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Effect of Ga content on luminescence and defects formation processes in $Gd_3(Ga,Al)_5O_{12}$:Ce single crystals

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ABSTRACT

Luminescence characteristics of Ce^{3+} - doped $Gd_3Ga_xAl_{5-x}O_{12}$ single crystals with different Ga contents (x = 1, 2, 3, 4, 5) are studied in the 9–500 K temperature range. The spectra of the afterglow, photoluminescence, radioluminescence, and thermally stimulated luminescence (TSL) of each crystal coincide. The increase of the Ga content results in the high-energy shift of the spectra while the radioluminescence intensity at 9 K remains practically constant up to x = 4. No Ce^{3+} emission is observed in case of x = 5. The total TSL intensity drastically increases, reaches the maximum value around x = 2–3, and then decreases due to the thermal quenching of the Ce^{3+} emission. The TSL glow curve maxima are gradually shifting to lower temperatures, and the dependence of the maxima positions and the corresponding trap depths on the Ga content is close to linear. However, the activation energy of the TSL peaks creation under irradiation of the crystals in the $4f - 5d_1$ absorption band of Ce^{3+} decreases drastically with the increasing Ga content (especially in the range of x = 1–2), and this dependence is found to be strongly nonlinear. Possible reasons of the nonlinearity are discussed.

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1. Introduction

In recent years, the Ce - doped $Gd_3Ga_xAl_{5-x}O_{12}$ single crystals, ceramics and epitaxial films with different Ga contents (x) were intensively studied as promising scintillator materials for their application in medical imaging because of their extremely high light yield, good energy resolution, relatively high density (6.63 g/cm^3), fast scintillation response, and high radiation stability and hardness [1–25]. Single crystals of $Gd_3Ga_xAl_{5-x}O_{12}$:Ce prepared by the micro-pulling down method were first reported in Ref. [1]. Their scintillation characteristics were found to depend on the Ga concentration. For the crystal with x = 3, the light yield of ≈42 000 photons/MeV, the dominating decay time of ≈53 ns, and the energy resolution of 8.3%@662 keV were obtained. With the decreasing Ga content, the light yield and energy resolution were found to improve, but the decay time increases. The first $Gd_3Ga_3Al_2O_{12}$:Ce single crystal grown by the Czochralski method with the light yield of 46 000 photons/MeV and energy resolution of 4.9%@662 keV was reported in Ref. [2]. Recently, an extremely high light yield of 58 000 photons/MeV and the best energy resolution of 4.2%@662 keV were obtained for the single crystals of $Gd_3Ga_xAl_{5-x}O_{12}$:Ce with x = 2.7 and x = 2.4, respectively, also grown by the Czochralski method [11]. Due to these characteristics, $Gd_3Ga_xAl_{5-x}O_{12}$:Ce with x = 2–3 was considered as a promising scintillator for the PET application [2].

The systematic photoluminescence study of $Gd_3Ga_xAl_{5-x}O_{12}$:Ce showed that the increasing Ga content results in the high-energy shift of the Ce^{3+} - related $5d_1 - 4f$ emission band and the lowest-energy $4f - 5d_1$ absorption (excitation) band as well as in the low-energy shift of the $4f - 5d_2$ excitation band [6,24]. The Stokes shift slightly increases as well. The decrease of both the crystal field strength and the band gap of the host material was reported (see, e.g. [4,6], and references therein). According to [6], the activation energy of thermal quenching of the $5d_1 - 4f$ emission determined from the temperature dependence of the luminescence decay time decreases linearly from $E_q \approx 0.6$ eV for x = 0 to $E_q \approx 0.07$ eV for x = 4. The TSL characteristics were studied for $Gd_3Ga_3Al_2O_{12}$:Ce crystals in Refs. [9,10,13], for the $Gd_3Ga_xAl_{5-x}O_{12}$:Ce epitaxial films

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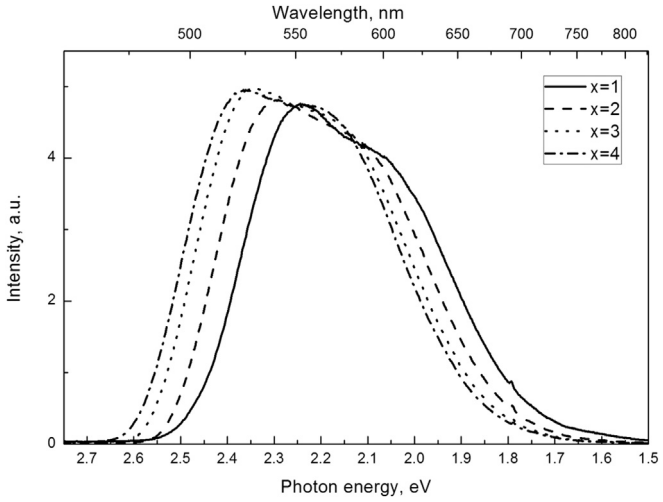

Fig. 1. X-ray excited luminescence spectra of the $Gd_3Ga_xAl_{5-x}O_{12}$:Ce single crystals with different Ga contents (shown in the legend) measured at the same conditions at T = 9 K.

with the Ga content varying from x = 2.7 to x = 3.54, in Ref. [23], and for the $Gd_3Ga_xAl_{5-x}O_{12}$:Ce single crystal with x = 2.83, in Ref. [25], i.e., the x values in the previous studies varied only from 2.7 to 3.54.

In the present work, the characteristics of the photo- and X-ray excited luminescence and TSL of the $Gd_3Ga_xAl_{5-x}O_{12}$:Ce single crystals with different Ga contents (x = 1, 2, 3, 4, 5) grown by the micro-pulling down method are measured in a wide temperature range (9–500 K) and compared. The aim of this work is to investigate the dependences of these characteristics on the Ga content, to explain the mechanisms of these dependences, and to clarify a possible role of Ga^{3+} ions in the defects formation processes in these scintillation materials.

2. Experimental procedure

Single crystals of $Gd_3Ga_xAl_{5-x}O_{12}$:Ce with the same (0.2 at.%) Ce content and different Ga contents (x = 1, 2, 3, 4, 5) were grown by the micro-pulling down method [1]. Pieces of the grown crystals were crushed and ground into a powder in a mortar. Powder X-ray diffraction (XRD) analysis was carried out in the 2θ range 15°–75° using a RINT Ultima (RIGAKU) diffractometer. Cu Kα X-ray source was used, and the accelerating voltage and current were 40 kV and 40 mA, respectively. Quantitative chemical analysis of the crystals for Al, Ce, Y and Gd content along the growth direction was performed by electron probe microanalysis (EPMA; JXA-8621MX, JEOL), for the details see Ref. [1]. Disks of about 1 mm thickness were cut from crystal rods of about 3 mm diameter and polished up to a optical grade.

The steady-state emission and excitation spectra in the 85–500 K temperature range were measured using a setup, consisting of the LOT - ORIEL xenon lamp (150 W), two monochromators (SF - 4 and SPM - 1) and nitrogen cryostat. The luminescence was detected by a photomultiplier (FEU - 39 or FEU - 79) connected with an amplifier and recorder.

The X-ray excited luminescence and afterglow spectra were measured at 9 K after a crystal was irradiated with the X-ray tube (40 kV, 15 mA) for 10 min to achieve the intensity saturation. The luminescence was detected using Andor Shamrock B-303i spectrograph coupled to Andor DU-401A-BV CCD camera.

TSL glow curves $I_{TSL}(T)$ were measured with a heating rate of 0.2 K/s at two different setups. At the first setup, the TSL glow curves were measured in the 85–500 K temperature range after selective ultraviolet (UV) irradiation of the crystals at different temperatures T_{irr} with different irradiation photon energies E_{irr}. A crystal located in a nitrogen cryostat was irradiated with the LOT - ORIEL xenon lamp (150 W) through a monochromator SF - 4. The spectral width of the monochromator slit did not exceed 5 nm. The TSL glow curves were measured with the monochromator SPM - 1 and detected with the photomultiplier FEU - 39 and recorder. For each TSL glow curve peak, the TSL peak creation spectrum, i.e., the dependence of the maximum TSL intensity (I_{TSL}^{max}) on the irradiation photon energy E_{irr}, was measured. From the dependence of the maximum TSL intensity (I_{TSL}^{max}) on the irradiation temperature T_{irr}, the activation energy E_a for the TSL peak creation was determined. To determine the trap depth E_t corresponding to each TSL peak, the partial cleaning method was used (for more details, see, e.g. [26], and references therein). The crystal, irradiated at the temperature T_{irr}, was cooled down to 85 K, heated up to a temperature T_{stop}, then quickly cooled down to 85 K and the TSL glow curve was recorded. In the next cycle, the same procedure was repeated for the different temperature T_{stop}, etc. From the slope of the $\ln(I_{TSL})$ as a function of the reverse temperature (1/T), the E_t value was calculated.

At the second setup, the TSL spectra and glow curves were measured in the 9–300 K temperature range after X-ray irradiation of the crystals with the X-ray tube (40 kV, 15 mA) for 5 min. The luminescence was detected using Andor Shamrock B-303i spectrograph coupled to Andor DU-40 1A-BV CCD camera. Trap depths were determined using fractional glow technique [27].

3. Experimental results and discussion

The radioluminescence spectra of $Gd_3Ga_xAl_{5-x}O_{12}$:Ce measured at the same conditions at 9 K are shown in Fig. 1. The position of the center of the doublet Ce^{3+} - related emission band (E_{em}) and its full width at half maximum (FWHM) for each crystal are presented in Table 1. It is evident that the emission band is shifting to high energies with the increasing Ga content while the radioluminescence intensity at 9 K remains practically constant up to x = 4. No Ce^{3+} emission is observed in case of x = 5. The photoluminescence spectra as well as the spectra of the afterglow and low-temperature TSL of each sample coincide with the spectra shown in Fig. 1. The photoluminescence excitation spectrum consists of several bands

Table 1
The position (E_{em}) and FWHM of the X-ray excited luminescence band at 9 K, activation energy E_q of the luminescence thermal quenching obtained in the present work from the I(T) dependences measured in the T > 430 K temperature region and in Ref. [6] from the temperature dependences of the decay times, activation energy E_a of the TSL peaks creation under irradiation in the $4f - 5d_1$ absorption band of Ce^{3+}, and the energy difference E_{dc} between the $5d_1$ level of Ce^{3+} and the bottom of the CB estimated in this work and reported in Ref. [29] for the $Gd_3(Ga,Al)_5O_{12}$:Ce single crystals with different Ga contents.

Crystal	E_{em}, eV	FWHM, eV	E_q, eV	E_q, eV [6]	E_a, eV	E_{dc}, eV	E_{dc}, eV [29]
$Gd_3Ga_1Al_4O_{12}$:Ce	2.15	0.46	0.40	0.43	1.04	1.04	0.56
$Gd_3Ga_2Al_3O_{12}$:Ce	2.20	0.47	0.42	0.49	0.32	0.76	0.52
$Gd_3Ga_3Al_2O_{12}$:Ce	2.23	0.49	0.33	0.27	0.16	0.50	0.36
$Gd_3Ga_4Al_1O_{12}$:Ce	2.25	0.50	0.34	0.07	0.08	0.25	0.18

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