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Lasing characteristics of $ZrO_2 - Y_2O_3 - Ho_2O_3$ crystal

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Abstract. **The spectral dependences of the gain cross section of the $^5I_8 \to {}^5I_7$, $^5I_7 \to {}^5I_8$ transition of Ho^{3+} ions in the ZrO_2 – 13.6 mol % Y_2O_3 – 0.4 mol % Ho_2O_3 crystal are calculated at different relative population inversions using the absorption and luminescence spectra of the $^5I_8 \to {}^5I_7$ and $^5I_7 \to {}^5I_8$ transitions of Ho^{3+} ions at T = 300 K. Lasing of these crystals at the $^5I_7 \to {}^5I_8$ transition is obtained for the first time under pumping by a Tm : $YLiF_4$ laser (λ_p = 1.905 μm). The lasing wavelength is 2.17 μm.**

Keywords: *absorption spectrum, luminescence spectrum, lasing, Ho^{3+} ions, $ZrO_2 - Y_2O_3 - Ho_2O_3$ crystals.*

The disordered structure of zirconium dioxide crystals stabilised with yttrium leads to a considerable inhomogeneous broadening of the absorption and luminescence lines of rare-earth dopants. This allows one to consider these crystals as active media of tunable lasers and as promising materials for development of pico- and femtosecond lasers.

The spectral and luminescent characteristics of stabilised zirconium dioxide crystals doped with Nd^{3+} and Yb^{3+} ions were studied in [1, 2]. The authors of [3, 4] published the results of laser experiments with $ZrO_2 - Y_2O_3 - Yb_2O_3$ crystals. In [5], we obtained two-micron lasing at the $^3F_4 \to {}^3H_6$ transition of Tm^{3+} ions in the $ZrO_2 - Y_2O_3 - Tm_2O_3$ crystals under diode laser pumping of the $^3H_4$ level of Tm^{3+} ions.

Lasers emitting in the two-micron spectral region are of interest for application in medicine and lidars. Laser radiation in this spectral region is obtained at the $^3F_4 \to {}^3H_6$ transition of Tm^{3+} ions and the $^5I_7 \to {}^5I_8$ transition of Ho^{3+} ions in various oxide and fluoride crystals. The characteristics of two-micron lasers based on oxide and fluoride materials doped with Tm^{3+} and Ho^{3+} ions are presented in review [6]. Investigation of the spectral and luminescent characteristics of various crystals and glasses doped with Ho^{3+} ions [6 – 8] show that the longest wavelength of laser radiation at the $^5I_7 \to {}^5I_8$ transition of Ho^{3+} ions is obtained in Ho : $Y_3Al_5O_{12}$ [6] and Ho : Lu_2O_3 crystals (2.12 μm) pumped by diode lasers and a thulium fibre laser [8]. Papers [9 – 12] report the development of two-micron fibre lasers with the longest wavelength of 2.15 μm.

In the present work, using the spectral dependences of the absorption and luminescence cross sections of the $^5I_8 \to {}^5I_7$ and $^5I_7 \to {}^5I_8$ transitions of Ho^{3+} ions in the crystals of the composition ZrO_2 – 13.6 mol % Y_2O_3 – 0.4 mol % Ho_2O_3, we obtained the spectral dependence of the gain cross section of the $^5I_7 \to {}^5I_8$ transition of Ho^{3+} ions in these crystals and performed laser experiments under pumping by a Tm : $YLiF_4$ laser.

The zirconium dioxide crystals stabilised with yttrium dioxide and doped with Tm^{3+} were grown in a 'Kristall-407' apparatus. The crystals of the composition ZrO_2 – 13.6 mol % Y_2O_3 – 0.4 mol % Ho_2O_3 were synthesised in a cold container 130 mm in diameter with a growth rate of 10 mm h^{-1}. The grown crystals were 40 – 50 mm long and 10 – 20 mm in cross section. From these crystals, we cut plane-parallel plates for spectral and luminescent investigations and active elements in the form of rectangular parallelepipeds for laser experiments. The concentration of Ho^{3+} ions in the crystals was 2.1×10^{20} cm^{-3}.

The absorption spectra of Ho^{3+} ions were recorded with a Lambda 950 (PerkinElmer) spectrophotometer. The spectra of luminescence from the $^5I_7$ level of Ho^{3+} ions were recorded using a computer-assisted setup based on an MDR-23 monochromator upon excitation to the $^5S_2$ level of Ho^{3+} ions by the second harmonic (532 nm) of a Nd : YVO_4 laser. As a detector, we used a PbS photoresistor.

An important characteristic of active laser media is the spectral dependence of the gain cross section σ_g of the laser transition of the rare-earth dopant ion. This characteristic allows one to estimate the wavelength region of laser radiation at this transition and is determined by the formula

$$\sigma_g = p\sigma_{em} - (1 - p)\sigma_{ab}, \qquad (1)$$

where σ_{em} is the luminescence cross section at the chosen wavelength, σ_{ab} is the absorption cross section at the same wavelength, $p = N_{ex}/(N_{ex} + N_f)$ is the relative population inversion, N_{ex} is the population of the upper laser level $^5I_7$ of Ho^{3+} ions, and $N_{ex} + N_f$ is the total number of particles (concentration of active ions).

The spectral dependence of the absorption cross section for the $^5I_8 \to {}^5I_7$ transition of Ho^{3+} ions was found by the formula

$$\sigma_{ab} = k(\lambda)/N, \qquad (2)$$

where $k(\lambda)$ is the absorption coefficient for the wavelength λ, and N is the concentration of Ho^{3+} ions.

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The spectral dependence of the luminescence cross section of the $^5I_7 \rightarrow {}^5I_8$ laser transition of Ho^{3+} ions was calculated by the Füchtbauer–Ladenburg formula

$$\sigma_{em}(\lambda) = \frac{\lambda^5}{8\pi c n^2} \frac{1}{\tau_{rad}} \frac{I(\lambda)}{\int \lambda I(\lambda) d\lambda}. \quad (3)$$

Here, τ_{rad} is the radiative lifetime of the $^5I_7$ level of Ho^{3+} ions, n is the refractive index of the material, and I is the luminescence intensity in relative units. Assuming that the probability of nonradiative relaxation from the $^5I_7$ level of Ho^{3+} ions is low, we used the value $\tau_{rad} = 1/A = 14.7$ ms, where

$$A = \frac{8\pi n_\lambda^2 c}{N\lambda^4} \frac{2J^{(1)}+1}{2J+1} \int k(\lambda) d\lambda \quad (4)$$

is the probability of the radiative transition from the level [$J^{(1)}$ and J are the total angular momenta of 4f electrons in the ground and excited states involved in the transition].

Figure 1 shows the absorption and luminescence cross sections for the $^5I_7 \leftrightarrow {}^5I_8$ transition of Ho^{3+} ions in the ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystal at T = 300 K, while Fig. 2 presents the spectral dependence of the gain cross section of the $^5I_7 \rightarrow {}^5I_8$ laser transition calculated by formula (1).

To perform laser experiments, we assembled a scheme given in Fig. 3. The scheme includes a Tm : $YLiF_4$ pump laser, whose active element (*4*) had a shape of a rectangular parallelepiped 3 × 3 × 10 mm in size with the faces antireflection coated for the laser wavelength 1.9 μm. The copper holder of the active element was thermostated at $T \approx 18$ °C. The pump laser cavity was formed by a spherical highly reflecting mirror (*3*) (R = 100 mm) and a plane output mirror (*6*) with a transmission coefficient of 4%. To match the pump laser spectrum with the absorption spectrum of Ho^{3+} ions, we placed into the laser cavity a Lyot filter (*5*), which allowed us to tune the laser wavelength within the range 1900–1930 nm. The Tm : $YLiF_4$ laser was pumped by a fibre-coupled (fibre diameter 400 μm) laser diode array (*1*). The pumped spot 600 μm in diameter was formed by lenses (*2*).

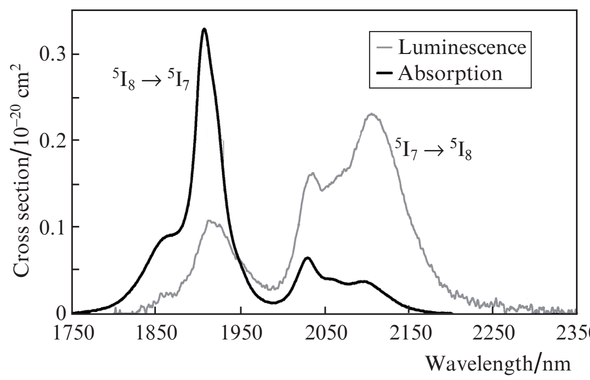

Figure 1. Spectral dependences of the absorption and luminescence cross sections for the $^5I_7 \rightarrow {}^5I_8$ transition of Ho^{3+} ions in the ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystals at T = 300 K.

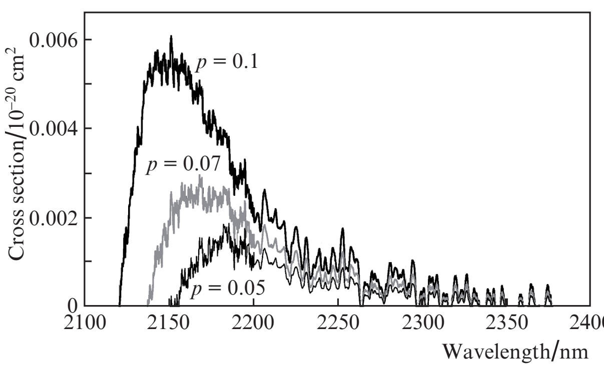

Figure 2. Spectral dependences of the gain cross section of the $^5I_7 \rightarrow {}^5I_8$ laser transition of Ho^{3+} ions in the ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystals at different population inversion p.

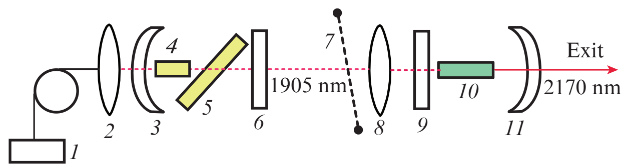

Figure 3. Optical scheme of the experiment with a laser based on the ZrO_2–13.6 mol % Y^2O_3–0.4 mol % Ho_2O_3 crystal.

An active rod (*10*) made of a ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystal had dimensions 3 × 3 × 20 mm and antireflection coatings for the laser wavelength ($\lambda_{gen} \sim$ 2.15 μm). The pump laser beam was focused into the active element by a lens (*8*) so that the pump beam waist was 300 μm. To decrease the thermal load on the active element, we used a chopper (*7*), which formed pump pulses with a duration of 30 ms and a repetition rate of ~3 Hz. We used a semiconfocal cavity formed by a plane mirror (*9*) (transmission coefficient in the pump wavelength region no less than 60%, reflection coefficient at λ_{gen} higher than 99%) and a spherical output mirror (*11*) (R = 100 mm) with a transmission coefficient at λ_{gen} below 1%.

Lasing at the $^5I_7 \rightarrow {}^5I_8$ transition of Ho^{3+} ions in the ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystal was obtained at the wavelength λ_{gen} = 2.17 nm. The laser threshold absorbed pump power was 380 mW; the laser spectrum is shown in Fig. 4.

The oscillograms of luminescence and laser pulses recorded using a GDS 720C digital oscilloscope are presented in Fig. 5.

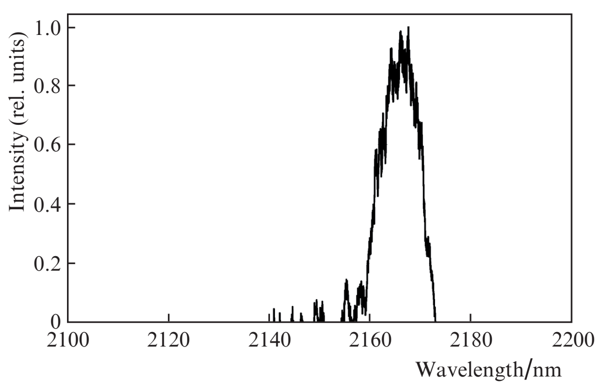

Figure 4. Spectrum of laser radiation at the $^5I_7 \rightarrow {}^5I_8$ transition of Ho^{3+} ions in the ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystals.

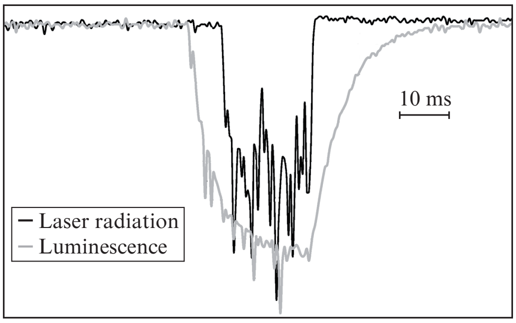

Figure 5. Oscillograms of luminescence and laser pulses of the ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystals.

Thus, in this work we received the spectral dependences of the absorption and luminescence cross sections for the $^5I_8 \leftrightarrow {}^5I_7$ transitions of Ho^{3+} ions in the ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystal. The spectral dependence of the gain cross section for the $^5I_7 \rightarrow {}^5I_8$ transition is calculated, which allowed us to determine the possible range of laser wavelength tuning to be 2120–2200 nm. Lasing at the $^5I_7 \rightarrow {}^5I_8$ transition (2.17 μm) of Ho^{3+} ions in these crystals pumped by a Tm:$YLiF_4$ laser is obtained for the first time. It should be noted that the wavelength of the laser based on the ZrO_2–13.6 mol % Y_2O_3–0.4 mol % Ho_2O_3 crystal is the longest among the wavelengths of all known solid-state lasers based on Ho^{3+}-doped crystals.

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