oxides taken in 1:4 mol/mol ratio. The Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> wafers of (100) or (111) orientation with the diameter of 15-36 mm and the thickness ~ 0.7 mm were used as substrates. The La<sub>2</sub>O<sub>3</sub>, Sc<sub>2</sub>O<sub>3</sub> and Gd<sub>2</sub>O<sub>3</sub> doping oxides were added in MS up to the concentrations of 7 and 10%, respectively, at which the stable growth of SCF was observed. In the case of doping with Gd the Gd<sub>2</sub>O<sub>3</sub> oxide concentration in MS was 7.2-5.9 mol.% what corresponded to the calculated composition of Lu<sub>3-x</sub>Gd<sub>x</sub>Al<sub>5</sub>O<sub>12</sub>:Ce with x = 0.9-1.0 and approximately equal to zero for the lattice mismatch between SCF and substrate.

When depositing the  $Lu_3Al_5O_{12}$  layers on the  $Y_3Al_5O_{12}$  substrates, the SCF contain the Y impurities as a result of substrate etching during the initial stage of the epitaxial growth, and concentration of these ions was raising with the number of samples obtained. Due to this fact, for the effect connected with doping by  $Y^{3+}$  ions to be enhanced we have obtained series of the  $Lu_3Al_5O_{12}$  SCF with  $La_2O_3/Y_2O_3$  ratio equal to  $9\div 13:1$  mol/mol in MS.

The activating  $CeO_2$  was sequentially introduced into MS with the aim of determining the optimal concentration at which the light yield of SCF reaches a maximum. The range of optimal concentrations amounts to 9-12 mas.% of garnet forming components. One must take into account that  $Ce^{3+}$  concentration in MS is an inversely

proportional function of the growth temperature  $T_g$  [5, 9], therefore in the series of SCF obtained from MS of a given composition there always takes place a scattering of SCF parameters which depends on the MS composition fluctuation. For each of the studied garnet compositions the series of SCF at various  $T_g$  from the 885-1040°C range was obtained. The growth rate was 0.6-2.4  $\mu$ m/min and the velocity of substrate rotation was ~ 80 rev./min.

The X-ray microanalysis (XRMA) of the composition of a number of SCF has been fulfilled for estimation of the segregation coefficients of different ions in the SCF based on of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>. For example, the sample obtained from MS which contains Lu<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>, Lu<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub> and CeO<sub>2</sub> with concentrations of 16.2, 64.5, 7.4, 0.80 and 11.1 mol.%, respectively, had the composition close to Lu<sub>3.6</sub>Y<sub>0.18</sub>La<sub>0.035</sub>Ce<sub>0.04</sub>Al<sub>4.42</sub>O<sub>12</sub>. This points to the possibility of localization of the significant part of the Lu<sup>3+</sup> ions in the octahedral sites of the Al<sup>3+</sup> ions which is, in particular, characteristic to the Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> crystal obtained from MS [10]. Obviously, this is promoted by SCF doped by the Ce<sup>3+</sup> and La<sup>3+</sup> ions with much greater ionic radii than in Lu<sup>3+</sup>.

The possibility of appearance of Lu<sup>3+</sup> ions in *a*-sites of the SCF Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce lattice also admits the elimination of lattice mismatch of SCF and Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> substrate without any additional conditions. As a confirmation, we have observed the stable growth of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce SCF from MS on the basis of Lu<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub> and CeO<sub>2</sub> (14 mol.%) despite the certain narrowing the range of homogeneity of this compound in comparison with the Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce,La,Y.

On the basis of XRMA-results we have found that segregation coefficients of  $La^{3+}$ ,  $Ce^{3+}$  and  $Y^{3+}$  ions at growing  $Lu_3Al_5O_{12}$  SCF are equal to 0.021, 0.032 and 1.0, respectively.

## 3. Results and discussion

Optical and luminous characteristics of SCF of investigated contents are presented in Figure and Table. As seen from Fig. a-h, curves 1, absorption in SCF is caused by the  ${}^{1}S_{0} \rightarrow {}^{3}P_{1}$  transitions in the Pb<sup>2+</sup> ions with  $\lambda_{max} = 260 \text{ nm}$ [9, 11], doping of which presents the permanent factor in the SCF synthesis from the Pb containing MS, as well as by the allowed electric dipole transitions with the E<sub>a</sub><sup>1</sup> and  $E_a^2$  energies from the  ${}^2F_{7/2}$  ground state of 4f-shell on the levels of the low doublet of 5d shell of the  $Ce^{3+}$  ion. The distinctive feature of SCF absorption consists in the difference of absorption intensity dependencies, for  ${}^{2}F_{7/2} \rightarrow 5d(E_{a}^{1}, E_{a}^{2})$  and  ${}^{1}S_{0} \rightarrow {}^{3}P_{1}$  transitions on the growth temperature  $T_g$ , which corresponds to the different values of Ce<sup>3+</sup>/Pb<sup>2+</sup> concentration ratio in the SCF content (see, for example, curves 1 in Figs b and c). An optimum is the maximum magnitude of this ratio in the range of optimal  $CeO_2$  concentration in MS that corresponds to maximum values of the light yield of the SCF of studied compositions (Table).

Varying the crystal field strength, which is proportional to the magnitude of  $\Delta E_a = E_a^{-1} - E_a^{-2}$ , results in the

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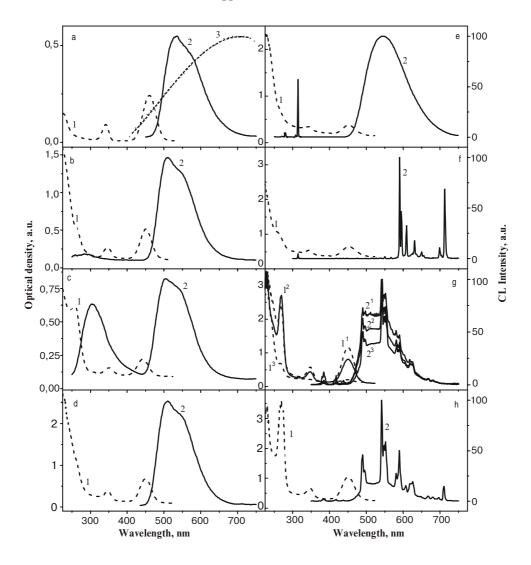


Figure. Absorption (1) and emission (2) spectra of  $Y_3Al_5O_{12}$ :Ce (a),  $Lu_3Al_5O_{12}$ :Ce, La, Y (b),  $Lu_3Al_5O_{12}$ :Ce, Sc (c),  $Lu_3Al_5O_{12}$ :Ce (d),  $Lu_2GdAl_5O_{12}$ :Ce (e),  $Lu_2GdAl_5O_{12}$ :Ce, Eu (f),  $Lu_3Al_5O_{12}$ :Ce, Eu (g),  $Lu_3Al_5O_{12}$ :Ce, Eu (h) SCF. Notations  $1^1$ ,  $1^2$ ,  $1^3$  and  $2^1$ ,  $2^2$ ,  $2^3$  in Fig. g correspond to the concentrations of  $Tb_4O_7$  oxide equal respectively to 0.165, 0.33 and 0.66 mol.% in MS. Curve 3 in Fig. a corresponds to the spectral sensitivity of frontside illuminated CCD camera.

shift of the given absorption bands for investigation compositions with respect to the similar bands for  $Y_3Al_5O_{12}$ :Ce (Table). This decrease in the crystal field strength is also a consequence of the shift of the doublet band of  $Ce^{3+}$  ion luminescence, which corresponds to the  $5d \rightarrow 4f(^2F_{5/2,7/2})$  allowed transition, towards the short wavelength region (Table, Figure) and changing the color of SCF emission from yellow ( $Y_3Al_5O_{12}$ :Ce, Fig. a, 2) to green ( $Lu_3Al_5O_{12}$ :Ce, La, Y, Fig. b, 2) and blue ( $Lu_3Al_5O_{12}$ :Ce, Sc, Fig. c, 2). The emission of  $Ce^{3+}$  in  $Lu_{3-x}Gd_xAl_5O_{12}$ :Ce (x=0.9-1.0) SCF is observed in the yellow-green spectral range (Fig. d, 2; Table) along with the  $Gd^{3+}$  ion luminescence in the UV spectral range ( $^6P_{7/2} \rightarrow ^8S_{7/2}$  transition).

The luminescence spectra of SCF based on of  $Lu_3Al_5O_{12}$ :Ce as well as  $Y_3Al_5O_{12}$ :Ce (Fig. a-d) substantially overlapped with the range of spectral sensitivity of CCD cameras used for radiation detection (Fig. a, curve

3). Therefore we used Si-photodiode with the sensitivity spectrum close to this of frontside illuminated CCD camera for estimation of the light yield of SCF of different compositions (Table). The carried out analysis of variation of the luminescence and light yield spectra versus the doping impurities and  $Ce^{3+}$  activator allows us to establish the following peculiarities of luminescence of SCF based on of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> of different compositions.

# 1) $Lu_3Al_5O_{17}$ : Ce, La, Y.

SCF of this composition have the light yield which exceeds by factor of 1.4 the best samples of  $Y_3Al_5O_{12}$ :Ce SCF. The distinguished feature of their emission is the presence (at small concentration of activator) of luminescence bands in the UV spectral range with  $\lambda_{max} = 267$ 

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Table. Optical and luminous characteristics of SCF on the basis of  $Lu_3Al_5O_{12}$ :Ce. m is the intergral light yield of samples,  $\mu_{FD}$  is the light yield measured by Si-photodiode with the spectral sensitivity close to the sensitivity of frontside illuminated CCD [3].

Location of Ce <sup>+3</sup> absorption		Location of the main emission	Relatively light yield		
maxima, eV					
$^2$ F <sub>7/2</sub> $\rightarrow$ 50	$d(E_a^1, E_a^2)$	$\Delta E_a$	maximum, eV	$\mu_{\mathrm{FD}}$ ,a.u.	μ, a.
3.618	2.695	0.923	2.32	1.00	1.00
3.60	2.75	0.85	2.43	1.40	1.65
3.54	2.80	0.74	2.46	0.17	0.305*
3.61	2.73	0.88	2.274	0.78	0.82
3.56	2.76	0.8	2.435	1.07	1.27
3.558	2.747	0.811	2.28	1.32	1.50
3.564	2.747	0.817	2.28	1.48	1.70
			2.28	1.15	1.30
3.569	2.747	0.822		0.37	0.40
3.587	2.731	0.856	2.09	0.285	0.11
	$\begin{array}{c} \text{ma} \\ ^{2}\text{F}_{7/2} \rightarrow 5 \\ 3.618 \\ 3.60 \\ 3.54 \\ 3.61 \\ 3.56 \\ 3.558 \\ 3.564 \\  3.569 \\  \end{array}$	$\begin{array}{c} \text{maxima, eV} \\ ^2F_{7/2} \rightarrow 5d(E_a^{-1}, E_a^{-2}) \\ 3.618 & 2.695 \\ 3.60 & 2.75 \\ 3.54 & 2.80 \\ 3.61 & 2.73 \\ 3.56 & 2.76 \\ 3.558 & 2.747 \\ 3.564 & 2.747 \\ \hline \end{array}$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	maxima, eV         main emission         yr ${}^2F_{7/2} \rightarrow 5d(E_a{}^1, E_a{}^2)$ $\Delta E_a$ maximum, eV $\mu_{FD}$ ,a.u.           3.618         2.695         0.923         2.32         1.00           3.60         2.75         0.85         2.43         1.40           3.54         2.80         0.74         2.46         0.17           3.61         2.73         0.88         2.274         0.78           3.56         2.76         0.8         2.435         1.07           3.558         2.747         0.811         2.28         1.32           3.564         2.747         0.817         2.28         1.15           3.569         2.747         0.822         0.37

<sup>\*</sup> taking into account UV component.

and 284 nm connected with the radiative recombination of electrons with holes located on the centers of  ${\rm La^{3+}}_{\rm Lu}$  and  ${\rm Lu^{3+}}_{\rm Al}$ , respectively [12, 13]. An intensity of the  ${\rm LU^{3+}}_{\rm AL}$  center emission is comparable with that of of the  ${\rm Lu^{3+}}_{\rm Lu}$  center and can be explained by substantial increasing  ${\rm Lu^{3+}}_{\rm Al}$  center concentration due to the known effect of "displacement" by ions with larger ionic radius ( ${\rm La^{3+}}$ ,  ${\rm Ce^{3+}}$ ) of ions with smaller ionic radius ( ${\rm Lu^{3+}}$ ) in a-sites of the garnet lattice [10] with taking into account that  ${\rm Y^{3+}}_{\rm Lu}$  ions in d-sites do not form radiative luminescence centers [12]. At the concentration  ${\rm CeO_2}$  9 mol.% the luminescence of  ${\rm La^{3+}}$  and  ${\rm Lu^{3+}}$  centers is practically quenched (Fig. b, 2).

# 2) Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, Sc.

The non-trivial peculiarity of properties of SCF of this composition consists in considerably lower magnitudes of light yield (not greater than 17% of Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, Table). The similar effect of luminescence has been earlier observed when in Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce and Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce substituting the a-sites of  $Al^{3+}$  by another II, namely  $Ga^{3+}$  [7, 14]. This allows, by analogy with the conclusions of [14], to suppose the presence for this garnet the excitation energy mechanism connected with transitions between the Ce<sup>3+</sup> levels and the band structure extrema whose probability essentially increases at significant (up to 1.3 per f.u.) concentration of Sc. The second cause of such a small quantum yield of the Ce<sup>3+</sup> emission in this system is great losses on the excitation of formed by Sc II centers of radiative recombination  $\text{Sc}^{3+}_{\text{Al}}$  and  $\text{Sc}^{3+}_{\text{Lu}}$  in the UV range of the spectrum with  $\lambda_{\text{max}} = 290$  and 324 nm (Fig. c, 2) [12, 13]. The comparable emission intensities of these centers with emission band of  $Ce^{3+}$  at  $CeO_2^{opt} = 8-12 \text{ mol.}\%$  testify in favour of the above statement.

# 3) Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce.

The possibility of manufacturing perfect SCF of this composition on the  $Y_3Al_5O_{12}$  substrates assumes the substitution a part of  $Al^{3+}$  a-sites by  $Lu^{3+}$  and formation of  $Lu^{3+}{}_{Al}$  centers of radiative recombination [12, 13]. Moreover the UV luminescence of these centers is quenched at  $CeO_2$  concentration in MS  $\geq$ 14.5mol.% (Fig. c, 2). A slightly lower light yield of SCF of this composition as compared with  $Lu_3Al_5O_{12}$ :Ce, La, Y may be explained by both the differences in  $Ce^{3+}$  ion segregation coefficients and by the different degree of substitution by Lu ions of a-sites of the garnet lattice in the case of presence  $La^{3+}$ ions with the large ionic radius in MS.

As a continuation of the investigations on creation of the effective screen of X-ray detectors based on Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, "engineering" of the SCF emission spectra matched with the range of CCD sensitivity is considered to be of practical importance. With this aim we have synthesized on Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> substrates the Lu<sub>3-x</sub>Gd<sub>x</sub>Al<sub>5</sub>O<sub>12</sub>:Ce (x = 0.9-1.0), Lu<sub>2</sub>GdAl<sub>5</sub>O<sub>12</sub>:Ce, Eu, Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, Tb and Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, Tb, Eu SCF and investigated their optical and light technical characteristics. Here, we proceed from the possibility of shifting the Ce<sup>3+</sup> ion emission in Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> towards long wavelength range in case of doping by Gd<sup>3+</sup> ions [7]; increasing the light yield of Ce<sup>3+</sup> ions by means of sensibilization by Gd<sup>3+</sup> [7] and Tb<sup>3+</sup> emission [4]; increasing the light yield of SCF in the red range of spectrum by doping with Eu<sup>3+</sup> ions [3].

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<sup>\*\*</sup> notations Tb<sup>1</sup>, Tb<sup>2</sup>, Tb<sup>3</sup> correspond to the concentrations of Tb<sub>4</sub>O<sub>7</sub> to 0.165, 0.33 and 0.66 mol.%, respectively.

# 4) $Lu_{3-x}Gd_xAl_5O_{12}$ : Ce (x = 0.9-1.0).

The emission spectrum of  $Ce^{3+}$  in SCF of this composition is close to that of  $Y_3Al_5O_{12}$  (Table), however the light yield of samples does not exceed 0.78 the value of the best  $Y_3Al_5O_{12}$ :Ce SCF samples. The possible reason for such a fact can be the losses due to the excitation of  $Gd^{3+}$  emission in the UV spectral range without marked signs of sensibilization.

# 5) Lu<sub>3</sub>Al<sub>5</sub>O<sub>17</sub>:Ce, Tb.

Doping by  $Tb^{3+}$  ions gives rise to increase of the light yield of SCF (Table) which at the optimal (0.33 mol.%)  $Tb_4O_7$  concentration in MS reaches the magnitudes exceeding by a factor of about 1.5 the light yield of  $Y_3Al_5O_{12}$ :Ce SCF. This fact correlates with the earlier observed noticeable increase of light yield of CL screens based on  $Y_3Al_5O_{12}$ :Ce,Tb SCF [4].

Under increasing  $Tb_4O_7$  concentration a decrease in the  $Ce^{3+}$  ion absorption (Fig. g,  $1^1$ - $1^3$ ) takes place that corresponds to reduction of Ce/Tb ratio in the SCF content. Using the determined by us segregation coefficients of Tb ions, 0.7, and Ce ions, 0.032, we have found the optimal Ce:Tb=1:2 ratio in  $Lu_3Al_5O_{12}:Ce:Tb$  (0.33 mol.% in MS) with maximum light yield. The emission spectrum of these SCF is a superposition of the  $Ce^{3+}$  and  $Tb^{3+}$  spectra. In case of increasing Tb concentration the luminescence quenching is realized in the blue range of spectrum (384-460 nm), that corresponds to  $^5D_3 \rightarrow ^7F_{6-0}$  transitions, and enhancement of emission in the yellow-green range (480-650 nm), what corresponds  $^5D_4 \rightarrow ^7F_{6-0}$  with  $\lambda_{max}=543$  nm (Fig. g, curves  $2^1$ - $2^3$ ).

# 6) Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce:Tb, Eu and Lu<sub>3,x</sub>Gd<sub>x</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, Eu.

Despite the contribution of Eu<sup>3+</sup> emission in the orange (589-600 nm,  ${}^5D \rightarrow {}^7F_1$  transitions) and the red spectral ranges (631-711 nm,  ${}^5D_0 \rightarrow {}^7F_{2,3,4}$  transitions), the doping by Eu ions resets in essential decrease of the light yield in SCF (Table). In particular, at the optimal concentration of EuO oxide in MS 4.35 mol.% according to [3] the luminescence of Ce<sup>3+</sup> ions in Lu<sub>2</sub>GdAl<sub>5</sub>O<sub>12</sub>:Ce:Eu is completely quenched (Fig. f, 2). In addition, taking into account segregation coefficient of Eu<sup>3+</sup> ions determined by us, i. e.1.0, the Ce:Eu ratio in the content of SCF is equal to 1:11.5. The emission spectrum of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce:Tb<sup>3</sup>:Eu at the EuO concentration in MS of 0.75mol.% is determined mainly by the Ce<sup>3+</sup> and Tb<sup>3+</sup> ion emission (Fig. h). The ratio of activators in these SCF may be estimated as Ce:Tb:Eu = 1:4:1.5. At the same time, for SCF of this composition more than threefold reduction of the light yield takes place in comparison with the Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce:Tb SCF.

The above mentioned data point to the fact that Eu<sup>3+</sup> ions are the effective quenching agents of Ce<sup>3+</sup> and Tb<sup>3+</sup> emission in the investigated compounds. Taking into account the relationships between the concentrations of acti-

vators in these SCF, we have determined that the light yield increase calculated per one  $Eu^{3+}$  atom with respect to one  $Ce^{3+}$  atom is not less than 45% of initial magnitude.

#### **Conclusions**

Among all studied SCF based on of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce SCF the best luminous characteristics for application as screens of X-ray detectors have the SCF based on Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, Tb, Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, La, Y and Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, light yield of which exceed that of SCF of basic Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce composition by factor of 1.1-1.5. The effect of embedding Lu<sup>3+</sup> ions in octa-sites of lattice, which we have revealed for these SCF, makes it possible to reach the limiting magnitude of  $Z_{\rm eff} = 60.65$ ,  $\rho = 7.35$  g/cm<sup>3</sup> in comparison with the stoichiometric Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> and Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> compounds and to increase the X-ray absorption coefficient by factors of 7–8 and 2.5–3 for the energy ranges of 5–17 and 18–60 keV, respectively. This allows, in accordance with the calculations using the procedure described in [1-3], to obtain (at the SCF thickness of 1.0–2.0 µm and the optical system aperture 1.0) a resolution of X-ray screens not less than 0.75–1.0 µm. In addition, the optimal degree of matching of emission spectra with the spectral sensitivity of CCD cameras is intrinsic to the SCF of Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce, Tb composition.

The SCF based on of  $Lu_3Al_5O_{12}$  doped by Eu and Sc have considerably smaller efficiency as compared to SCF based on  $Lu_3Al_5O_{12}$ :Ce, Tb. In this respect the choice of material, made by the authors [3], for screens of X-ray detectors based on  $Lu_3Al_{5-x}Sc_xO_{12}$ :Eu is not successful. It is inherent to this system to have both the large losses of excitation energy on emission of the centers formed by Sc ions and small, as compared to  $Ce^{3+}$  ions, quantum yield of  $Eu^{3+}$  emission.

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