

Terbium-Doped Garnet Single Crystals as X-ray-Sensitive Phosphors

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Abstract—The spectra of luminescence induced by synchrotron radiation in the fundamental absorption range were measured at 10 and 300 K for Czochralski-grown bulk $\text{Y}_3\text{Al}_5\text{O}_{12}$ single crystals doped with Tb^{3+} ions and for $(\text{Tb},\text{La},\text{Gd})\text{Ga}_5\text{O}_{12}$ single crystal films grown by liquid phase epitaxy from a $\text{PbO}-\text{B}_2\text{O}_3$ based supercooled solution melt on $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ substrates.

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Terbium-doped gadolinium oxysulfide ($\text{Gd}_2\text{O}_3\text{S}:\text{Tb}^{3+}$) has been used for about three decades as an effective powder phosphor [1–4]. The efficiency of X-ray to light conversion, that is, the coefficient of energy transfer from incident X-ray photons to the emitted visible light is about 22% [3]. The high conversion of ionizing and other types of radiation into visible light is provided by a combination of the rare earth matrix ($\text{Gd}_2\text{O}_3\text{S}$), which possesses a high absorption capacity in a broad energy range, and the activator (Tb^{3+}) characterized by a large capture cross section. The conversion efficiency can be improved by decreasing the level of losses caused by intrinsic and extrinsic (impurity) defects [1]. In the systems under consideration, the photon energy absorbed during the $^8\text{S}_0 \rightarrow ^6\text{Pr}_J$ transition between Gd^{3+} ion levels is nonradiatively transferred to the upper levels of Tb^{3+} ion and then radiated upon the $^5\text{D}_4 \rightarrow ^7\text{F}_5$ transition [1, 5].

The main advantages of ceramic phosphors over powder ones are (i) a 1.5–2-times higher density of the former materials (amounting to 99.9% of the X-ray density), which increases the transparency of a screen for the intrinsic radiation, (ii) higher mechanical strength, and (iii) greater stability under irradiation [1]. The further progress toward decrease in the density of intrinsic and extrinsic defects is provided by the passage from ceramics to single crystals.

This Letter presents the results of investigations into the luminescence of bulk and thin-film single crystals of garnets doped with Tb^{3+} ions.

The single crystals of $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Tb}^{3+}$ were grown using the Czochralski technique as described in [5]. The samples of $(\text{Tb},\text{La},\text{Gd})\text{Ga}_5\text{O}_{12}$ single crystal films were obtained by liquid phase epitaxy from a $\text{PbO}-\text{B}_2\text{O}_3$ -based supercooled solution melt on (111)-oriented $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ substrates [6]. Data presented below for the bulk garnet crystal refer to an $\text{Y}_3\text{Al}_5\text{O}_{12}$ plate with a thickness of $h = 2.5$ mm containing 0.7 at. % Tb^{3+} ions. The $(\text{Tb},\text{La},\text{Gd})\text{Ga}_5\text{O}_{12}$ films deposited on the two sides of an 0.5-mm-thick $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ substrate had a total thickness of $h = 11$ μm and contained 3.3 at. % Tb^{3+} ions.

The luminescence was excited by synchrotron radiation (SR) [7] in the range of fundamental absorption of the single crystals under study. The measurements were performed on Superlumi setup arranged in the SR channel of DORIS positron storage ring [8] at DESY (Hamburg, Germany). The luminescence spectra were processed with allowance for the instrumental function, a difference between the SR intensity for $\lambda = 80$ and 85 nm, the accumulation time, and the storage ring current variations during measurements.

Figure 1 shows the SR-excited luminescence spectra of a bulk $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Tb}^{3+}$ single crystal measured at a low temperature (curve 1) and at room temperature (curve 2). As can be seen, the spectra of luminescence excited in the fundamental absorption range (~ 15 eV) of the crystal at low and high temperatures exhibit no principal qualitative differences. The main emission bands exhibit maxima at $\lambda = 495, 550, 590, 630, 676,$ and 689 nm, which correspond to the following interconfiguration transitions in Tb^{3+} ion: $^5\text{D}_4 \rightarrow ^7\text{F}_6$,

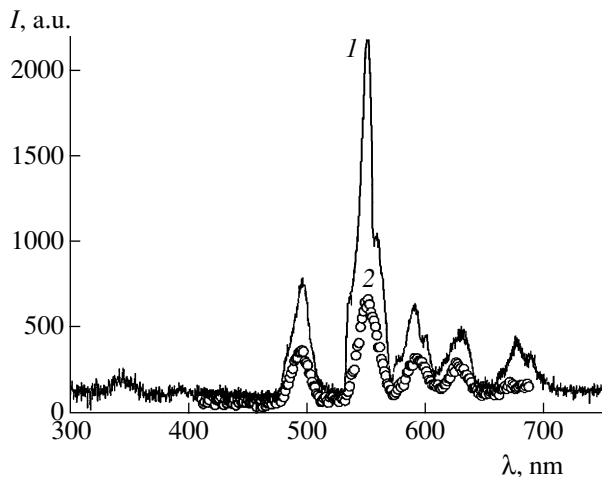


Fig. 1. The luminescence spectra of a bulk $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Tb}^{3+}$ single crystal excited by SR with a photon energy of (1) $E = 15.5$ eV ($\lambda = 80$ nm) at $T = 10$ K and (2) $E = 14.6$ eV ($\lambda = 85$ nm) at $T = 300$ K.

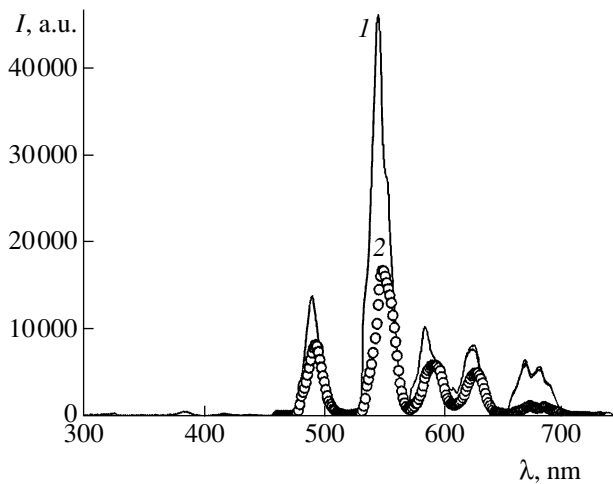


Fig. 2. The luminescence spectra of a $(\text{Tb,Lu,Gd})\text{Ga}_5\text{O}_{12}$ single crystal film excited by SR with a photon energy of (1) $E = 15.5$ eV ($\lambda = 80$ nm) at $T = 10$ K and (2) $E = 14.6$ eV ($\lambda = 85$ nm) at $T = 300$ K.

$^5\text{D}_4 \rightarrow ^7\text{F}_5$, $^5\text{D}_4 \rightarrow ^7\text{F}_4$, $^5\text{D}_4 \rightarrow ^7\text{F}_3$, $^5\text{D}_4 \rightarrow ^7\text{F}_2$, and $^5\text{D}_4 \rightarrow ^7\text{F}_1$, respectively. The peak positions remain virtually unchanged when the sample temperature is increased from 10 to 300 K. As can be seen from Fig. 1, the amplitude of the most intense emission band ($\lambda = 550$ nm) at $T = 300$ K is lower by a factor of about 3.3 as compared to that at $T = 10$ K.

Figure 2 shows the luminescence spectra of a $(\text{Tb,Lu,Gd})\text{Ga}_5\text{O}_{12}$ single crystal film excited by SR with $E \sim 15$ eV and measured at a low temperature (curve 1) and at room temperature (curve 2). As can be seen, this spectrum is similar to that of the bulk $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Tb}^{3+}$ single crystal presented in Fig. 1. The positions of band maxima at $\lambda = 490, 545, 585, 625, 667$, and 689 nm for the film (also assigned to the inter-configuration transitions $^5\text{D}_4 \rightarrow ^7\text{F}_6$, $^5\text{D}_4 \rightarrow ^7\text{F}_5$, $^5\text{D}_4 \rightarrow ^7\text{F}_4$, $^5\text{D}_4 \rightarrow ^7\text{F}_3$, $^5\text{D}_4 \rightarrow ^7\text{F}_2$, and $^5\text{D}_4 \rightarrow ^7\text{F}_1$, respectively, in Tb^{3+} ion) are close to the positions of the corresponding peaks in the spectrum of the bulk $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Tb}^{3+}$ crystal. The most intense luminescence band in the spectrum of epitaxial films has a peak at $\lambda = 545$ nm, that is, exhibits a 5-nm shift toward shorter wavelengths as compared to the analogous peak in the spectrum of the terbium-doped garnet single crystal.

Based on the above results, we conclude that Tb^{3+} ions in the $\text{GdGa}_5\text{O}_{12}$ and $\text{Y}_3\text{Al}_5\text{O}_{12}$ garnet matrices retain both the efficiency and the spectral distribution of luminescence in a broad temperature range (10–300 K). This circumstance ensures stability of the luminescent characteristics of X-ray sensitive screens made of these crystalline materials.

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