

Scintillation properties of new ultrafast crystals Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂

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New crystal scintillators Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂ having ultrashort decay time were grown for the first time. The pulsed cathodoluminescence spectra of the new scintillators, light output, and their decay times have been investigated. It is shown that the decay time constant of the scintillation pulse of Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂ crystals is 0.9/6 ns and 0.7/4.8 ns, respectively.

Keywords: scintillating crystals, gamma radiation, scintillation time, pulsed cathodoluminescence.

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Inorganic scintillators based on Yb³⁺ charge transfer luminescence with ultrashort decay times are promising materials for future applications in high-energy physics and experimental setups with high-intensity X-ray free-electron lasers. It was demonstrated that Lu₂O₃:Yb ceramics have scintillation decay time $\tau = 1.1$ ns [1].

Current high-energy physics experiments at the CMS collider in CERN (Switzerland) utilize 78 000 scintillation modules based on PbWO₄ crystals with a light output of 25–35 photons/MeV [2]. This PbWO₄ crystal was chosen primarily for its short scintillation decay time and the low cost of production of PbWO₄ modules.

The scintillation decay time of PbWO₄ depends on small concentrations of impurities: for example, the decay times are $\tau_1 = 4.0$ ns (34 %), $\tau_2 = 17$ ns (54 %), and $\tau_3 = 129$ ns (12 %) for 0.5 at. ppm La and $\tau_1 = 3.7$ ns (84 %) and $\tau_2 = 53$ ns (16 %) for 85 at. ppm La [3].

The cost of producing scintillation modules from new crystals (ceramics) with ultra-short scintillation decay times will be important for future colliders. Highly efficient Ce:Lu₂SiO₅ crystals are inferior to ytterbium-based crystals due to the high cost of lutetium, since Yb₂O₃ is several times cheaper than Lu₂O₃. Another important advantage of the scintillator material is the feasibility of growth of large-diameter crystals (~ 100 mm [4]) and production of transparent optical ceramics [5]. Crystals with a garnet structure (specifically, Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂) satisfy these requirements.

In our earlier study [6], we have grown and examined a new laser garnet crystal Tm:Ho:Yb₃Al₅O₁₂. The growth technique was developed, and Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂ boules 80 mm in length with high optical quality were grown using the Czochralski method in a crucible 40 mm in diameter with radio frequency heating. The iridium purity was 99.989 %, and a protective atmosphere

of 99.9 % Ar+0.01 % O₂ was used. The growth rate was 6–7 mm/h; the rotation rate was 10 rpm. Crystal growth from a single-crystal seed with the (001) crystallographic orientation was considered optimal. The concentrations of ions Tm³⁺ and Ho³⁺ in the melt were 5.7 and 0.7 at.%, respectively. Spectral and luminescent characteristics were examined. Lasing of the Ho³⁺ ion in the Tm:Ho:YbAG crystal with $\lambda = 2100$ nm pumped at $\lambda = 1678$ nm to the absorption line at the ³H₆–³F₄ transition of the Tm³⁺ ion was observed in experiments. In the present study, we report the scintillation characteristics of ultrafast garnet crystals Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂ with a density of 6.6 g/cm³ and effective atomic number $Z = 57.9$. The spectra of absorption coefficients of polished Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂ crystal samples were measured at room temperature with a UNICO 2800 UV-VIS spectrophotometer (Fig. 1). The electron transitions found in the absorption spectrum originate at the ³H₆ ground state and end at higher-lying levels ³H₄, ³F₃, ¹G₄, ¹D₂, and ³P₂ of the Tm³⁺ ion with electron shell configuration ⁴f₁₂. In the Ho³⁺ ions, electrons transit from the ⁵I₈ ground state to higher-lying levels ⁵F₅, ⁵S₂, ⁵F₄, ⁵F₃, and ⁵G₅.

Pulsed cathodoluminescence (PCL) of crystals Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂ induced by a high-intensity (~ 10 MW/cm²) short (~ 2 ns) exposure to an electron beam with an average electron energy of 150 keV from the RADAN-EXPERT accelerator [7] was examined at room temperature. The electron beam with a diameter of 20 mm was directed from the top onto the sample under study. Emission from the sample entered a light guide located at a distance of 20 mm from the electron-irradiated surface of the crystal and was transported to the input of an OCEAN USB2000 spectrometer with a spectral range of 200–800 nm.

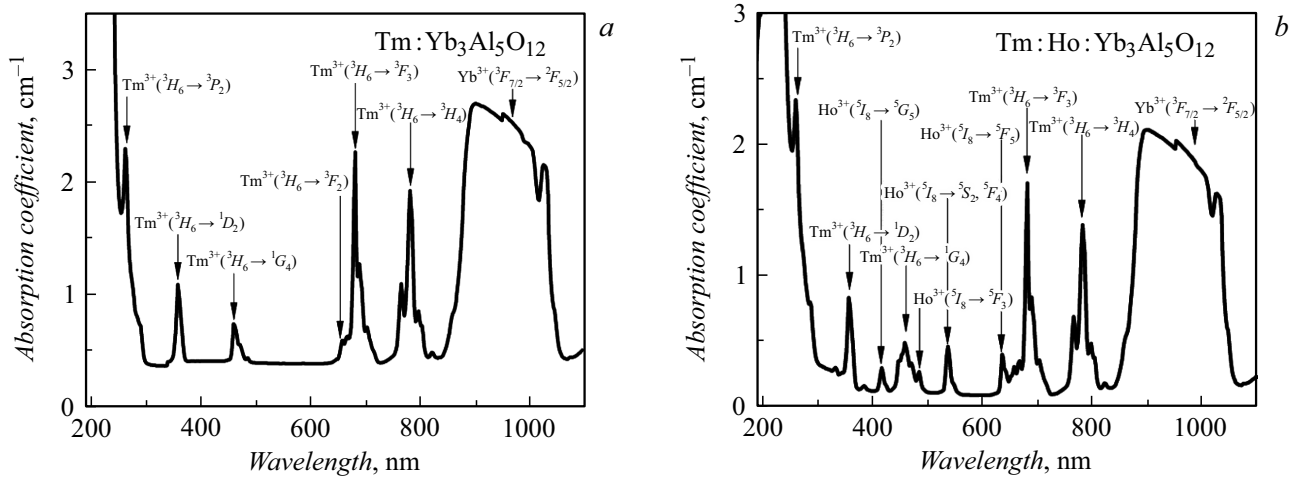


Figure 1. Absorption coefficient spectra for polished samples of crystals Tm:Yb₃Al₅O₁₂ (a) and Tm:Ho:Yb₃Al₅O₁₂ (b).

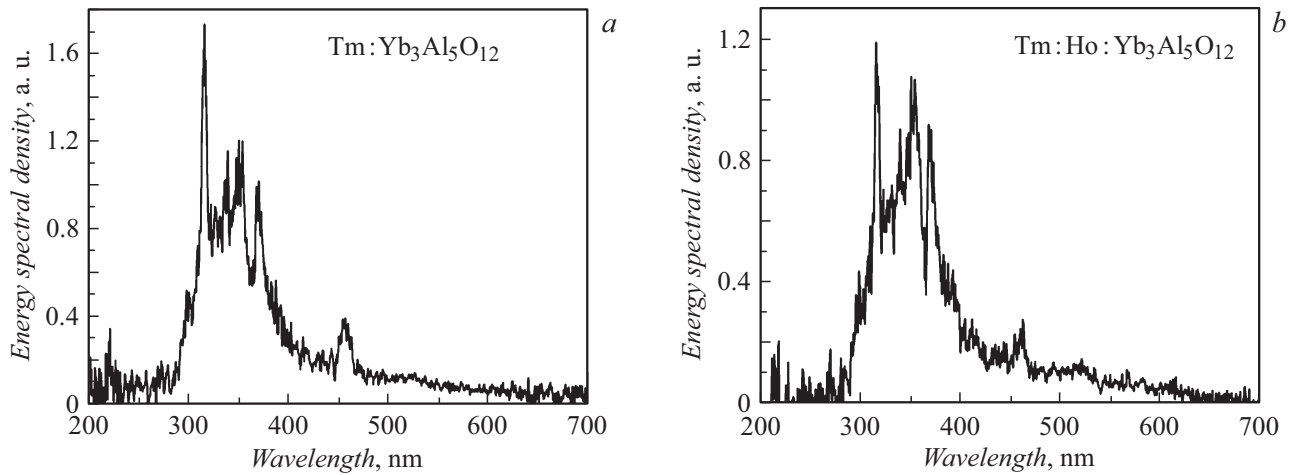


Figure 2. Room-temperature PCL spectra for crystals Tm:Yb₃Al₅O₁₂ (a) and Tm:Ho:Yb₃Al₅O₁₂ (b).

The spectral sensitivity of the OCEAN spectrometer together with a quartz fiber optic cable connected to it was calibrated using OCEAN DH3-plus reference halogen and deuterium lamp with their emission spectra known to an accuracy of $\sim 1\%$.

The obtained PCL spectra are the sum of intensities of all light signals emitted over 30 s and represent the result of averaging of 30 luminescence pulses (Fig. 2).

The well-known CeF₃ scintillation crystal with a light output of 2400 photons/MeV was used to estimate the light output of crystals Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂.

The scintillation crystal under examination was coupled to a Hamamatsu R4125Q photomultiplier with a quartz window using Dow Corning Q2-3067 optical grease. All the other faces of the crystal were wrapped in 8–10 layers of Teflon tape. Scintillation of the crystal was excited by a radioactive ¹³⁷Cs γ -source. The light output of the new crystals (in photons/MeV) was determined by comparing their photocurrent with the one from the CeF₃ crystal. Assuming

that the sensitivity of the photomultiplier to the emission of CeF₃ and the new crystals is the same (since their emission spectra are closely adjacent), we find that the light output of the Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂ crystals is 380 ± 65 and 340 ± 70 photons/MeV, respectively.

The delayed coincidence method was used to determine the decay time of crystals Tm:Yb₃Al₅O₁₂ and Tm:Ho:Yb₃Al₅O₁₂. This method consists in measuring the distribution of time intervals Δt between the excitation of trigger scintillator BaF₂ by gamma quanta of the ²²Na radioactive source (start channel) and the formation of a photoelectron at the PMT photocathode (stop channel) to which the crystal under study was attached. The signals from R4125Q PMTs of start and stop channels were fed to CAEN C808 constant fraction discriminators and then to a KA-317 2048-channel time-to-digital converter (TDC). The TDC output data was stored in the memory of a personal computer. Experimental data were approximated by a function in the form of a sum of two exponentials

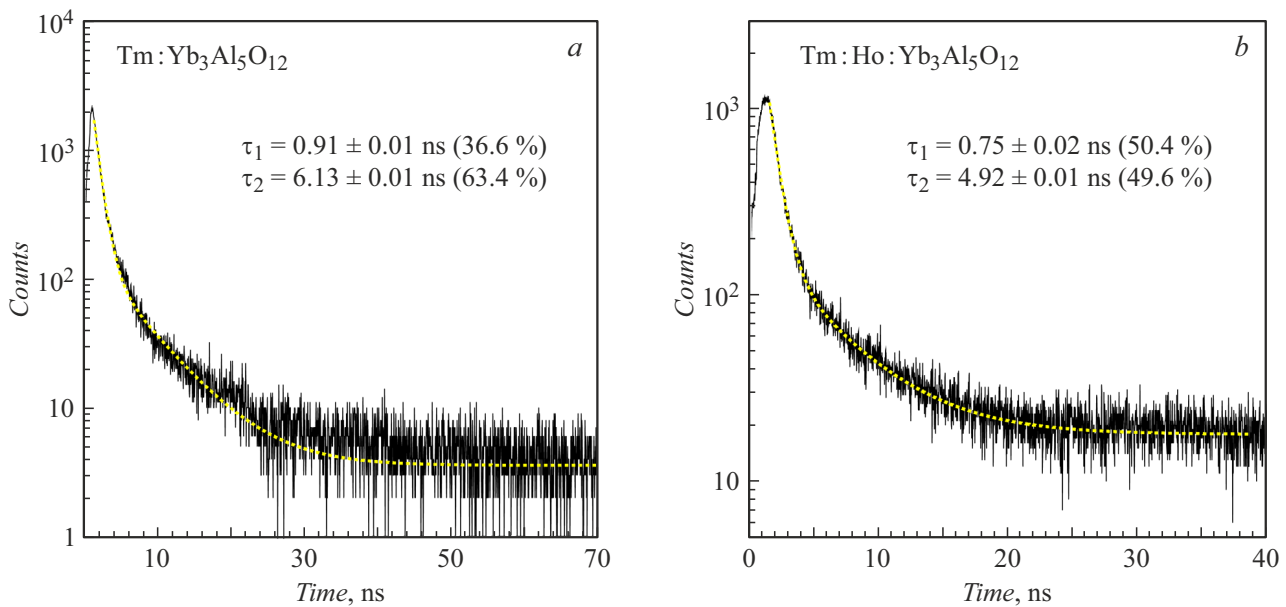


Figure 3. Time dependence of the emission intensity of crystals $\text{Tm:Yb}_3\text{Al}_5\text{O}_{12}$ (a) and $\text{Tm:Ho:Yb}_3\text{Al}_5\text{O}_{12}$ (b).

$I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) + I_0$, where A_1 and A_2 are the amplitudes of two components, τ_1 and τ_2 are their decay constants, and I_0 is the constant component. The contributions of components were calculated as the ratio of the component integral to the function integral. Figure 3 shows the time dependence of the emission intensity for the new crystals.

Comparing the PCL and absorption spectra of crystals $\text{Tm:Yb}_3\text{Al}_5\text{O}_{12}$ and $\text{Tm:Ho:Yb}_3\text{Al}_5\text{O}_{12}$, we find that scintillation-luminescence light with a maximum at 340 nm escapes the crystal without significant absorption by levels 3F_3 , 1G_4 , and 1D_2 of Tm ions and 5S_2 , 5F_4 , and 5G_5 of Ho ions within the 320–650 nm ytterbium scintillation range. This leads to a reduction in scintillation time, which is an important and novel result.

The results presented above suggest that additional doping with Tm and Ho ions should reduce the scintillation time of $\text{Lu}_2\text{O}_3\text{:Yb}$ ceramics.

Simple Lu_2O_3 and Yb_2O_3 oxides have a melting point above 2400 °C, while the melting point of crystals $\text{Tm:Yb}_3\text{Al}_5\text{O}_{12}$ and $\text{Tm:Ho:Yb}_3\text{Al}_5\text{O}_{12}$ is close to 1970 °C, which is a significant advantage in industrial production of bulk ceramic scintillation modules. These crystals are also easy to process mechanically and polish.

New fast $\text{Tm:Yb}_3\text{Al}_5\text{O}_{12}$ and $\text{Tm:Ho:Yb}_3\text{Al}_5\text{O}_{12}$ scintillators hold promise for application in experiments at the new Russian NICA (Nuclotron-based Ion Collider fAcility, Joint Institute for Nuclear Research, Dubna) collider.

Conflict of interest

The authors declare that they have no conflict of interest.

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