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Cr-Tm and Cr-Er energy transfer in GSAG and YSGG crystals

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Sensitization of rare-earth doped solid-state lasers by chromium ions can greatly improve their flashlamp-pumped performance. Along with the widely used Cr,Er:YAG and Cr,Tm:YAG crystals, the similar systems, based on GSAG (gadolinium scandium aluminum garnet) and YSGG (yttrium scandium gallium garnet) crystals [1,2], are of intense interest currently.

In this paper the results of the systematic study of series of Cr,Tm:GSAG, Cr,Tm:YSGG and Cr,Er:YSGG crystals with Tm and Er concentrations of $2 \times 10^{20} \text{ cm}^{-3}$ to $40 \times 10^{20} \text{ cm}^{-3}$ are presented. The dynamics of Cr-Tm and Cr-Er energy transfer were studied by analysis of Cr decay kinetics and Er fluorescence build-up at the selective excitation of Cr ions.

The analysis of Cr decay curves with the help of computer simulation allowed to identify the mechanisms and the character of energy transfer as well as to evaluate the corresponding microparameters of the transfer in the studied crystals.

The example of Cr^{3+} fluorescence decay curve in the presence of Tm^{3+} ions in GSAG crystal is depicted in Fig. 1a. At low concentrations of Tm^{3+} ($\sim 2 \times 10^{20} \text{ cm}^{-3}$) the energy transfer is dipole-dipole, has a static disordered character and is well described by the following relation:

$$I(t) = I_0 \exp\left(-\frac{t}{\tau_{\text{Cr}}}\right) - \gamma\sqrt{t}, \quad (1)$$

where I_0 is the normalized initial fluorescence intensity, $\gamma = -4/3\pi^{3/2}n_{\text{A}}\sqrt{C_{\text{DA}}}$ is the parameter of the direct static interaction $\text{Cr}^{3+}\text{-Tm}^{3+}$, n_{Tm} is the concentration of Tm^{3+} , and C_{DA} is the microparameter equal to the probability of Cr^{3+} and Tm^{3+} ions interaction at the distance of 1 cm. The quantum efficiency of the energy transfer in this crystal even at low $n_{\text{Tm}} = 2 \times 10^{20} \text{ cm}^{-3}$ turned out to exceed 80%.

The same mechanisms of interaction between Cr^{3+} and Er^{3+} ions in YSGG crystal have been observed at the n_{Er} up to $2 \times 10^{20} \text{ cm}^{-3}$.

Thus, at low concentrations of Tm and Er ($2 \times 10^{20} \text{ cm}^{-3}$) the transfer is dipole-dipole, which demonstrates Fig. 1b (slope is equal to $1/2$), and has a static disordered character. In this Fig. $\Pi(t)$ is the "energy transfer function" in the following formula, describing the fluorescence decay of donor ions (D) at the relative concentration of acceptor ions (x_{A}) and donor-acceptor interaction probability $w_{\text{DA}} = C_{\text{DA}}/R^6$ (for dipole-dipole case):

$$N_{\text{D}}(t) = N_{\text{D}}(0) \exp\left(-\frac{t}{\tau_{\text{D}}} - \Pi(t)\right), \quad (2)$$

where

$$\Pi(t) = \sum_i \ln(1 - x_{\text{A}} + x_{\text{A}}e^{-(w_{\text{DA}})_i t}) \quad (3)$$

At higher concentrations the fast ($\tau < 100$ ns) energy transfer has been observed.

In Fig. 2 b the dependence of the part of Cr ions, rapidly transferring their energy (P), upon the relative concentration of acceptors (Tm and Er ions), is presented (circles). The calculated probabilities of the presence of at least one acceptor in the first (solid line) and the first two (dashed line) nearest coordination shells are also plotted. This probability is equal to $P = 1 - (1 - n_A)^p$, where n_A is the relative concentration of acceptors, p is the number of acceptors in the first ($p=8$) and in the first two ($p=12$) coordination shells. It becomes evident, that for both Tm and Er ions the good agreement with the experimental data is achieved under the first coordination shell approximation. Thus, the radius of this strong interaction was found to be ~ 3.5 Å.

The Cr decay kinetics at the Er concentration of $40 \times 10^{20} \text{ cm}^{-3}$, shown in Fig. 2 a is of special interest. It consists of the two exponential parts. The initial (rapid) part corresponds to the fast energy transfer. About 75% of all Cr^{3+} excitations transfer their energy rapidly to Er^{3+} ions within the first 100ns. The second (slow with the lifetime $\tau \sim 1.1 \mu\text{s}$) part is not described within the dipole-dipole energy transfer model, but it can be satisfactorily described within the "excitation trapping" model at the strong interaction of excited Cr ions with the nearest Er ions at the $^4I_{9/2}$ level (with a lifetime of less, than $1 \mu\text{s}$). The observed Cr lifetime turned out to be equal to the calculated one within the mentioned above model of "excitation trapping" [3].

Thus, the energy transfer mechanisms and microparameters of interaction between Cr^{3+} and Tm^{3+} ions in GSAG were established. In YSGG:Cr, Er and YSGG:Cr, Tm crystals the "rapid energy transfer" to the Er^{3+} and Tm^{3+} ions was investigated. The radius of this strong interaction was found to be ~ 3.5 Å. It was also shown, that in both cases of the energy transfer to Tm^{3+} and Er^{3+} ions, the strong interaction occurs within the first coordination shell of Cr^{3+} ions. The model of the "excitation trapping" between Cr^{3+} and Er^{3+} ions at their strong interaction was suggested to describe the Cr^{3+} fluorescence decay at large concentrations of Er^{3+} ions ($40 \times 10^{20} \text{ cm}^{-3}$).

References

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Figure 1: *a*: The Cr^{3+} fluorescence decay curve in the presence of Tm^{3+} ions ($n_{\text{Tm}} = 2 \times 10^{20} \text{cm}^{-1}$) in GSAG; *b*: Logarithm of the “energy transfer function” vs. logarithm of time in the same crystal.

Figure 2: Circles: dependence of the part of Cr^{3+} ions involved in the "fast energy transfer" to Er^{3+} and Tm^{3+} ions (P) on the relative concentration of the latter (n_A). Solid line: results of the calculations, taking into account the energy transfer only to the first coordination shell. Dashed line: same taking into account both the first and the second coordination shells.

Figure 3: The Cr^{3+} fluorescence decay in the presence of Er^{3+} ions ($\lambda = 740 \text{ nm}$, $N_{\text{Er}} = 40 \times 10^{20} \text{ cm}^{-3}$).