

Efficiency of population of the ${}^4I_{13/2}$ level of the Er^{3+} ion and the possibility of lasing at $1.5\ \mu\text{m}$ in Yb, Er:YAG at high temperatures

B.I. Galagan, B.I. Denker, V.V. Osiko, S.E. Sverchkov

Abstract. It is shown that, as the temperature of an Yb, Er:YAG crystal is increased from room temperature to $400\text{--}600\text{ }^\circ\text{C}$, the efficiency of energy accumulation on the ${}^4I_{13/2}$ level of the Er^{3+} ion increases by several times upon optical pumping through Yb^{3+} ions. Under these conditions, up to 60% of erbium ions can be excited, which gives promise that lasing at a wavelength of $1.54\ \mu\text{m}$ can be achieved in the Yb, Er:YAG crystal in the three-level scheme.

Keywords: yttrium–aluminium garnet, $1.5\text{-}\mu\text{m}$ ytterbium–erbium laser.

Erbium lasers emitting at the $1.54\text{-}\mu\text{m}$ ${}^4I_{13/2} - {}^4I_{15/2}$ transition of Er^{3+} ions are quite attractive for a number of applications. At present the list of materials from which the active bulk (not fibre) elements for these lasers are fabricated is virtually limited by ytterbium–erbium phosphate glasses. The Yb^{3+} ions in them play the role of sensitizers which absorb pump radiation in the region from 0.9 to $1\ \mu\text{m}$ and transfer their excitation energy to Er^{3+} ions due to nonradiative transitions (Fig. 1).

A specific feature of phosphate glasses is a combination of a high (close to 100%) quantum yield of luminescence from the upper ${}^4I_{13/2}$ laser level of the Er^{3+} ion (with the lifetime $\sim 7\ \text{ms}$) with the rather short ($1\text{--}2\ \mu\text{s}$) lifetime of the ${}^4I_{11/2}$ level of this ion. Due to rapid multiphonon relaxation from the ${}^4I_{11/2}$ level, the inverse excitation transfer from the ${}^4I_{11/2}$ level of the Er^{3+} ion to the resonance ${}^4F_{5/2}$ level of the Yb^{3+} ion and a number of parasitic up-conversion processes are drastically weakened in phosphate glasses (Fig. 1). This allows the use of ytterbium–erbium phosphate glasses as the efficient active medium for lasing in Er^{3+} ions at $1.54\ \mu\text{m}$ in the three-level scheme.

A disadvantage of glasses as a laser material is a low heat conduction (an order of magnitude worse than that for many known laser crystals). This means that glasses can be easily thermally damaged by pump radiation and therefore the average lasing power in glasses is limited.

Unfortunately, crystals with the combination of the excited-state lifetimes of erbium ions pointed out above

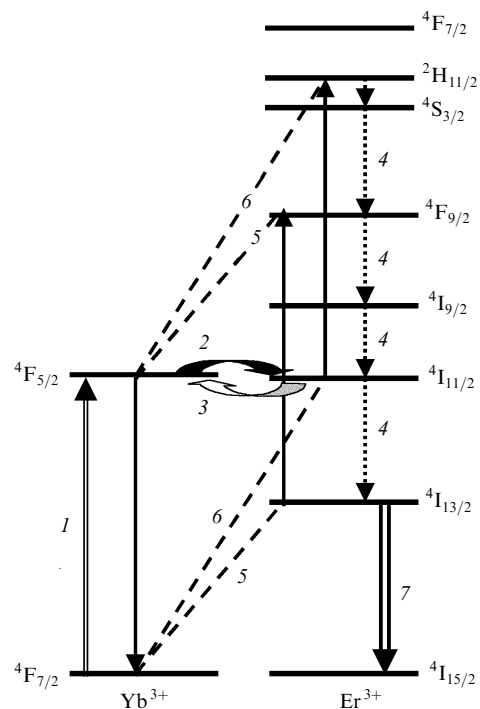


Figure 1. Energy level diagram and processes of electron energy transfer in ytterbium–erbium systems: (1) optical pumping into the absorption band of ytterbium; (2, 3) direct and reverse nonradiative transitions; (4) multiphonon relaxation; (5, 6) up-conversion processes; (7) laser transition.

are not known. Nevertheless, an yttrium–aluminium garnet crystal is an interesting matrix being studied in this direction. Thus, lasing at $\sim 1.6\ \mu\text{m}$ with the efficiency $\sim 7\%$ was observed upon pumping a Yb, Er:YAG crystal into the absorption band of ytterbium [1]. This was achieved by using the quasi-four-level scheme of the laser transition from the metastable ${}^4I_{13/2}$ level of Er^{3+} to the upper Stark sublevels of the ${}^4I_{15/2}$ ground state. We have found [2] that the spectral and kinetic properties of a Yb, Er:YAG crystal change considerably with increasing crystal temperature by a few hundreds of degrees. In particular, the lifetime of the ${}^4I_{11/2}$ level of the Er^{3+} ion decreases from 100 to $6\ \mu\text{s}$ with increasing temperature from room temperature to $800\text{ }^\circ\text{C}$ and the rate of nonradiative $Yb^{3+} \rightarrow Er^{3+}$ energy transfer increases by several times, the lifetime of the upper laser level being virtually invariable. This gives promise that heated Yb, Er:YAG crystals may be used to obtain lasing at the $1.54\text{-}\mu\text{m}$ ${}^4I_{13/2} - {}^4I_{15/2}$ transition of erbium ions in the

B.I. Galagan, B.I. Denker, V.V. Osiko, S.E. Sverchkov A.M. Prokhorov
General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38,
119991 Moscow, Russia; e-mail: galagan@ran.gpi.ru, denker@Lst.gpi.ru

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three-level scheme (as in ytterbium–erbium glasses at room temperature). We have estimated [2] the optimal concentration of activators $[(1 - 1.4) \times 10^{21} \text{ cm}^{-3}$ for Yb^{3+} and $(7 - 10) \times 10^{19} \text{ cm}^{-3}$ for $\text{Er}^{3+}]$ and temperature ($\sim 600^\circ\text{C}$) to achieve lasing. However, these studies were performed only at low excitation levels of erbium ions in the absence of nonlinear up-conversion processes. The inevitable development of these processes with increasing the pump power can considerably affect the achievement of the inverse population at the ${}^4\text{I}_{13/2} - {}^4\text{I}_{15/2}$ transition because, as is known, the three-level lasing scheme requires the excitation of no less than 50% of active particles. Thus, the question of the possibility of obtaining lasing in Yb, Er:YAG at $1.54 \mu\text{m}$ remains open.

In this paper, we studied the dependences of the population of the upper ${}^4\text{I}_{13/2}$ laser level of erbium ions on the pump energy density and temperature of a Yb, Er:YAG crystal.

Samples in the form of plane–parallel 1-mm-thick polished plates of diameter 7 mm were cut from a crystal grown by the method of horizontal crystallisation with concentrations of Er^{3+} and Yb^{3+} ions equal to $6.9 \times 10^{19} \text{ cm}^{-3}$ and $1.38 \times 10^{21} \text{ cm}^{-3}$, respectively.

The population of the upper laser level of erbium was measured by the method that we used earlier [3] to study ytterbium–erbium glasses, which were also used in this paper for comparison and additional calibration. Ytterbium–erbium samples were excited into the absorption band of ytterbium by a neodymium phosphate glass laser at $1.055 \mu\text{m}$. The laser emitted 1-ms, 10-J pulses in the free-running mode, which were focused into a spot of diameter 2 mm on samples. The dependence of the luminescence intensity at $1.5 \mu\text{m}$ on the absorbed power was studied. To provide the sufficient measurement accuracy, care was taken to ensure the spatial uniformity of excitation of samples:

- (i) Samples had a low optical density;
- (ii) pump laser radiation was highly uniform over the beam cross section due to the use of a diffusively reflecting lamp illuminator;
- (iii) incident energy was varied by using a set of calibrated neutral filters for fixed pump energy;
- (iv) luminescence emitted only from the central, most uniformly excited part of a sample was incident on a germanium photodetector.

The density of excited erbium ions was determined by the following method. Because the luminescence intensity at low excitation levels depends linearly on the pump energy, the concentration N^* of excited erbium ions can be determined from the expression

$$N^* = \frac{E_{\text{abs}}\eta}{SLhv},$$

where E_{abs} is the absorbed energy; SL is the pumped-region volume; S is the cross section of the pump laser beam; L is the sample thickness; hv is the pump photon energy; η is the quantum efficiency of electron energy transfer from ytterbium to erbium, which can be written at low temperatures as $\eta = 1 - \tau_{\text{Yb}}/\tau_{0\text{Yb}}$, where τ_{Yb} and $\tau_{0\text{Yb}}$ are the luminescence lifetimes of ytterbium in the presence and absence of erbium ions, respectively.

The energy absorbed in samples was determined from absorption coefficients k measured at $1.055 \mu\text{m}$. The absorption coefficient of a phosphate glass with ytterbium and

erbium concentrations equal to 4×10^{21} and $5 \times 10^{19} \text{ cm}^{-3}$ was 0.13 m^{-1} at room temperature. The temperature dependence of the absorption coefficient measured for a Yb, Er:YAG crystal with the ytterbium concentration of $1.38 \times 10^{21} \text{ cm}^{-3}$ is presented in Fig. 2. This dependence is caused by the broadening of the absorption spectrum of ytterbium with increasing temperature and is virtually linear. Figure 3 demonstrates the dependences of the relative population of the upper ${}^4\text{I}_{13/2}$ laser level of the Er^{3+} ion in YAG on the pump energy and temperature. Also, the corresponding dependence for a phosphate glass at 20°C is presented. On the abscissa the dimensionless pump energy $\varepsilon = E_{\text{abs}}/(SLhvN)$ (i.e. the number of absorbed pump photons per erbium ion, where N is the concentration of erbium ions in a sample) is plotted, and on the ordinate – the ratio $\beta = N^*/N$.

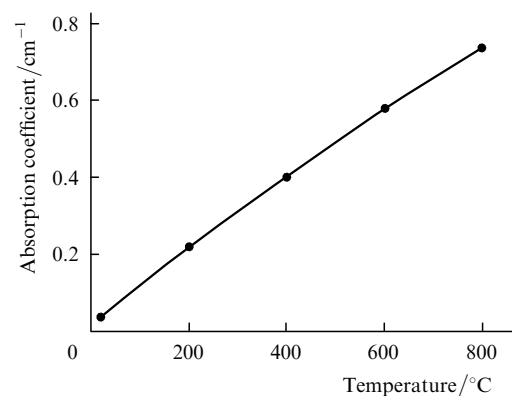


Figure 2. Temperature dependence of the absorption coefficient k at $1.055 \mu\text{m}$ for the Er, Yb:YAG sample with the ytterbium concentration $1.38 \times 10^{21} \text{ cm}^{-3}$.

One can see from Fig. 3 that, as temperature is increased from room temperature to 600°C , the sensitisation of luminescence of erbium ions upon pumping into the absorption band of ytterbium noticeably increases. This is explained, as in the case of low excitation levels [2], by the decrease in the lifetime of the ${}^4\text{I}_{11/2}$ state of erbium ions with increasing temperature. We explain the decrease in the luminescence sensitisation efficiency observed at $T > 600^\circ\text{C}$ by the thermal population of this state from the ${}^4\text{I}_{13/2}$ level. The maximum achievable value of β in the Yb, Er:YAG crystal at any temperatures proves to be considerably lower than that in the phosphate glass. It follows from Fig. 3 that the pump energy providing the achievement of the inversion threshold ($\beta \geq 0.5$) in the Yb, Er:YAG crystal is approximately five times higher than that in phosphate glass. The maximum value of β for the Yb, Er:YAG crystal obtained in the temperature range between 400 and 600°C is ~ 0.6 . We explain such a difference between the dependences $\beta(\varepsilon)$ in the phosphate glass and YAG crystal first of all by different lifetimes τ of the ${}^4\text{I}_{11/2}$ state of erbium ions in glass ($1 - 2 \mu\text{s}$) and YAG ($6 - 8 \mu\text{s}$) at $400 - 600^\circ\text{C}$. This probably leads to higher losses in the $\text{Yb}^{3+} \rightarrow \text{Er}^{3+}$ excitation efficiency in YAG compared to the phosphate glass due to the inverse $\text{Yb}^{3+} \leftarrow \text{Er}^{3+}$ energy transfer and up-conversion processes developing at high pump levels.

Note that the dependences $\beta(\varepsilon)$ for the Yb, Er:YAG crystal in the temperature range between 400 and 600°C are

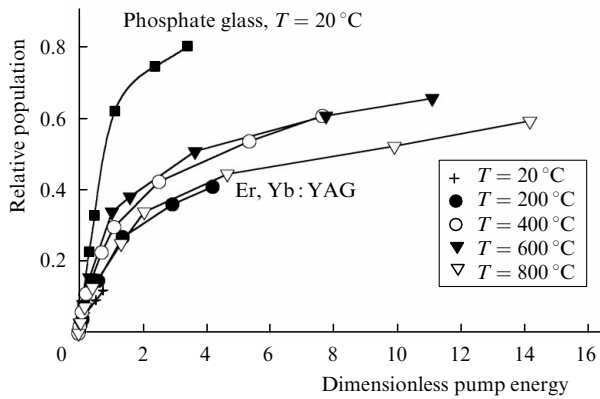


Figure 3. Dependences of the relative population ε of the $^4I_{13/2}$ level of erbium ions on the dimensionless pump energy β in the phosphate glass at temperature 20 °C and in the 0.5% Er, 10% Yb:YAG crystal at temperatures between 20 and 800 °C.

similar to the corresponding dependences obtained for the ytterbium–erbium system in a silicate glass at room temperature [4]. Taking into account that the lifetimes of the $^4I_{11/2}$ level of the Er^{3+} ion in both cases are close (8–20 μ s in silicate glasses), this result seems reasonable.

We attempted to obtain lasing in the Yb, Er:YAG crystal at 1.54 μ m upon laser pumping. The crystal described above, whose quality was not good enough unfortunately, was used to fabricate a cylindrical active element of diameter 5 mm and length 30 mm. The active element (without optical coatings on its ends) was placed into a ceramic tube heated up to 600 °C and was pumped longitudinally by the neodymium glass laser. The resonator of length \sim 150 mm was formed by concave mirrors with the radii of curvature of 100 mm with the reflectance at 1.54 μ m higher than 99.5%, which were mounted outside the heated tube. We have failed to obtain lasing under these conditions at the pump energy density \sim 250 J cm $^{-2}$ (at which the crystal was damaged by pump radiation scattered by inclusions), which corresponds according to Fig. 3, to the value $\beta \approx 0.5 - 0.55$.

Thus, we have shown that the efficiency of energy accumulation on the $^4I_{13/2}$ level of the Er^{3+} ion in the Yb, Er:YAG crystal can be considerably increased by heating the crystal by a few hundreds of degrees above room temperature. We have found the optimal temperature region (400–600 °C) in which up to 60% of erbium ions can be excited to the $^4I_{13/2}$ state upon pumping through Yb^{3+} ions, which gives promise to achieve lasing at \sim 1.54 μ m in the three-level scheme by optimising all elements of the laser.

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