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Interaction of Ho^{3+} and Tm^{3+} Ions in YSGG : Cr^{3+}, Ho^{3+}, Tm^{3+} at Strong Selective Excitation

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Abstract – The processes of the population and relaxation of excited states of the Ho^{3+} and Tm^{3+} ions in YSGG : Cr^{3+}, Tm^{3+}, Ho^{3+} laser crystals are investigated for the excitation modes which simulate real laser pumping. The conditions are stated under which the interaction of excited ions affects the ratio of the populations for the levels $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$, resulting in a considerable relative decrease of the population of the upper laser level of $^5I_7Ho^{3+}$.

The appearance of powerful diode lasers has promoted further investigations and development of new types of lasers employing diode pumping, including lasers with a two-micron spectrum band. Generation at the transition $^5I_7 \to {}^5I_8$ of the ions Ho^{3+} ($\lambda_{gen} = 2.1$ µm) for diode pumping was obtained in the crystals YAG : Cr^{3+}, Ho^{3+}, Tm^{3+} and YSGG : Cr^{3+}, Ho^{3+}, Tm^{3+} [1 - 3]. In these crystals the formation of inverse population in Ho^{3+} ions involves several different schemes of interaction of additive particles, such as energy transfer from Cr^{3+} ions to Tm^{3+} ions, cross-relaxation excitation exchange in Tm^{3+} ions, energy migration over Tm^{3+} ions, energy transfer from Tm^{3+} ions to Ho^{3+} ions and in reverse order, and finally pairwise interaction of excited additive ions. As was found in Ref. [4], at operating values of concentration of excited particles and pumping density which really occur (especially at laser excitation), the main channel of excitation losses from the upper laser level $^5I_7Ho^{3+}$ in the crystal YSGG : Cr^{3+}, Ho^{3+}, Tm^{3+} is the processes of interaction of excited ions Ho^{3+} with excited ions Tm^{3+} ($^3H_4 \to {}^3H_6Tm^{3+}$, $^5I_7 \to {}^5I_5$, $^5I_6Ho^{3+}$). Then, due to a fast process of intracentral relaxation, $^5I_5 \to {}^5I_6Ho^{3+}$, nonradiative transfer $^5I_6Ho^{3+} \to {}^3H_5Tm^{3+}$ and intracentral relaxation $^3H_5 \to {}^3H_4Tm^{3+}$, the excitation quantum returns to the level $^3H_4Tm^{3+}$.

Usually all attempts to develop a quantitative description of such a system involve the application of kinetic equations with constant probabilities for energy transfer between the subsystems of Tm^{3+} and Ho^{3+} ion. Occasionally, however, the validity of this approach is not clear (specifically, if the probability dispersion of pairwise interaction of different particles is observed). At large pumping densities and, therefore, at high probability of the processes of up-conversion the situation becomes even more complicated.

The aim of the present work is to experimentally study the dynamics of population and relaxation of the upper laser level $^5I_7Ho^{3+}$ and the level $^3H_4Tm^{3+}$ in YSGG : Cr^{3+}, Ho^{3+}, Tm^{3+} crystal, including the case of interaction of excited Ho^{3+} and Tm^{3+} ions at various durations and powers of exciting light pulses simulating both lamp and diode laser pumping.

The following samples of YSGG : Cr^{3+}, Ho^{3+}, Tm^{3+} crystals (which were polished plates about 1.5 mm thick) have been investigated: $n_{Cr} = 2.5 \times 10^{20}$ cm^{-3}, $n_{Ho} = 1 \times 10^{19}$ cm^{-3}, $n_{Tm} = 8 \times 10^{20}$ cm^{-3}. By means of a shutter, the crystals were excited by square pulses of radiation generated by a continuous wave Kr laser ($\lambda = 647.1$ nm) (its radiation was focused on a spot with a diameter of 100 µm). The duration of pumping pulses was varied in the range from one to 15 ms, and their power – from 0.15 up to 3.4 W. The excitation of crystals corresponded to the absorption band of Cr^{3+} ions at the transition $^4A_2 \to {}^4T_2$, ^2E with further fast energy transfer $^4T_2$, ^2E (Cr^{3+}) $\to {}^3F$ (Tm^{3+}) and cross-relaxation exchange $^3F_4 \to {}^3H_4$, $^3H_6 \to {}^3H_4$ [5]. In the experiments, the kinetics of luminescence of Ho^{3+} ions at the transition $^5I_7 \to {}^5I_8$ ($\lambda = 1.99$ µm) and Tm^{3+} ions at the transition $^3H_4 \to {}^3H_6$ ($\lambda = 1.7$ µm) was detected.

The results of conducted experiments argue that the dependence of the populations of the levels $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$ on the power density of exciting radiation is not nonlinear (Fig. 1) and efficient decay time of luminescence from these levels decreases with a growth in the density of excitation (this is consistent with the results of Refs. [4, 6]). Both phenomena described above are associated with the interaction of excited Tm^{3+} and Ho^{3+} ions. It was also shown that the efficiency of these processes in the crystal with larger ion concentration Ho^{3+} ($n_{Ho} = 5 \times 10^{19}$ cm^{-3}) is higher than in the crystal with a lower ion concentration ($n_{Ho} = 1 \times 10^{19}$ cm^{-3}). This fact agrees with quantitative esti-

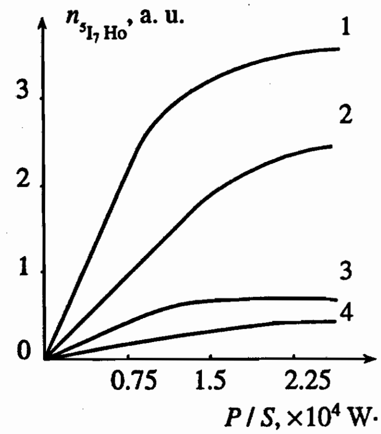

Fig. 1a. *The dependence of luminescence intensity from the level $^5I_7Ho^{3+}$ on pump power density in the crystal YSGG: Cr^{3+}, Tm^{3+}, Ho^{3+} ($n_{Cr} = 2.5 \times 10^{20}$ cm^{-3}, $n_{Tm} = 8 \times 10^{20}$ cm^{-3}, $n_{Ho} = 5 \times 10^{19}$ cm^{-3} (1), (3) and $n_{Ho} = 1 \times 10^{19}$ cm^{-3} (2), (4)). Pumping pulse durations are 1 ms (3), (4), 15 ms (1), (2).*

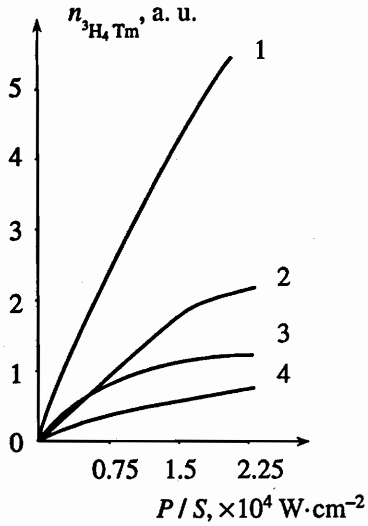

Fig. 1b. *The dependence of luminescence intensity from the level $^3H_4Tm^{3+}$ on pump power density in the crystal YSGG: Cr^{3+}, Tm^{3+}, Ho^{3+} ($n_{Cr} = 2.5 \times 10^{20}$ cm^{-3}, $n_{Tm} = 8 \times 10^{20}$ cm^{-3}, $n_{Ho} = 5 \times 10^{19}$ cm^{-3} (3), (4) and $n_{Ho} = 1 \times 10^{19}$ cm^{-3} (1), (2)). Pump pulse durations are 3 ms (2), (4), 15 ms (1), (3).*

mates made using the rate constants for mentioned "nonlinear" interactions of Tm^{3+} and Ho^{3+} ions measured in Ref. [4]. At the same time, for the values of excitation densities implemented in this experiment (which are specific for real lasers), luminescence intensity of Ho^{3+} ions is always higher for the crystals with larger ion concentrations ($n_{Ho} = 5 \times 10^{19}$ cm^{-3}) than for those with the lower ($n_{Ho} = 1 \times 10^{19}$ cm^{-3}).

An analysis of obtained results allows us to conclude that at excitation densities ($\gtrsim 10^4$ W·cm^{-3}) typical for real laser systems, interactions between excited activator ions produces an essential effect on a formation of populations of metastable laser levels.

It was experimentally established that at realized densities of exciting radiation and long pumping pulses ($\tau = 15$ ms), the ratio of maximum values of the populations at the levels $^5I_7$ (Ho^{3+}) and $^3H_4$ (Tm^{3+}) is constant in the entire range of variation of pumping power (Fig. 3a) and equal to its value for weak stationary excitation. At the same time for more short pumping pulses the ratio of maximum values of the populations for Ho^{3+} and Tm^{3+} ions declines as pumping power grows (Fig. 3b). In addition, in the case of short pulses excitation energy decreases and the total population of the levels $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$ is lower than that for long pulses.

Therefore, it is found experimentally that, at quasi-stationary pumping with realized values of power density, the processes of excitation "exchange" between the levels $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$ advance, on average, the processes of interaction of excited thulium and holmium ions ($^3H_4 \rightarrow {}^3H_6Tm^{3+}$, $^5I_7 \rightarrow {}^5I_5$, $^5I_6Ho^{3+}$; $^5I_5 \rightsquigarrow {}^5I_6Ho^{3+}$; $^5I_6 \rightarrow {}^5I_8Ho^{3+}$, $^3H_6 \rightarrow {}^3H_5Tm^{3+}$; $^3H_5 \rightsquigarrow {}^3H_4Tm^{3+}$). Finally, the ratio of excited particles corresponds to its stationary value for weak pumping. But, at the same time, at short pumping pulse the interactions of excited ions dominate; in this case the ratio of excited particles (Ho^{3+} and Tm^{3+}) does not achieve its stationary value and depends on power density of exciting radiation.

Deviation of the ratio of maximum populations for excited Ho^{3+} and Tm^{3+} ions from its stationary value results in abnormal dependence of maximum values of luminescence intensity both for Ho^{3+} and Tm^{3+} ions on duration of exciting pulse, if its energy (3 mJ) is fixed (for specified energy and weak pumping the efficiency of nonlinear interactions was sufficiently high and comparable with the relaxation rate for metastable levels $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$). As one can see from Fig. 3, if the duration of the exciting pulse is decreased less than 6 ms the maximum value of luminescence intensity of Ho^{3+} ions in the crystal with ion concentration equal to 5×10^{19} cm^{-3} does not increase, as one may expect, but remains constant (taking into account that the time $\tau \approx 6$ ms is commensurate to the effective lifetime for the levels $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$).

The circumstance established here that maximum values of the populations of Ho^{3+} and Tm^{3+} ions are not equal to their stationary values leads to the fact that the time needed for the population of Ho^{3+} ions to reach its maximum value depends not only on pumping pulse duration, but also on its power. For example, if YSGG crystal ($n_{Cr} = 2 \times 10^{20}$ cm^{-3}, $n_{Tm} = 8 \times 10^{20}$ cm^{-3}, $n_{Ho} = 5 \times 10^{19}$ cm^{-3}) is excited by light pulse of 1 ms duration and 1 W power, luminescence of Ho^{3+} ions adopts its approximate maximum value in $\Delta t \sim 1$ ms after switching off the pumping pulse. But, if the same crystal is excited by small power pulses and the duration is 1 ms, the magnitude Δt is 0.5 ms [4]. Apparently, this phenomenon (i.e., a shift of luminescence maximum of

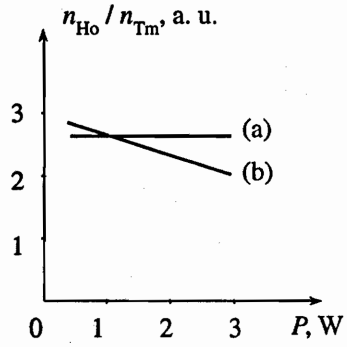

Fig. 2. *The dependence of the ratio for maximum values of the populations at the levels* $^5I_7Ho^{3+}$ *and* $^3H_4Tm^{3+}$ *on the power of exciting light in the crystal YSGG :* Cr^{3+}, Tm^{3+}, Ho^{3+} ($n_{Cr} = 2.5 \times 10^{20}$ cm^{-3}, $n_{Tm} = 8 \times 10^{20}$ cm^{-3}, $n_{Ho} = 5 \times 10^{19}$ cm^{-3})*: exciting pulse duration is* $\tau = 15$ *ms (a); exciting pulse duration is* $\tau = 3$ *ms (b).*

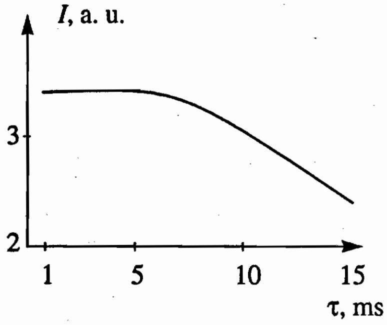

Fig. 3. *The dependence of maximum value of luminescence from the level* $^5I_7Ho^{3+}$ *on pump pulse duration at* $E_p = 3$ *mJ in the crystal YSGG :* Cr^{3+}, Tm^{3+}, Ho^{3+} ($n_{Cr} = 2.5 \times 10^{20}$ cm^{-3}, $n_{Tm} = 8 \times 10^{20}$ cm^{-3}, $n_{Ho} = 5 \times 10^{19}$ cm^{-3}).

Ho^{3+} ions with a growth of pumping power density) should be taken into account at optimization of operating modes of Ho lasers, including the case when optimum shutter exposure time at *Q*-modulation mode is to be obtained.

Distinctive behavior of the ratio of the populations for excited Ho^{3+} and Tm^{3+} ions at long and short pumping pulses (in particular, the equality of the ratio of mentioned populations to its stationary value at long pumping pulse and relative decrease of the fraction of excited Ho^{3+} ions at short pulses which depends on pumping power density) can be interpreted in the following manner. In accordance with Ref. [4], a strong dispersion of probability for energy transfer to both non-excited and excited ions takes place in studied crystals. It should be pointed out that at different pumping durations and intensities these or other Tm^{3+} and Ho^{3+} ion subsystems which contribute to either "initial" fast or "far" slow stages of energy transfer can be populated in different time moments relative to each other. Accordingly, at various pumping modes these or another subsystems of interacting ions can deliver a dominating contribution to the processes of population and depopulation of the levels $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$ (additionally, in principle, the cases of energy transfer to non-excited and excited ions imply different subsystems of interacting particles). Thus, in general, depending on particular conditions of excitation, either the processes of energy transfer from excited ions to non-excited ones ($^3H_4 \rightarrow {}^3H_6Tm^{3+}$, $^5I_8 \rightarrow {}^5I_7Ho^{3+}$ and $^5I_7 \rightarrow {}^5I_8Ho^{3+}$, $^3H_6 \rightarrow {}^3H_4Tm^{3+}$) or the processes of pairwise interaction of excited ions ($^3H_4 \rightarrow {}^3H_6Tm^{3+}$, $^5I_7 \rightarrow {}^5I_5$, $^5I_6$ Ho^{3+}) can be more efficient.

Another possible reason which ensures a non-proportionately slow increase of the population of Ho^{3+} ions with a growth of the power of short ($\lesssim 1$ ms) pumping pulses is that the populations of the levels $^3H_4Tm^{3+}$ and $^5I_7Ho^{3+}$ are approximately equal during this time interval. It appears evident that, at the same total population of levels of $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$, the interaction rate of excited ions is higher in the case where the populations of excited ions Tm^{3+} and Ho^{3+} are equal than in the case where these populations are significantly different (according to Ref. [4] in YSGG : Cr^{3+}, Tm^{3+}, Ho^{3+} crystal $n_{Tm} = 8 \times 10^{20}$ cm^{-3}, $n_{Ho} = 5 \times 10^{19}$ cm^{-3}, the ratio of the stationary populations of Ho^{3+} and Tm^{3+} is about 2.5).

Therefore, the processes of populating and relaxation of excited states of the ions Ho^{3+} and Tm^{3+} in laser crystals YSGG : Cr^{3+}, Tm^{3+}, Ho^{3+} are investigated in this work for the excitation modes, which simulate real laser pumping.

It is shown that in crystals YSGG : Cr^{3+}, Tm^{3+}, Ho^{3+} ($n_{Tm} = 8 \times 10^{20}$ cm^{-3}) interaction of excited ions Tm^{3+} and Ho^{3+}, which decreases the population of the upper laser level $^5I_7Ho^{3+}$, becomes more intensive at larger concentrations of the ions Ho^{3+} ($n_{Ho} = 5 \times 10^{19}$ cm^{-3}) rather than at lower values of this concentration ($n_{Ho} = 1 \times 10^{19}$ cm^{-3}). Nevertheless, for all pumping durations and intensities experimentally studied, an application of the crystals with higher concentrations of the ions Ho^{3+} in the lasers is more efficient with respect to the formation of inverse populations at the transition $^5I_7 \rightarrow {}^5I_8Ho^{3+}$.

The conditions are stated at which interaction of excited ions affect the ratio of the populations for the levels $^5I_7Ho^{3+}$ and $^3H_4Tm^{3+}$ resulting in considerable relative decrease of the population of the upper laser level $^5I_7Ho^{3+}$. In particular, a non-proportionately strong influence of non-linear processes on the maximum population of the level $^5I_7Ho^{3+}$ at short pumping pulses (about 1 ms), rather than at long ones (about 15 ms), is established.

Simple kinetic equations usually applied cannot provide an adequate description of the host of experimental results obtained. This is associated with the fact that, in the system we studied, a probability dispersion for energy transfer to both non-excited and excited ions takes place.

The results yielded can be relevant in optimizing operating modes for particular Ho lasers, using both lamp and diode pumping.

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