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LASERS

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Lasing in a Tm:Ho:$Yb_3Al_5O_{12}$ crystal pumped into the $^3H_6-^3F_4$ transition

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Abstract. **A growth technology has been developed, and a Tm:Ho:$Yb_3Al_5O_{12}$ laser crystal of high optical quality has been grown by Czochralski method. Its spectral and luminescent characteristics are studied. Lasing at a wavelength of 2100 nm is obtained under pumping into the absorption line on the $^3H_6-^3F_4$ transition of the Tm^{3+} ion at a wavelength of 1678 nm. The slope and total (optical) efficiencies of the laser at an output power of up to 320 mW reach 41 % and 30 %, respectively.**

Keywords: *Tm, Ho, YbAG, laser, luminescence, lasing, lifetime.*

1. Introduction

Two-micron lasers based on crystalline active elements doped with Tm^{3+} and Ho^{3+} ions are effectively used in various fields of science and technology [1–3]. In order to further improve the characteristics of these lasers, an active search for new crystal matrices is being conducted. For example, recently we have studied new Tm:SSO and Tm:Ho:SSO scandium silicate (Sc_2SiO_5) crystals and a Tm:YbAG crystal, which demonstrated high lasing efficiency [4, 5].

Our attention has been drawn to the previously unstudied (as an active laser element) $Yb_3Al_5O_{12}$ crystal doped with Tm^{3+} and Ho^{3+} ions (Tm:Ho:YbAG). This crystal is an analogue of the YAG crystal with replacement of the Y atom by Yb. Its growth technology at moderate temperatures is relatively simple and is combined with a high growth rate. Unlike the YAG crystal, in the YbAG matrix an additional energy level exists at $E \approx 10000$ cm^{-1}, which can create a channel of excitation energy decay.

In this paper we report for the first time new regimes of the Tm:Ho:YbAG laser crystal growth and present the results of study of its laser characteristics under pumping into the $^3H_6-^3F_4$ transition of the Tm^{3+} ion at the wavelength λ = 1678 nm. As a pump source we use a single-mode erbium-doped fibre laser with Raman wavelength conversion.

2. Laser crystal

Tm:Ho:YbAG crystals were grown in a 'Kristall-2' industrial facility by the Czochralski method. The crystal growth using a single crystal seed with the ⟨001⟩ crystallographic orientation was considered optimal. The concentrations of Tm^{3+} and Ho^{3+} ions in the melt were 5.7 at. % and 0.7 at. %, respectively. The crystal growth rate was 6–7 mm h^{-1} at a seed rotation rate of 10 revolutions per minute. The melt was in an iridium crucible with an outer diameter of 40 mm.

The above crystal growth parameters were determined in experiments on their optimisation. It was found that the growth rate of the Tm:Ho:YbAG boule can reach 6–7 mm h^{-1} without deteriorating the optical quality of the crystal, while for the widely used YAG crystal it is 0.5 mm h^{-1}. A higher growth rate of the Tm:Ho:YbAG crystal is explained by the use of the optimised composition of the melt: $\{Yb_{2.784}Tm_{0.192}Ho_{0.024}\}[Yb_{0.02}Al_{1.98}]Al_3O_{12}$ crystals have a cubic space group symmetry and therefore are isotropic, which permits an industrial growth of large crystals with a diameter up to 80 mm.

In the experiments we sought for an optimal composition of the growth atmosphere. To reduce the loss of iridium due to evaporation from crucible walls, use was made of a gas atmosphere with a minimum oxygen content, because even a low concentration of oxygen leads to the destruction of the walls of costly iridium crucibles. However, from a chemical point of view, it is desirable to have some amount of oxygen in the atmosphere in order to prevent the dissociation of the melt and the partial loss of oxygen from the melt at high temperatures. As a result, the Tm:Ho:YbAG crystal was grown in nitrogen atmosphere (99.5 %) with a small amount of oxygen (0.5 %) at a melting temperature of about 2000 °C. Under these conditions, the formation of macroscopic defects in the form of gas bubbles is completely eliminated. From the grown crystals we cut samples with polished end faces, a study of which showed their high optical quality and a complete lack of microscopic defects.

3. Spectral characteristics

The diagram of low-lying levels of Tm^{3+}, Ho^{3+} and Yb^{3+} ions is presented in Fig. 1. This diagram shows the wavelengths of the pump (1678 nm), luminescence of the Yb^{3+} ion (1031 nm) and lasing (2100 nm), as well as the transitions due to the transfer of energy between the ions of Tm^{3+} (arrow 1), Tm^{3+} and Yb^{3+} (arrows 2) and Tm^{3+} and Ho^{3+} (arrow 3).

The use of an YbAG crystal matrix in lasers based on thulium and holmium ions stipulates the presence of the $^2F_{5/2}$ level in the energy diagram. However, this level is not involved in the lasing, but can create additional channels of pump energy decay. Luminescence was observed at λ = 1031 nm, but its intensity was small compared with the luminescence intensity at $\lambda \approx$ 2100 nm. We assume that the channels of

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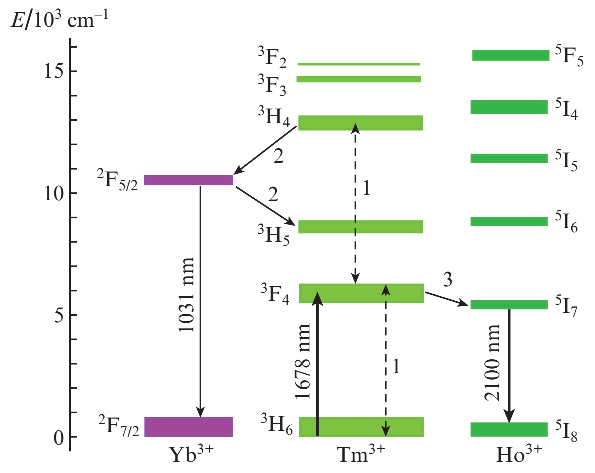

Figure 1. Diagram of low-lying levels of Tm^{3+}, Ho^{3+} and Yb^{3+}.

excitation energy decay produced by the ytterbium level should not significantly impair the lasing efficiency because under pumping into the $^3H_6-^3F_4$ transition band of the thulium ion the $^2F_{5/2}$ level of the ytterbium ion can be populated only by means of up-conversion transitions and thereafter $^3F_4$ (Tm) and $^5I_6$ (Ho) levels will be partially populated due to the energy of the $^2F_{5/2}$ (Yb) level.

Apart from the transitions shown in the diagram, other interionic transitions participate in the energy exchange (see, for example, [6, 7]). In particular, the population of the $^2F_{5/2}$ (Yb) level suggests the possibility of interionic cascade transition $^2F_{5/2}(Yb) + {^5I_8}(Ho) \to {^2F_{7/2}}(Yb) + {^5I_6}(Ho)$ and $^5I_6(Ho) + {^5I_6}(Ho) \to {^5S_2}(Ho) + {^5I_8}(Ho)$, as was shown for the Yb : Ho : YLF crystal in [6]. All these transitions contribute to the up-conversion energy loss and reduce the efficiency of the laser, but, as was shown by our study, the presence of the Yb^{3+} ion in the matrix does not significantly impairs the lasing efficiency.

The absorption spectrum of the crystal under study is shown in Fig. 2. The scale of the vertical axis is chosen such that the parts of the spectrum containing absorption lines of Tm^{3+} and Ho^{3+} ions are shown in greater detail, the absorption peaks of Yb^{3+} being outside the boundaries of the figure.

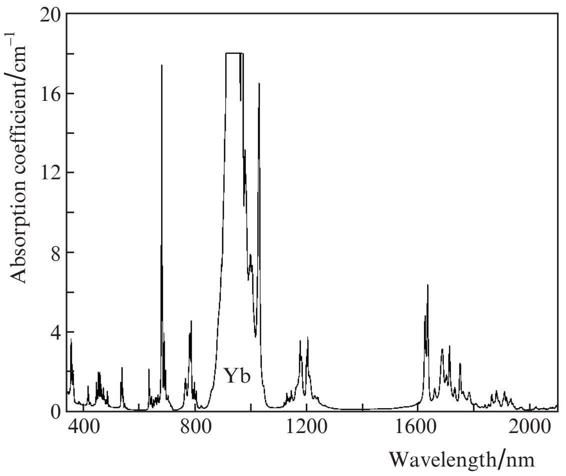

Figure 2. Absorption spectrum of the Tm : Ho : YbAG crystal. The absorption coefficient of the Yb^{3+} ion in the maximum exceeds 68 cm^{-1}.

The measurements were carried out using a Shimadzu UV-3600 spectrophotometer. About 60 % of the pump power was absorbed in the length of the active element. It should be noted that the pump wavelength of 1678 nm used in our experiments does not coincide with the wavelength of the maximum of the strongest absorption line and, hence, is not optimal for achieving the highest efficiency of a quasi-three-level laser. Thus, the laser efficiency can be increased by choosing a laser pump source compatible with the most intense absorption line.

The luminescence spectra of the active element of the crystal were recorded at the excitation wavelength $\lambda = 1678$ nm. The active element was placed in front of the slit of an MDR-204 monochromator (LOMO-Fotonika). The optical scheme of the experiment was identical to that described in [5]. This geometry ensured minimal distortions of luminescence spectra caused by light reabsorption at the exit from the crystal.

The radiation was detected in the wavelength range 1000–2200 nm by a G8373-01 photodiode (Hamamatsu). The pump radiation was modulated by an MC2000 optical chopper (Thorlabs) within the modulation frequency range 2–1000 Hz. The signal from the photodiodes was amplified by a SR830 lock-in amplifier (Stanford Research Systems) and recorded by a personal computer using the MDR-204 monochromator software. Figure 3 shows the luminescence, lasing and absorption spectra of the Tm : Ho : YbAG crystal in the IR region. A slight shift in the wavelengths of the absorption and luminescence peaks is caused by the error in the measurement of wavelengths by the monochromator, which was about 1.6 nm. The widths of the input and output slits of the monochromator were equal to 100 μm.

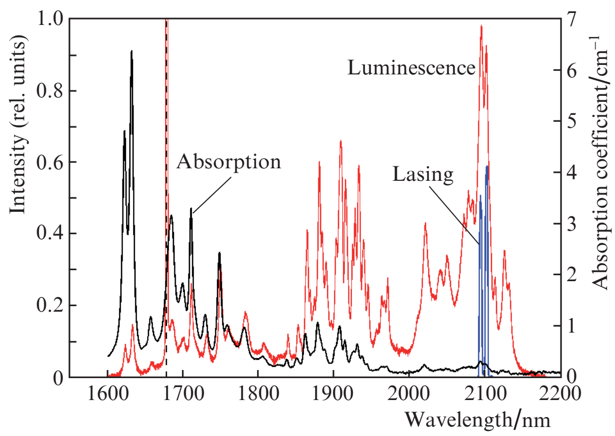

Figure 3. Luminescence, lasing and absorption spectra for the Tm : Ho : YbAG crystal in the IR region. The vertical dashed line shows the pump wavelength (1678 nm).

The IR luminescence spectrum lies in the wide range (~1600–2150 nm). The laser wavelength of 2100 nm almost coincides with the wavelength of the intense luminescence line. Because the absorption of the active element at wavelengths longer than 2020 nm is insignificant, one can expect that the laser wavelength can be tuned in a wide range when using a spectrally selective cavity.

The two-micron luminescence decay time τ_{lum} of the Tm : Ho : YbAG crystal was measured by the method described in [5] upon pump beam modulation by a disk chopper in the frequency range 2–1000 Hz. In the crystal in question the luminescent level $^5I_7(Ho^{3+})$ is populated not directly by pump radiation, but through the $^3F_4(Tm^{3+}) \leftrightarrow {}^5I_7(Ho^{3+})$ transition so that the luminescence decay time is determined by the lifetimes of both levels involved in the process. The measured time $\tau_{lum} = 3.8 \pm 0.14$ ms. Within the approach used it is impossible to separate the contributions of the lifetimes of different levels in the resulting luminescence decay time.

4. Laser experiments

Lasing in the Tm:Ho:YbAG crystal was achieved using a nearly semi-concentric cavity. The active element was pumped through a highly reflecting plane dichroic mirror. A spherical output mirror with a radius of curvature of 50 mm had a reflection coefficient of 98% at the laser wavelength. The active element was placed near the plane mirror. The pump radiation was focused by a lens with a focal length of 80 mm inside the crystal into a spot having a diameter of ~80 μm. The active element measuring 3 × 3 × 3 mm and cut along the [001] crystallographic direction was mounted using indium foil on a copper heat sink. The working faces of the active element were not AR-coated. In most experiments the temperature of the active element was maintained at 20 °C.

Lasing in the Tm:Ho:YbAG crystal was obtained at λ = 2100 nm. Figure 4 shows the dependences of the output power of the Tm:Ho:YbAG laser on the pump power absorbed by the active element. Measurements were performed in pulsed and cw pump regimes. The pulsed regime was obtained using a disk chopper, which was open for 1/20 of the rotation period and rotated with a frequency of 10 Hz.

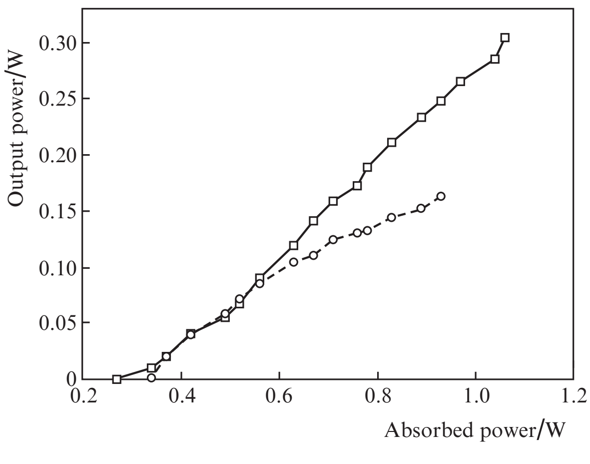

Figure 4. Dependences of the output power of the Tm:Ho:YbAG laser on the pump power absorbed in the active element in the pulsed (□) and cw (○) regimes.

The lowest threshold pump power was obtained in the case of pulsed pumping and was ~270 mW. For pulsed pumping we observed a linear increase in the output radiation intensity over the entire range of the used powers (up to 1 W). The slope and total (optical) efficiencies of the laser reached 41% and 30%, respectively, at an output power up to 320 mW, which was limited only by the maximum output power of our pump laser.

It should be noted that within the experimental error the Tm:Ho:YbAG crystal demonstrates the lasing efficiency that is comparable with that of the Tm:YbAG crystal [5]. This result, obtained under identical experimental conditions, indicates the high efficiency of the energy transfer of Tm^{3+} ions to Ho^{3+} ions. Note also that the presence of the energy level $^2F_{5/2}$ of the Yb^{3+} ion has no significant effect on the efficiency of energy transfer from the Tm^{3+} ion to the Ho^{3+} ion.

In the cw pump regime at a slight excess of the threshold (approximately twofold) the laser efficiency is almost the same as in the pulsed regime. With increasing pump power, the efficiency of cw lasing is 20%–25% less than that in the pulsed regime. The maximum power of the laser in the cw regime reached 180 mW at a slope and total laser efficiencies of 30% and 18%, respectively. A decrease in the lasing efficiency, which was observed in the cw regime, can be due to the heating of the channel inside the crystal by pump radiation. This heating increases the intracavity losses of the laser because lasing is realised according to the quasi-three-level scheme.

Figure 5 shows the lasing spectra of the Tm:Ho:YbAG crystal at different pump powers. The spectra consist of one or two lines, depending on the pump power. When the pump power exceeds the threshold, the lasing line wavelength is 2100 nm with a line width of ~10 nm.

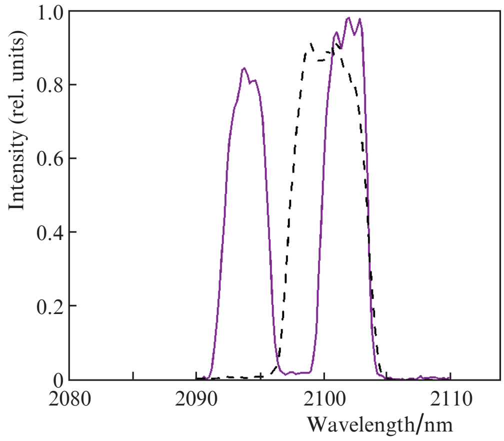

Figure 5. Lasing spectra at a pump power of 0.4 (solid curve) and 0.8 W (dashed curve).

5. Conclusions

Thus, we have grown and investigated a new Tm:Ho:YbAG laser crystal. It is shown that the crystal growth rate can be large enough and, in this respect, the crystal is superior to most known laser crystals.

We have obtained lasing of the Ho^{3+} ion in the Tm:Ho:YbAG crystal with λ = 2100 nm, pumped at λ = 1678 nm into the absorption line on the $^3H_6-^3F_4$ transition of the Tm^{3+} ion. The lasing threshold with respect to the pump power is equal to approximately 180 mW. The slope and total efficiencies of the laser are quite large, reaching 41% and 30%, respectively, at the output power of 320 mW. The measured luminescence decay time at λ = 2100 nm is 3.8 ms.

The energy level diagram of Tm:Ho:YbAG is more complicated than that of the previously investigated Tm:YbAG crystal [5]. In particular, the frequency of the $^2F_{5/2} - ^2F_{7/2}$ transition of the Yb^{3+} ion is in resonance with the frequency of the $^5I_7 - ^5F_5$ transition of the Ho^{3+} ions, which can lead to an undesirable depopulation of the upper laser level. Comparison of the energy characteristics of the Tm:Ho:YbAG and Tm:YbAG laser crystals under identical experimental conditions shows almost the same efficiency, given that in the first case, lasing occurs on the transition of the Tm^{3+} ion, and in the second – on the transition of the Ho^{3+} ion. This is indicative of a high efficiency of the energy transfer from the Tm^{3+} ion to the Ho^{3+} ion and the lack of a significant influence of the $^2F_{5/2}$ level of the Yb^{3+} ion on the laser efficiency.

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