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# Scintillation properties of new ultrafast crystals $Tm{:}Yb_3Al_5O_{12}$ and $Tm{:}Ho{:}Yb_3Al_5O_{12}$

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New crystal scintillators $Tm{:}Yb_3Al_5O_{12}$ and $Tm{:}Ho{:}Yb_3Al_5O_{12}$ 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 $Tm{:}Yb_3Al_5O_{12}$ and $Tm{:}Ho{:}Yb_3Al_5O_{12}$ 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 $Yb^{3+}$ 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 $Lu_2O_3{:}Yb$ 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 $PbWO_4$ crystals with a light output of 25–35 photons/MeV [2]. This $PbWO_4$ crystal was chosen primarily for its short scintillation decay time and the low cost of production of $PbWO_4$ modules.

The scintillation decay time of $PbWO_4$ 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 $Ce{:}Lu_2SiO_5$ crystals are inferior to ytterbium-based crystals due to the high cost of lutetium, since $Yb_2O_3$ is several times cheaper than $Lu_2O_3$. Another important advantage of the scintillator material is the feasibility of growth of large-diameter crystals ($\sim$ 100 mm [4]) and production of transparent optical ceramics [5]. Crystals with a garnet structure (specifically, $Tm{:}Yb_3Al_5O_{12}$ and $Tm{:}Ho{:}Yb_3Al_5O_{12}$) satisfy these requirements.

In our earlier study [6], we have grown and examined a new laser garnet crystal $Tm{:}Ho{:}Yb_3Al_5O_{12}$. The growth technique was developed, and $Tm{:}Yb_3Al_5O_{12}$ and $Tm{:}Ho{:}Yb_3Al_5O_{12}$ 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 % $O_2$ 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 $Tm^{3+}$ and $Ho^{3+}$ in the melt were 5.7 and 0.7 at.%, respectively. Spectral and luminescent characteristics were examined. Lasing of the $Ho^{3+}$ ion in the Tm:Ho:YbAG crystal with $\lambda = 2100$ nm pumped at $\lambda = 1678$ nm to the absorption line at the $^3H_6{-}^3F_4$ transition of the $Tm^{3+}$ ion was observed in experiments. In the present study, we report the scintillation characteristics of ultrafast garnet crystals $Tm{:}Yb_3Al_5O_{12}$ and $Tm{:}Ho{:}Yb_3Al_5O_{12}$ with a density of 6.6 g/cm$^3$ and effective atomic number $Z = 57.9$. The spectra of absorption coefficients of polished $Tm{:}Yb_3Al_5O_{12}$ and $Tm{:}Ho{:}Yb_3Al_5O_{12}$ 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 $^3H_6$ ground state and end at higher-lying levels $^3H_4$, $^3F_3$, $^1G_4$, $^1D_2$, and $^3P_2$ of the $Tm^{3+}$ ion with electron shell configuration $^4f_{12}$. In the $Ho^{3+}$ ions, electrons transit from the $^5I_8$ ground state to higher-lying levels $^5F_5$, $^5S_2$, $^5F_4$, $^5F_3$, and $^5G_5$.

Pulsed cathodoluminscescence (PCL) of crystals $Tm{:}Yb_3Al_5O_{12}$ and $Tm{:}Ho{:}Yb_3Al_5O_{12}$ induced by a high-intensity ($\sim$ 10 MW/cm$^2$) short ($\sim$ 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.

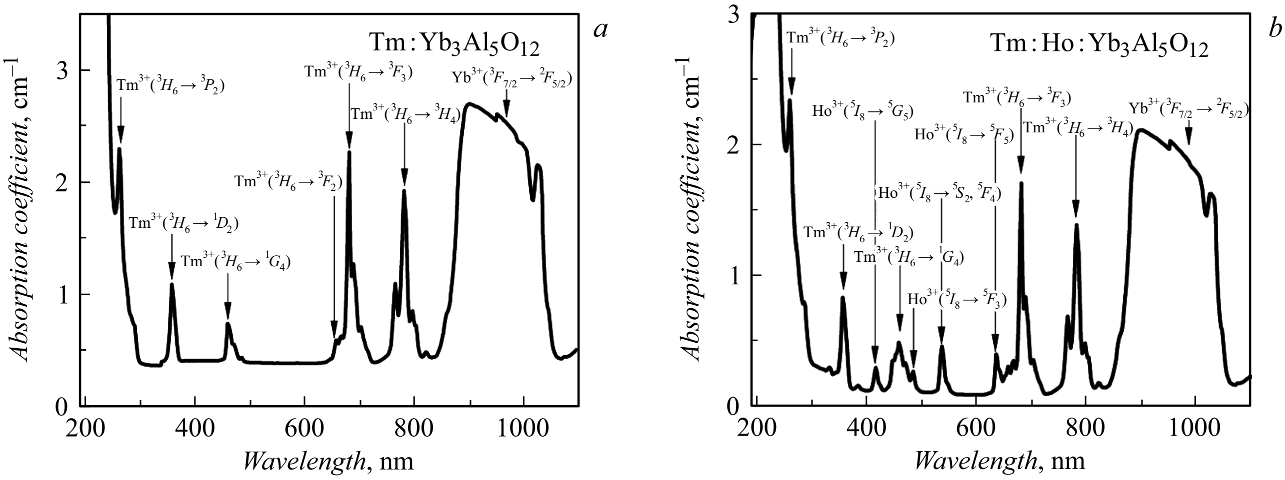

**Figure 1.** Absorption coefficient spectra for polished samples of crystals Tm:$Yb_3Al_5O_{12}$ (*a*) and Tm:Ho:$Yb_3Al_5O_{12}$ (*b*).

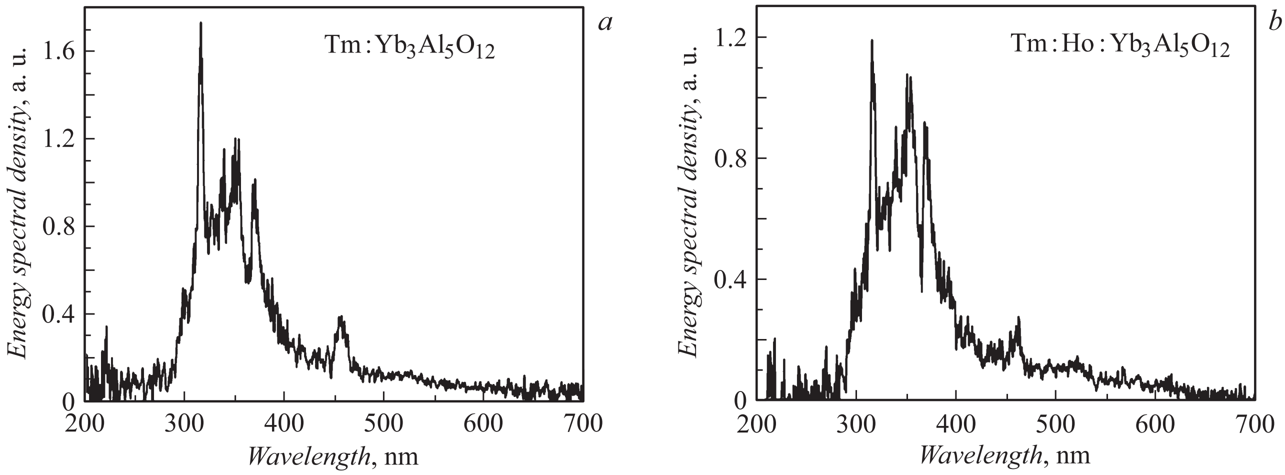

**Figure 2.** Room-temperature PCL spectra for crystals Tm:$Yb_3Al_5O_{12}$ (*a*) and Tm:Ho:$Yb_3Al_5O_{12}$ (*b*).

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 $CeF_3$ scintillation crystal with a light output of 2400 photons/MeV was used to estimate the light output of crystals Tm:$Yb_3Al_5O_{12}$ and Tm:Ho:$Yb_3Al_5O_{12}$.

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 $^{137}$Cs $\gamma$-source. The light output of the new crystals (in photons/MeV) was determined by comparing their photocurrent with the one from the $CeF_3$ crystal. Assuming that the sensitivity of the photomultiplier to the emission of $CeF_3$ and the new crystals is the same (since their emission spectra are closely adjacent), we find that the light output of the Tm:$Yb_3Al_5O_{12}$ and Tm:Ho:$Yb_3Al_5O_{12}$ crystals is $380 \pm 65$ and $340 \pm 70$ photons/MeV, respectively.

The delayed coincidence method was used to determine the decay time of crystals Tm:$Yb_3Al_5O_{12}$ and Tm:Ho:$Yb_3Al_5O_{12}$. This method consists in measuring the distribution of time intervals $\Delta t$ between the excitation of trigger scintillator $BaF_2$ by gamma quanta of the $^{22}$Na 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

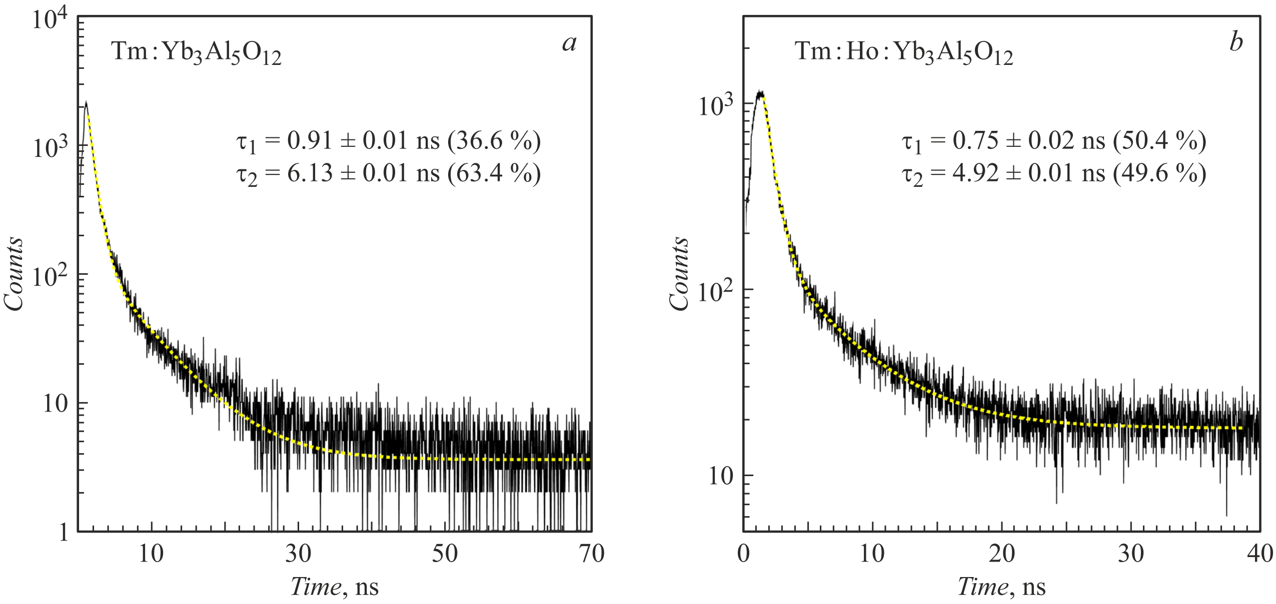

**Figure 3.** Time dependence of the emission intensity of crystals Tm:$Yb_3Al_5O_{12}$ (*a*) and Tm:Ho:$Yb_3Al_5O_{12}$ (*b*).

$I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) + I_0$, where $A_1$ and $A_2$ are the amplitudes of two components, $\tau_1$ and $\tau_2$ 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 Tm:$Yb_3Al_5O_{12}$ and Tm:Ho:$Yb_3Al_5O_{12}$, 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 $Lu_2O_3$:Yb ceramics.

Simple $Lu_2O_3$ and $Yb_2O_3$ oxides have a melting point above 2400 °C, while the melting point of crystals Tm:$Yb_3Al_5O_{12}$ and Tm:Ho:$Yb_3Al_5O_{12}$ 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 Tm:$Yb_3Al_5O_{12}$ and Tm:Ho:$Yb_3Al_5O_{12}$ 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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