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# Synthesis and structural properties of thin YAG:$Ce^{3+}$ films produced by pulsed laser deposition

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Thin YAG:$Ce^{3+}$ films were synthesized by pulsed laser deposition. For the first time, the dependences of the thickness, phase composition, and photoluminescence of films on the substrate temperature during synthesis were studied. It is shown that, regardless of the synthesis temperature in the range of 100−400°C, the resulting films had a polycrystalline structure. With an increase in the substrate temperature, an increase in the film growth rate was observed. Despite the polycrystalline structure of the films, after annealing at 900°C, YAG:$Ce^{3+}$ thin films exhibited pronounced photoluminescence, which opens up the possibility of their application as modifying coatings for photoconverters, as well as for X-ray detectors.

**Keywords:** pulsed laser deposition, YAG:$Ce^{3+}$, functional coatings, x-ray diffraction, luminescence.

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Yttrium aluminum garnet and solid solutions with a garnet structure are used widely at present as materials for electronic and photonic devices. Cerium-doped yttrium aluminum garnet (YAG:$Ce^{3+}$) is the most practically relevant of garnet materials. A well-pronounced photoluminescence band in the yellow spectral region [1] makes YAG:$Ce^{3+}$ an efficient luminophore for white light-emitting diodes [2]. In addition, YAG:$Ce^{3+}$ exhibits scintillation properties and is used to fabricate X-ray radiation detectors [3,4]. The modification of characteristics of photoconverters [5] is a promising field of application of YAG:$Ce^{3+}$ films. Being highly transparent in the visible range, YAG:$Ce^{3+}$ films have the capacity to convert ultraviolet (320−380 nm) and blue (400−500 nm) solar radiation into longer-wave radiation (500−680 nm).

Since YAG:$Ce^{3+}$ is a high-temperature material, its films may be fabricated using only a select few methods, such as liquid-phase epitaxy [6], plasma sputtering [7], or pulsed laser deposition (PLD) [8].

The aim of the present study is to synthesize thin YAG:$Ce^{3+}$ films on Si by pulsed laser deposition and examine the influence of the substrate temperature on their structural properties and luminescence.

Thin YAG:$Ce^{3+}$ films were fabricated using an experimental PLD system [9,10]. A target was formed by uniaxial cold pressing of stoichiometric YAG:$Ce^{3+}$ powder with a composition characterized by formula $Y_{2.98}Ce_{0.02}Al_5O_{12}$. This powder was synthesized by inverse coprecipitation. Single-crystalline *p*-Si (100) was used as substrates. Silicon substrates were prepared in accordance with the modified Shiraki technique detailed in [11]. The residual pressure in the chamber was as high as $10^{-4}$ Pa. Second-harmonic radiation (532 nm) of a YAG−$Nd^{3+}$ laser was used for sputtering. The substrate temperature was varied from 100 to 400°C. The laser radiation energy density was 3.5 J/cm$^2$, and the deposition time was 90 min. The pulse duration was 10 ns at a pulse repetition rate of 15 Hz. Thermal annealing of the synthesized films was performed in air at a temperature of 900°C for 120 min.

The phase composition of films was examined by small-angle X-ray diffraction using an ARL X'TRA (Thermo Fisher Scientific) diffractometer with a Cu$K_\alpha$ radiation source in the $\omega-2\theta$ geometry at angle $\omega = 0.5°$. The surface morphology of YAG:$Ce^{3+}$ films was studied with a MIRA3-LMH (Tescan) scanning election microscope (SEM) with an AZtecEnergy Standart/X-max20 system for determination of the elemental composition. Photoluminescence spectra were measured with an SFL-MDR (OOO „Spektr", Russia) spectrofluorimetric setup. The excitation wavelength was 447 nm, and the luminescence intensity was measured within the 460−740 nm wavelength range. The film thickness was determined ellipsometrically using an SE800 (Sentech Instruments GmbH) spectroscopic ellipsometer.

The surface morphology of YAG:$Ce^{3+}$ films was examined after thermal annealing (Fig. 1). Spherical droplets were observed on the surface of YAG:$Ce^{3+}$ films synthesized at a substrate temperature of 400°C (Fig. 1, *a*). The mean size of these droplets was 124 nm. Large 1−3 $\mu$m droplets were also found. The mean size of droplets on the YAG:$Ce^{3+}$ film surface after annealing (Fig. 1, *a*) was 83 nm. Large droplets on the film surface were associated with smaller ones. Figure 1, *b* presents the microscopic image of a cleaved face of the YAG:$Ce^{3+}$ film sample after annealing. The film is continuous and has a thickness of approximately 111 nm. Figure 2 shows the

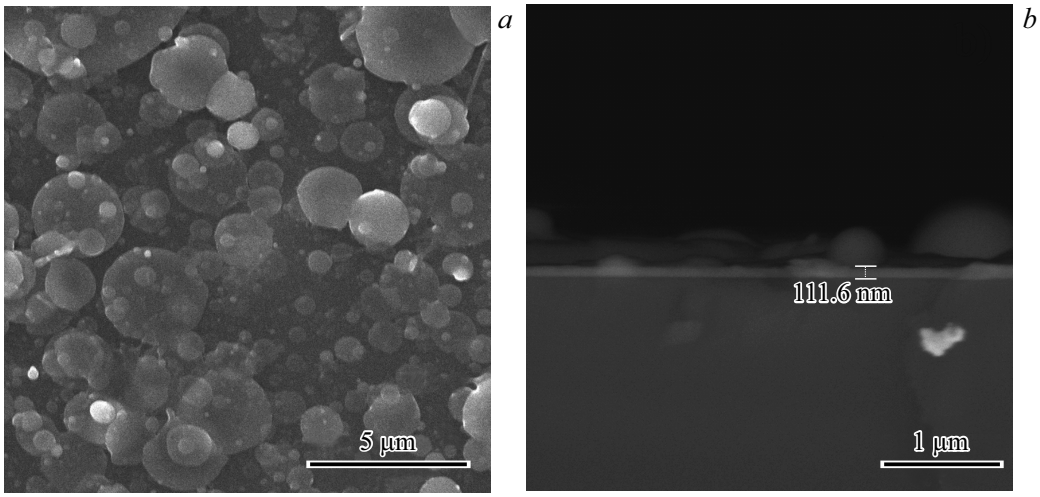


**Figure 1.** SEM images of the surface (*a*) and the cleaved face (*b*) of thin YAG:$Ce^{3+}$ films synthesized on a Si substrate at a temperature of 400°C.

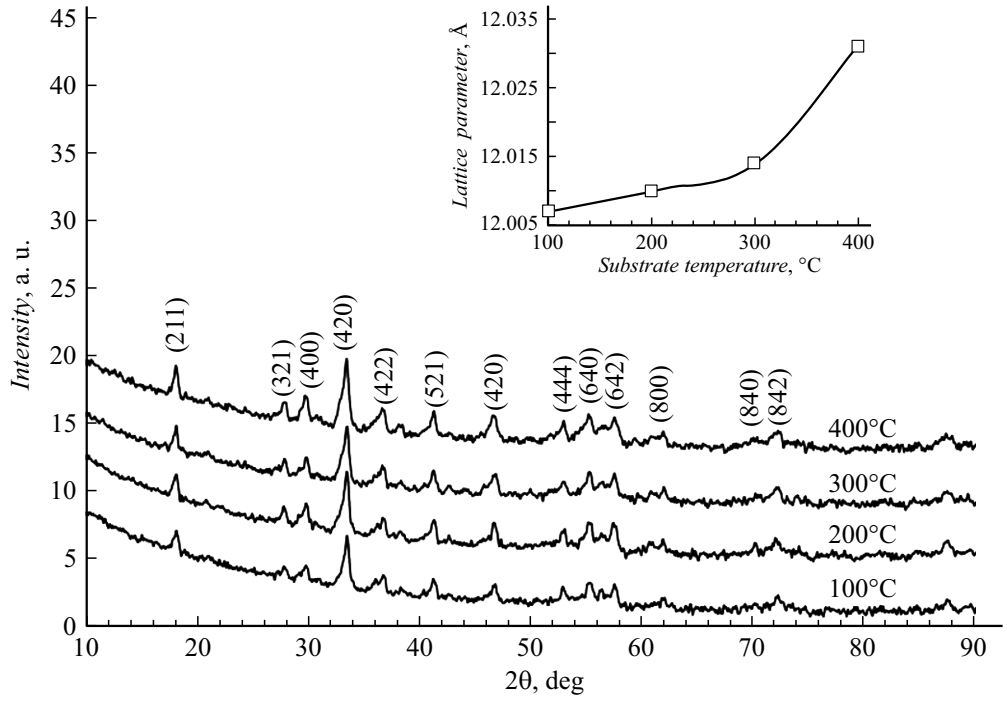


**Figure 2.** Diffraction patterns of thin YAG:$Ce^{3+}$ films, which were synthesized at different substrate temperatures, after annealing. The dependence of the YAG:$Ce^{3+}$ lattice parameter on the substrate temperature in shown in the inset.

diffraction patterns of thin YAG:$Ce^{3+}$ films after annealing. These diffraction patterns revealed that films are single-phase and have a garnet structure with a composition of $Y_{2.98}Ce_{0.02}Al_5O_{12}$. The films were X-ray amorphous prior to annealing. Reflections corresponding to planes (211) and (420) had the highest intensity in all samples; less intense reflections (321), (400), (422), and (521) were also observed. This is indicative of a polycrystalline structure of thin films and of the lack of a preferred orientation matching the crystal-lattice Si (111) substrate orientation. The lattice parameter of YAG:$Ce^{3+}$ in thin films increased from 12.007 to 12.031 Å with substrate temperature. This may imply that the fraction of embedded cerium cations increases (inset in Fig. 2).

Figure 3, *a* presents the luminescence spectra of thin YAG:$Ce^{3+}$ films, which were synthesized at different substrate temperatures, after annealing. All the obtained film samples exhibit marked luminescence with its intensity maximum at a wavelength of 528 nm. This maximum did not shift at higher substrate temperatures. A broad (480–680 nm) luminescence band was observed. This spectrum shape is typical of YAG:$Ce^{3+}$. Having analyzed

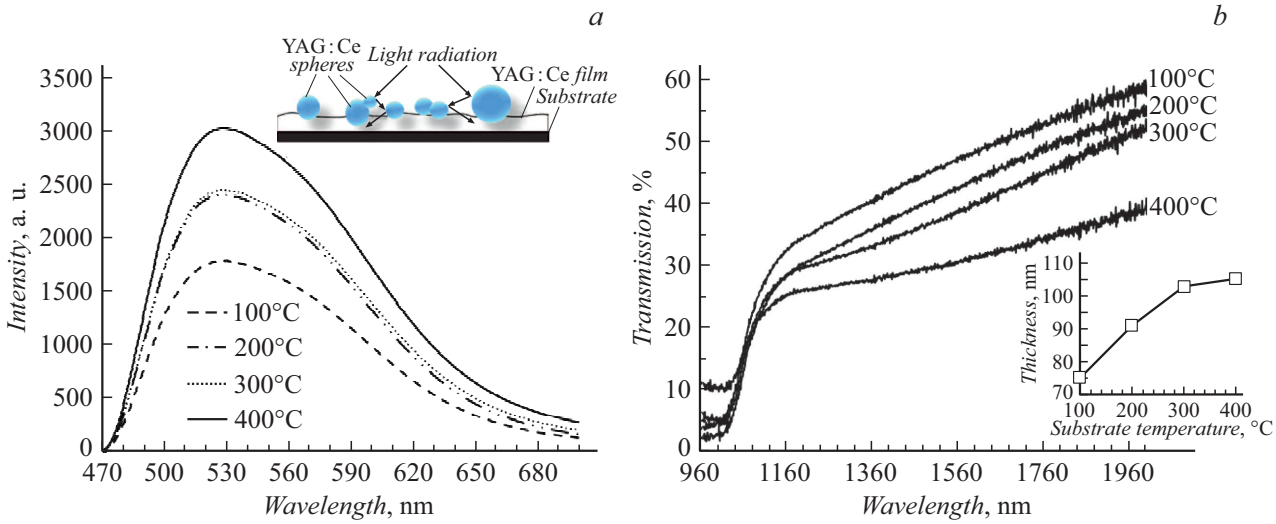


**Figure 3.** Luminescence ($a$) and IR transmission ($b$) spectra of thin YAG:Ce$^{3+}$ films after annealing.

the results of luminescence measurements, we concluded that an increase in substrate temperature during PLD does not affect the spectrum shape and does not induce the emergence of new luminescence bands, but does result in a significant enhancement of luminescence intensity. The transmission spectra of samples in the 960–1960 nm range (Fig. 3, $b$) revealed that the transmission coefficient decreases insignificantly as the substrate temperature grows within the 100–300°C interval, while the transmission of films at 400°C decreases by more than 5%. This agrees with the results of ellipsometry: as the substrate temperature varied from 100 to 400°C, the film growth rate increased, and the thickness of YAG:Ce$^{3+}$ films grew accordingly from 75 to 105 nm (inset in Fig. 3, $b$).

The obtained experimental data suggest that pulsed laser deposition allows one to fabricate thin polycrystalline YAG:Ce$^{3+}$ films. These films are characterized by a developed structured surface that enhances the efficiency of capture of excitation radiation due to rereflections (inset in Fig. 3, $a$). Although the annealing temperature remains the same, the luminescence intensity increases with substrate temperature maintained in the PLD process. This implies that temperature activation facilitates the embedding of activator ions (Ce$^{3+}$). The synthesized films may be used efficiently as luminescent markers (sensitive to blue and X-ray radiation) and in the fabrication of optical modifying converting coatings.

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## Conflict of interest

The authors declare that they have no conflict of interest.

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