Cross-relaxation emptying of the ground state of rare-earth ions in crystals

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The ground state of the ions of rare-earth elements can be effectively emptied during pumping from an excited state of these ions. A necessary condition for the realization of this method is a cross-relaxation "breeding" of excitations in a system of interacting particles.

The branching energy-level diagram of trivalent rare-earth ions (TR^{3+}) makes it possible in several cases to effectively empty their ground state without directly pumping an external agent to it. The deactivation is instead achieved through an optical pumping from excited states of the TR^{3+} ion. A necessary condition for the realization of this method for exciting TR^{3+} ions is a cross-relaxation "breeding" of excitations in a system of interacting particles. The unconventional new pumping schemes which are being proposed could in principle be organized around a number of TR^{3+} ions, including Tm^{3+} and Er^{3+}.

As an example we consider one possible realization of this new method. The filling and relaxation of excited states of Tm^{3+} ions in an yttrium–scandium–gallium garnet (YSGG) crystal are described by the system of equations

$$\begin{cases} dn_3/dt = k_2 n_2 - \beta n_1 n_3 - (w_{31} + w_{32}) n_3, \\ dn_2/dt = -k_2 n_2 + 2\beta n_1 n_3 + w_{32} n_3 - w_{21} n_2 + k_1 n_1, \\ n_1 + n_2 + n_3 = 1, \end{cases} \tag{1}$$

where subscripts 1, 2, 3 refer to the $^3H_6$, $^3H_4$, and $^3F_4$ states, respectively, of the Tm^{3+} ion (Fig. 1); w_{ij} is the probability for an intracenter $i \rightarrow j$ relaxation; and $\beta n_1 n_3$ is the rate of cross-relaxation emptying of the $^3F_4$ state of the Tm^{3+} ion. Equations (1) reflect the excitation of Tm^{3+} ions on transitions from both the ground state ($^3H_6$) and an excited state ($^3H_4$). Here $k_1 n_1$ and $k_2 n_2$ are the excitation rates on the transitions $^3H_6 \rightarrow {}^3H_4$ and $^3H_4 \rightarrow {}^3F$, respectively.

At a Tm^{3+} ion density of 8×10^{20} cm^{-3}, during the absorption of light on the $^3H_4 \rightarrow {}^3F$ transition (we assume $n_2, n_3 \ll n_1$), the excitation of the $^3F_4$ level is expended, in a time ~ 30 μs and with a quantum yield approaching 2, on two $^3H_4$ excitations[1] ($^3F_4 \rightarrow {}^3H_4$, $^3H_6 \rightarrow {}^3H_4$). This process makes possible an effective optical pumping of this crystal on a transition from the $^3H_4$ metastable state of Tm^{3+}.

In an experiment, an yttrium–scandium–gallium garnet crystal doped with Cr

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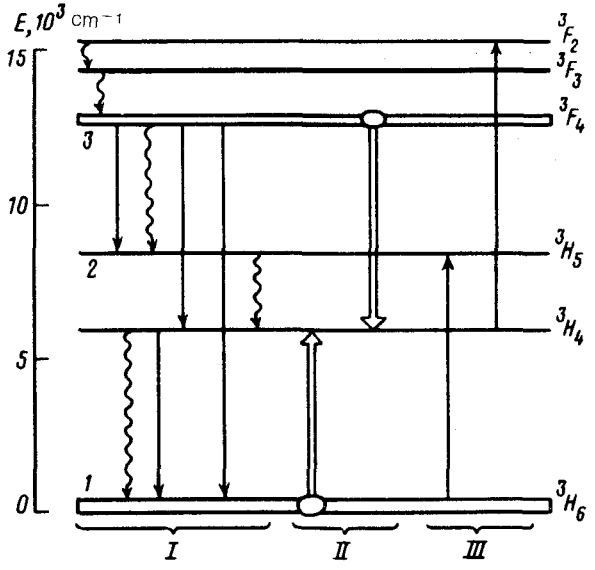

FIG. 1. Energy-level diagram of the Tm^{3+} ions. I—Intracenter decay; II—cross relaxation; III—pumping.

(2.5×10^{20} cm^{-3}) and Tm (8×10^{20} cm^{-3}) was excited by the beams from pulsed neodymium lasers, with excitation wavelengths (λ_{exc}) of 1.08 and 1.064 μm. The length of the exciting pulses was 400 μs, and the energy in a pulse ranged up to 4 J.

Figure 2 shows the experimental results on the population of the $^3H_4$ level of Tm^{3+} as a function of the pump energy at $\lambda_{exc} = 1.064$ μm (curve 2). This curve runs well above the theoretical curve (curve 3) calculated under the assumption that there is no absorption from the $^3H_4$ excited state of Tm^{3+} (with the values $w_{21} = 100$ s^{-1}, $w_{31} = 50$ s^{-1}, and $w_{32} = 1000$ s^{-1}). Also shown in Fig. 2 is the theoretical behavior in the absence of a cross-relaxation breeding of excitations ($\beta = 0$; curve 4). The population of the $^3H_4$ level reaches a maximum of 10% of the total concentration of Tm^{3+} ions; as a result, according to estimates, lasing on the $^3H_4 \rightarrow {}^3H_6$ transition of Tm^{3+} becomes possible. Note also that the filling of the $^3H_4$ in this experiment was due primarily to absorption of light from specifically the $^3H_4$ excited state of Tm^{3+}. The cross section for absorption at the wavelength 1.064 μm, on the transition $^3H_4 \rightarrow {}^3F$, was 6.6×10^{-22} cm^2, or ~ 60 times the cross section for absorption on the transition from the $^4H_6$ ground state of Tm^{3+} at this wavelength.

During excitation of the YSGG:(Cr^{3+},Tm^{3+}) crystal by light with $\lambda = 1.08$ μm, the excited states of Tm^{3+} were filled much less efficiently (despite the fact that the cross section for absorption from the ground state is larger than at $\lambda_{exc} = 1.064$ μm). This result is attributed to the small cross section ($\sim 5\times10^{-23}$ cm^2) for absorption from the $^3H_4$ metastable state at this wavelength.

This system has an interesting feature. It follows from (1) that in the quasisteady pproximation ($dn_3/dt = 0$), which is valid at high Tm^{3+} densities ($\geqslant 8\times10^{20}$

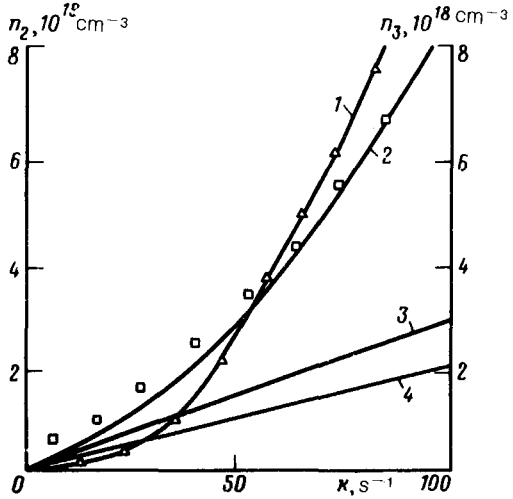

FIG. 2. Populations of levels of the Tm^{3+} ion versus the pump energy. △, 1: The $^3F_4$ level. △—Experimental; 1—theoretical, calculated with $\sigma_{^3H_4 \to {}^3F_4} = 6.6 \times 10^{-22}$ cm^2 and $\beta = 3 \times 10^4$ s^{-1}. □, 2, 3, 4: The $^3H_4$ level. □—Experimental; 2—calculated with $\sigma_{^3H_4 \to {}^3F_4} = 6.6 \times 10^{-22}$ cm^2 and $\beta = 3 \times 10^4$ s^{-1}; 3—calculated with $\sigma_{^3H_4 \to {}^3F_4} = 0$ cm^2; 4—calculated with $\sigma_{^3H_4 \to {}^3F_4} = 6.6 \times 10^{-22}$ cm^2 and $\beta = 0$.

cm^{-3}), there is a certain critical intensity of steady-state pumping, k_2^{crit} (with $k_1 = 0$),

$$k_2^{\text{crit}} = w_{21}[(1 + (w_{32} + 2w_{31}) / (\beta n_1 - w_{31})], \tag{2}$$

at which the entire picture of the filling of the excited states of the Tm^{3+} ion changes radically. Under the conditions $\beta n_1 \gg w_{31}, w_{32}$ we find $k_2^{\text{crit}} \approx w_{21}$ from (2). For the population of level 2 we have

$$dn_2/dt \approx k_2 n_2 - w_{21} n_2, \tag{3}$$

With a constant pump k_2, and under the condition $w_{21} = k_2$, the excitations which reach level 2 remain there for an unlimited time. At $k_2 > k_2^{\text{crit}}$, the dumping of excitations to the second level leads to a further increase in the population there, while at $k_2 < k_2^{\text{crit}}$ the population of this level decreases with a time constant which may be much longer than the intrinsic lifetime of the $^3H_4$ level of Tm^{3+}.

Analysis shows that situations similar to those described above can be realized in YSGG:Er^{3+} crystals. In this case the role of the second level would be played by the $^4I_{13/2}$ state, which is filled as a result of cross-relaxation processes involving the level $^4S_{3/2}$ ($^4S_{3/2} \to {}^4I_{13/2}$; $^4I_{15/2} \to {}^4I_{9/2} \gg {}^4I_{11/2} \gg {}^4I_{13/2}$, etc.).

In summary, a new method has been proposed for exciting TR^{3+} ion on transitions from excited states. The results found here open up a new approach for searching for, and evaluating the efficiency of, active media for solid state lasers, especially for lasers which operate with selective pumping and diode pumping. In the analysis of

absorption spectra of activator ions, one should abandon the conventional wisdom that absorption of pump light on a transition from an excited state is obviously a harmful factor. On the contrary, one should seek situations in which an excited absorption will provide an efficient energy storage in the active medium and thus increase the working efficiency of the laser.

[1]E. V. Zharikov, S. P. Kalitin, V. V. Laptev *et al.*, Kvant. Elektron (Moscow) **13**, 216 (1986) [Sov. J. Quantum Electron. **16**, 145 (1986)].

Translated by D. Parsons