Experimental observation of "excitation trapping" in a system of strongly interacting particles

T. T. Vasiev, Yu. K. Voron'ko, V. V. Osiko, A. M. Prokhorov, and I. A. Shcherbakov

P. N. Lebedev Physics Institute, USSR Academy of Sciences
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An experimental study is made of processes of electronic excitation exchange between Er^{3+} and Yb^{3+} ions in fluorite crystals. From observation of the kinetics of decay of $^2F_{5/2}$ metastable states of the Yb^{3+} ion and $^4I_{11/2}$ states of the Er^{3+} ion on selective excitation of Yb^{3+} ions ($\lambda_{exc} = 0.91\ \mu$) and Er^{3+} ions ($\lambda_{exc} = 0.532\ \mu$) it is concluded that there is a strong interaction between these ions. We have observed experimentally a lengthening of the decay time of the $^2F_{5/2}$ excited state by a factor of 2.5 above the spontaneous decay time (excitation trapping effect) and also a saturation of the process of excitation transfer from Er^{3+} to Yb^{3+}. Such effects had previously been predicted theoretically. It is proposed to use the observed excitation trapping effect to increase the effective lifetime of the upper laser level $^4I_{11/2}$ of Er^{3+} ions in an erbium garnet laser.

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The lifetime τ of an excited level of an ensemble of particles is one of the most important parameters which determine the behavior of this ensemble in an excited state. In a number of physical problems the need arises to change the effective lifetimes of multilevel systems. Thus, in quantum electronics to achieve the maximum efficiency of lasers definite relations must be satisfied between the lifetimes of the excited levels, between the lifetime of the metastable level and the duration of the pumping pulse, and so forth. [1]

Effects of a decrease in τ under the influence of various types of excitation are well known (stimulation by a radiation field, quenching). The question of increasing the lifetime above the spontaneous lifetime in such systems has received practically no consideration. An analysis of theoretical studies on the transfer of electronic excitation shows that an increase in τ can be expected if there is a strong interaction between the particles. For example, Agabekyan[2] predicts the possibility of increased excitation lifetime with a fast reversible transfer of energy between particles with different τ. This effect was named by him "excitation trapping" by analogy with the radiation trapping occurring in optically dense media radiating in resonance.

In the present work we have studied experimentally processes leading to an increase in the lifetime of one of the ions in doubly activated crystals with a strong interaction between the impurity particles. Weak and strong interactions have been studied most completely theoretically for the simplest double system—a donor D and acceptor A. A number of studies[3-17] devoted to transfer of excitation in a donor-acceptor pair have shown that the kinetics of decay should carry a wealth of information on the elementary interaction processes in such systems.

In the strong-coupling case in which we are interested,[1] when the rate of reversible transport $(D \rightleftarrows A)P_{DA}$ is much greater than the reciprocal lifetime of the acceptor $1/\tau_A^0$ $(P_{DA} \gg 1/\tau_A^0)$, the decay of excited levels of the donor and acceptor in times $t > P_{DA}^{-1}$ should occur

with a single rate $1/2\tau_A^0$ if $\tau_D^0 \gg \tau_A^0$. On excitation of the donor D this situation is often called "outflow saturation in the acceptor" since here it is just the relaxation in the acceptor, as the slowest process, which limits the rate of disappearance of the donor excitation: $\tau_D = 2\tau_A^0$. By exciting the acceptor ion in the very simple two-particle system under consideration, we reveal the effect of excitation trapping, since the lifetime of the acceptor excited state (τ_A) is drawn out by a factor of two in comparison with its spontaneous lifetime as the result of capture of radiation by the weakly relaxing donor: $\tau_A = 2\tau_A^0$. In spite of the fact that the theoretical analysis of the interactions in the elementary D–A system leads to very interesting conclusions, the search for such systems and their experimental investigation has turned out to be extremely difficult.[2] Usually in an experiment it is necessary to deal with large groups of particles. Here, in addition to the processes indicated above of donor-acceptor transfer and relaxation, it is necessary to take into account donor-donor and acceptor-acceptor interactions. A crystal activated with two different types of ions is just such a system.

In the impurity system let the concentration of particles of type I be n_1 and that of type II be n_2. If the lifetime of the excited level of the ions of type I (τ_1) is greater than the lifetime of the excited level of the ions of type II (τ_2), then in the resonance interaction between these levels the ions of type I are often called donors, and the ions of type II—acceptors. In the case of a strong interaction these terms lose their direct meaning. For a donor concentration less than the acceptor concentration $(n_1 \ll n_2)$ the strong coupling case has been studied theoretically by a number of workers[3,9,12,14,16] and most completely by Burshteĭn and Pusep.[19] This is a limiting case of the well known static quenching of donor excitations by acceptors. If the reverse inequality is satisfied, $n_1 \gg n_2$, quenching of donor excitations occurs most frequently by means of migration of donors over the system of donors (ions of type I) to the acceptors (ions of type II).[20] Here the limiting case (strong coupling) is extremely rapid migration (supermigration) of excitation to the ac-

ceptor. [2,21-23] As a result of the fact that the exact solution of the many-particle problem for arbitrary n_1 and n_2 and with account being taken of donor-donor (P_{11}), acceptor-acceptor (P_{22}), and donor-acceptor (P_{12}) interactions is extremely complicated, Heber[24] and Kingsley[24] analyzed the conditions of applicability of the collective kinetic equations for description of the interactions between two impurity subsystems. On the basis of solution of such equations these authors[24] determined parameters which characterize the decay of the excited levels (of ensembles) of the donors and acceptors in the presence of an interaction between them. A similar approach was applied previously to electron paramagnetic resonance in study of the harmonic cross relaxation between spin sublevels. [25] There the equalization of the populations between different levels as the result of excitation exchange was investigated theoretically and experimentally. However, the question of the rate of removal and the lifetime of spin excitation in such a system was not discussed; in addition, the assumption was made of equality of the spin-lattice relaxation times of different levels (i.e., in our case $\tau_1 = \tau_2$).

The most important theoretical results of the studies cited above, which refer to the case of strong interaction, can be summarized as follows. On rapid selective excitation of one of the two ensembles (for example, I) of ions interacting in resonance there occurs in the first stage a very rapid equalization (in a time $t \sim P_{12}^{-1}$) of the populations of the excited levels of the two subsystems. Here after a time $t > P_{12}^{-1}$, regardless of the initial conditions, the relative populations of the excited levels of the two subsystems will be equal independently of the probabilities of transitions to them, i.e., $n_1^*/n_1 = n_2^*/n_2$. The further decay of the excited levels of the two systems occurs with an identical rate equal to

$$A_{av} = \frac{n_1 \tau_1^{-1} + n_2 \tau_2^{-1}}{n_1 + n_2}. \tag{1}$$

It is evident from this that the limiting rate of disappearance of donor excitations does not depend on the probabilities P_{11}, P_{22}, and P_{12} as occurs for a weak interaction, but is determined, as in the case discussed above of a pair of ions D-A, only by the rate of relaxation in the acceptor τ_2^{-1} (for $\tau_1 \gg \tau_2$) and by the ratio n_1/n_2 (the case of discharge saturation in the acceptor).

On excitation of subsystem II, which has a shorter lifetime $\tau_2 < \tau_1$, depending on the relation between τ_1 and τ_2 and n_1 and n_2 an extension of the decay time of the excited state of this subsystem should be observed (excitation trapping):

$$\tau_{av} = A_{av}^{-1} = \frac{n_1 + n_2}{n_1 \tau_1^{-1} + n_2 \tau_2^{-1}}. \tag{2}$$

In spite of the fact that at the present time the theoretically limiting situations have been discussed rather completely, there is practically no experimental information on them.[3)]

We have synthesized and studied single crystals of $CaF_2 + ErF_3$ (1-10 weight%), $CaF_2 + YbF_3$ (1-10 weight%), $CaF_2 + Er_{0.5}Yb_{0.5}F_3$ (0.1, 0.3, 1.0, 3.0, 5.0, 7.0, and

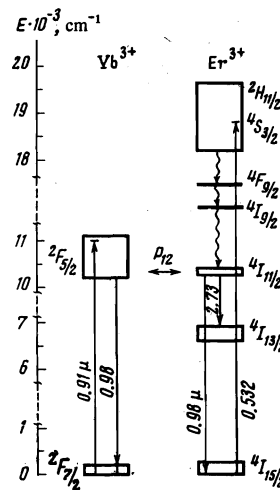FIG. 1. Diagram of interaction of Yb^{3+} and Er^{3+} ions in fluorite.

10.0 weight%), and $CaF_2 + Er_x Yb_{1-x} F_3$ 10 weight % (x = 0.1, 0.25, 0.38, 0.41, 0.43, 0.5, 0.54, 0.74, 0.8, 0.85, and 0.87). Crystals of fluorite activated with Er^{3+} and Yb^{3+} were grown by the method of directed crystallization in a fluorinating atmosphere. Monitoring of the value of x and the ratio of concentrations of Er and Yb ions was carried out on the basis of the absorption spectra of the crystals, taken in an SF-8 spectrophotometer.

Study of the absorption and luminescence spectra of the transitions $^2F_{5/2} - ^2F_{7/2}$ (the Yb^{3+} ion) and $^4I_{11/2} - ^4I_{15/2}$ (the Er^{3+} ion) has shown that they are in good agreement and we can expect a rather effective interaction between the metastable levels of Yb^{3+} and Er^{3+} ions in fluorite crystals. A diagram of the interaction $Er^{3+} \rightarrow Yb^{3+}$ in fluorite is shown in Fig. 1. We have measured the damping time of Er^{3+} and Yb^{3+} ions in the crystals studied with use of selective excitation of these ions. Excitation of Yb^{3+} ions was accomplished in the upper Stark sublevel of the excited state $^2F_{5/2}$ by a semiconductor injection laser. Here no excitation of Er^{3+} ions occurred, as a result of the absence of its absorption bands at the frequency of the semiconductor laser. The principal parameters of the semiconductor laser were $\lambda_{gen} = 9100$ Å, $P_{peak} = 40$ W, $t_{pulse} = 300$ nsec, $f \leq 100$ Hz. For selective excitation of Er^{3+} ions we used a LTIPCh-7 laser employing neodymium-activated garnet, with doubling of the radiated frequency by means of a lithium niobate crystal. The principal parameters of this laser were $\lambda = 5320$ Å, $P_{peak} = 1.5 \times 10^5$ W, $t_{pulse} = 15$ nsec, $f \leq 100$ Hz. To separate the spectral lines in the luminescence of the crystals we used an MDR-2 monochromator with a diffraction grating.

Determination of the lifetime was carried out by analysis of the kinetics of damping, obtained on the screen of an EMG-1555 oscilloscope. The photodetectors used were an FÉU-62 photomultiplier and a PbS photosensitive resistor cooled with liquid nitrogen. The range of variation of the intensities for the kinetic measurements was $\sim 10^2$. The accuracy in determination of the lifetime was 10%. Analysis of the kinetics of damping of the luminescence of the $^4I_{11/2}$ level of Er^{3+} ions in $CaF_2 + ErF_3$

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crystals (1 to 10 weight%) showed that it is strictly exponential with a constant $\tau_{Er}^0 = 8.8$ msec and does not change with increasing ErF_3 concentration to 10 weight %. Studies of the kinetics of luminescence of various optical centers in fluorite crystals activated with Yb^{3+} ions were carried out in detail by Voron'ko et al.[26] It was shown that for a YbF_3 concentration from 1 to 10 weight% the Yb^{3+} ions can be considered as forming one complex low-symmetry center, and the probability of interaction between different Yb^{3+} ions is $P \gg 10^3$ sec^{-1}. The damping kinetics of the luminescence of the $^2F_{5/2}$ level of Yb^{3+} ions also has an exponential form with a constant $\tau_{Yb}^0 = 3$ msec which does not depend on the Yb content in the indicated region of concentrations. Measurements made by us of the damping times of Yb^{3+} ions in fluorite have confirmed these results.

Investigations of the interaction processes were carried out in doubly activated fluorite crystals with equal concentrations of Er^{3+} and Yb^{3+} ions, $CaF_2 + Er_{0.5}Yb_{0.5}F_3$ (0.1, 0.3, 1.0, 3.0, 5.0, 7.0, and 10.0 weight%). Analysis of the kinetics of these crystals on selective excitation of various ions showed that for concentrations less than 1 weight% a complex nonexponential damping kinetics is observed, which indicates a weak interaction between different optical centers of ions of the same and different types. With a concentration greater than 3 weight%, exponential damping kinetics is observed for luminescence in the transitions $^2F_{5/2}-^2F_{7/2}$ of the Yb^{3+} ion, $^4I_{11/2}-^4I_{15/2}$ of the Er^{3+} ion ($\lambda = 0.98 \mu$), and $^4I_{11/2}-^4I_{13/2}$ of the Er^{3+} ion ($\lambda = 2.73 \mu$), i. e., strong coupling exists: $P_{12} \gg 1/\tau_{Er}^0$, $1/\tau_{Yb}^0$. Here the measured luminescence damping times at 2.73 and 0.98 μ were in good agreement and did not depend on the means of excitation ($\tau = 4.4$ msec).

The equality of the damping times at different wavelengths permits us to conclude that the lifetime of the $^4I_{11/2}$ level (τ_{Er}) is equal to the lifetime of the $^2F_{5/2}$ level (τ_{Yb}) and is equal to 4.4 msec. It turns out that only Er^{3+} ions take part in luminescence at $\lambda = 2.73 \mu$, and Yb^{3+} ions give the greatest contribution to luminescence at $\lambda = 0.98 \mu$, since the oscillator strength of the $^2F_{5/2}-^2F_{7/2}$ (Yb^{3+}) transition is at least a factor of three greater than the oscillator strength of the transition $^4I_{11/2}-^4I_{15/2}$ of the Er^{3+} ion. Let us compare the measured times $\tau_{Er} = \tau_{Yb} = 4.4$ msec with the calculated τ_{av}. From Eqs. (1) and (2) in the case of a system of strongly bound ions ($P_{11}, P_{12}, P_{22} \gg 1/\tau_{Yb}^0, 1/\tau_{Er}^0$) for $n_{Yb} = n_{Er}$, $\tau_1 = \tau_{Er}^0 = 8.8$ msec, and $\tau_2 = \tau_{Yb}^0 = 3.0$ msec we find $\tau_{av} = 4.43$ msec. It is evident that the measured value is in good agreement with the theoretical value.

Thus, in this system the Er^{3+} damping time τ_{Er} falls to 4.4 msec, while the time for Yb^{3+} in the presence of Er^{3+} increases to 4.4 msec. Here it turned out that a simultaneous increase in the concentration of Er and Yb from 3 to 10 weight% does not lead to any change in τ_{Er} or increase in quenching, while in the region of low concentrations less than 3 weight% the quenching increases very rapidly. These facts confirm the existence of strong coupling in this resonance system and, as a consequence, the appearance of discharge saturation in the acceptor (Yb^{3+}) if we are discussing excitation of
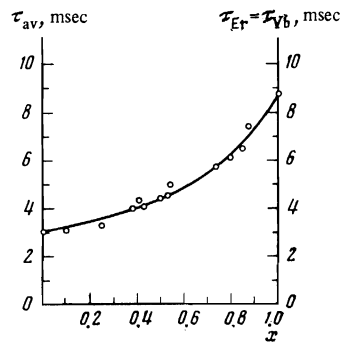

FIG. 2. Concentration dependences of measured lifetimes $\tau_{Er} = \tau_{Yb}$ (points) and calculated τ_{av} (the line) for fluorite.

Er^{3+} ions, and excitation trapping in pumping to the Yb^{3+} absorption bands.

A more detailed analysis of the effect observed by us has been carried out on the system $CaF_2 + Ef_xYb_{1-x}F_3$ 10 weight% ($x = 0.1, 0.25, 0.38, 0.41, 0.43, 0.5, 0.54, 0.74, 0.8, 0.85,$ and 0.87). The damping kinetics of these crystals also was measured in transitions with wavelengths of 0.98 and 2.73 μ with selective excitation of Er^{3+} ($\lambda = 5320$ Å) and Yb^{3+} ($\lambda = 9100$ Å). Here it turned out to be exponential and the lifetimes in all measurements agreed with the error of ± 10% for each single crystal.

The points in Fig. 2 show the concentration dependence on the parameter x of the lifetimes $\tau_{Er} = \tau_{Yb}$ averaged over all measurements. Here the solid line shows the theoretical dependence of τ_{av} in accordance with Eq. (2). As can be seen from comparison of these dependences, they agree quite well. Let us analyze the dependence obtained, $\tau_{Er} = \tau_{av} = f(x)$. It can be seen from Fig. 2 that in the region of small x, i. e., when $n_{Yb} \gg n_{Er}$, quenching of erbium occurs (the limiting case of static quenching). The lifetime of the excited level of the donor (erbium) in this case is simply equal to intrinsic lifetime of the acceptor (ytterbium) $\tau_{Er} = \tau_{Yb}^0 = 3$ msec, which agrees with the theoretical[19,25] and experimental results.[26,27] In the case of large x ($n_{Er} \gg n_{Yb}$) quenching is the limiting case of migration (supermigration) and in our case is very weak. This is due to the low rate of relaxation in the acceptor ($1/\tau_{Yb}^0$), which cannot provide a rapid delivery of excitations from the donor system.

It should be noted that the essential independence (Fig. 2) of the lifetime of the excited level of the Er^{3+} donors (τ_{Er}) on the concentration of the Yb^{3+} acceptor ions for $n_{Yb} \gg n_{Er}$ does not signify constancy of the quantum yield of luminescence from this level. This is due to the fact that the kinetic measurements made by us did not include the very first stage of the process when for a time P_{12}^{-1} a sharp drop in the donor level population should occur in order to provide equalization of the populations between the two subsystems $n_1^*/n_1 = n_2^*/n_2$. It is just this fact, as assumed by Burshtein and Pusep,[19] which will lead to a drop in the quantum yield of the donors with increasing acceptor concentration, even for a constant τ of the donors: $\eta \sim n_D/(n_D + n_A)$. The experi-

mental verification of this assumption is still waiting to be carried out.

Let us analyze now the dependence $\tau_{Yb} = \tau_{av} = f(x)$. It can be seen from Fig. 2 that for large concentrations ($x \ll 1$) ytterbium has an intrinsic lifetime $\tau_{Yb} = \tau_{Yb}^0 = 3$ msec. However, an increase of the relative concentration of erbium ions, which have a lower probability of decay of the excited level, leads to trapping of part of the excitation in them and, as a consequence, to a reduction in the total rate of relaxation of excitations in this system, which appears in a lengthening of the lifetime of the $^2F_{5/2}$ excited level of Yb^{3+} ions (the excitation trapping effect). The maximum lifetime τ_{Yb} observed by us in this system is 7.4 msec, i.e., 2.5 times greater than the intrinsic lifetime.

Thus, in doubly activated crystals it is actually possible to obtain a significant increase in the lifetime of a working laser level by introducing a second impurity which has a level resonant to the laser but with a larger intrinsic lifetime τ_1^0. As can be seen from Eq. (2), τ_2 will increase with increase of the ratio n_1/n_2, i.e., with increase of the concentration of the additional impurity n_1. The maximum value is $\tau_2^{max} = \tau_{av} = \tau_1^0$, when $n_1/n_2 \gg 1$. However, there is also a limitation on the value of n_1/n_2. In the case where τ_1^0 is not very large, but of the order of τ_2^0, large losses of excitation arise to radiation through the resonance level of the additional impurity (1). The expression for the quantum yield of the working level can be written

$$\eta_2 = \frac{\tau_{av}}{\tau_2^0} \frac{n_2}{n_1+n_2} = \frac{n_2/\tau_2^0}{n_1/\tau_1^0 + n_2/\tau_2^0}. \tag{3}$$

From Eqs. (2) and (3) it is evident that a large increase of τ_2 with a high quantum yield can be achieved only by choosing a system with greatly differing spontaneous lifetimes: $\tau_1^0 \gg \tau_2^0$. We note that exchange of electronic excitation between the impurity subsystems occurs with a finite rate P_{12}. The duration of the illumination Δt_{gen}, depending on the mode, can be varied over a very wide range, extending in the giant-pulse mode down to 10^{-8} sec. Here, to realize the effect is necessary to assure fulfillment of the condition of rapid equalization of the populations between the buffer and working ions ($P_{12}^{-1} < \Delta t_{gen}$). The experimental increase of P_{12} can be achieved by building up the total concentration of impurities ($n_1 + n_2$). The reduction occurring in this case of the distances between the impurities of the same and different types (R_{11}, R_{22}, R_{12}) leads to a significant increase in the interactions P_{11}, P_{22}, and P_{12}. We shall discuss a specific example of the possible use of the excitation trapping effect.

It is well known that Er^{3+} and Yb^{3+} ions implanted in various matrices are often used to obtain stimulated radiation (see the references cited by Kaminskiĭ[29]). In particular, lasing has recently been obtained with wavelength λ near 3 μ in the transition $^4I_{11/2}-^4I_{13/2}$ of Er^{3+} ions in crystals of CaF_2 (Ref. 30) and $Y_3Al_5O_{12}$ (Ref. 31). In addition to such undoubted advantages of a garnet base as low lasing threshold, good spectral and lasing properties, and also high optical, thermal, and mechanical qualities, it also has deficiencies. The most important
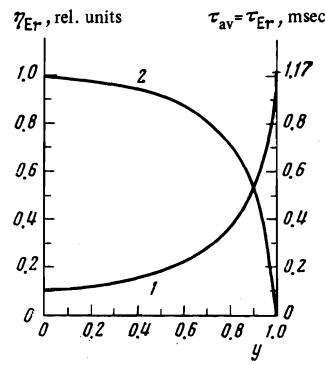

η_{Er}, rel. units $\tau_{av} = \tau_{Er}$, msec

FIG. 3. Theoretical concentration dependences of $\tau_{Er} = \tau_{av}$ (curve 1) and η_{Er} (curve 2) for yttrium–aluminum garnet.

of them is the very short lifetime of the upper laser level $^4I_{11/2}$ of the Er^{3+} ion in garnet in comparison with the lower level. Our measurements have shown that $\tau(^4I_{11/2})$ for small concentrations of Er^{3+} ions is 0.1 msec, while the τ of the lower working level $^4I_{13/2}$ is 6.4 msec. This ratio of the lifetimes strongly limits the possibilities of operation of an erbium laser just to the regime of self blocking and does not permit efficient use of the energy of pumping lamps, which have a duration $\Delta t \sim 1$ msec. In fact, as can be seen from the work of Zharikov et al.,[31] lasing in ytterbium-erbium garnet occurs only at the beginning of the pumping pulse in a narrow time interval and then is rapidly cut off as a result of the rise of population of the lower level. Introduction of a small quantity of Ho^{3+} ions permits some reduction of the lifetime of the $^4I_{13/2}$ level, but this is insufficient to remove the self-blocking effect.

In an earlier study we investigated the lifetimes of Yb^{3+} ions in garnet crystals. We showed that for sufficiently pure materials it is possible to obtain garnet crystals with 50% Yb and $\tau(^2F_{5/2})$ at 300 K close to the radiative value $\tau_{Yb}^0 = 1.17$ msec. Comparison of the level schemes of Er^{3+} (Ref. 31) and Yb^{3+} (Ref. 32) in garnet showed good agreement of the energies of the transitions $^2F_{5/2}-^2F_{7/2}$ of the Yb^{3+} ion and $^4I_{11/2}-^4I_{15/2}$ of the Er^{3+} ion, which permits us to expect an effective $Er^{3+} \rightarrow Yb^{3+}$ interaction. The analysis of these data and consideration of the experimental results described above on observation of strong $Er^{3+} \rightarrow Yb^{3+}$ coupling in fluorite permits suggestion of the excitation trapping effect to increase the effective lifetime of the upper laser level $^4I_{11/2}$ of an erbium-garnet laser. As can be seen from Eq. (2), the maximum possible lifetime of the $^4I_{11/2}$ level in this case is $\tau_{Er} = \tau_{Yb}^0 = 1.17$ msec (when $n_{Yb}/n_{Er} \gg 1$). The limitation on the maximum value of n_{Yb}/n_{Er} is due to the need for absence of a rapid drop in quantum yield of the $^4I_{11/2}$ level of the Er^{3+} ion on introduction of Yb^{3+}.

The proposed dependences of $\tau_{Er} = \tau_{av}$ and η_{Er} on $y = n_{Yb}/(n_{Yb} + n_{Er})$ in accordance with Eqs. (2) and (3) for the (Yb_yEr_{1-y}) system in yttrium–aluminum garnet are shown in Fig. 3. It can be seen from this figure that for $y = 0.8$ it is possible to obtain a lengthening to $\tau_{Er} = 3.7 \tau_{Er}^0$ with rather high quantum yield $\eta_{Er} = 0.75$, and for $y = 0.9$ we have $\tau_{Er} = 5.6 \tau_{Er}^0$ and $\eta_{Er} = 0.56$. We recall that the drop in quantum yield is not due to radia-

tionless transitions, but is due to radiation of Yb^{3+} ions. Reabsorption of this radiation in laser crystals by Yb^{3+} ion themselves and also by Er^{3+} can result in some increase in quantum yield of the $^4I_{11/2}$ level of the Er^{3+} ions in comparison with the theoretical value.

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[1] Here and subsequently we mean by strong interaction $\delta \gg P_{DA} > 1/\tau_D^0$, $1/\tau_A^0$. That is, the probability of interaction (P_{DA}) does not exceed the rate of phase relaxation (δ) in the system. For rare-earth ions in crystals the reverse situation $(\delta < P_{DA})$ is unlikely. This is due to the high probability of direct transitions between the Stark sublevels of one multiplet in absorption and emission of phonons (a large value of δ) and to the strong forbiddenness of the electronic transitions which determine the value of P_{DA}. Our investigations have shown that the condition $P_{DA} \gg 1/\tau_D^0$, $1/\tau_A^0$ can be realized experimentally.

[2] An example of the investigation of such systems but in the case of weak coupling is study of the transfer of energy between rare-earth ions in paired rhombic centers in fluorite crystals. [18]

[3] The only examples we can give are: 1) study of the transfer of energy between cubic and complex centers for Yb^{3+} ions[26] and from tetragonal to complex centers for Nd^{3+} ions[27] in CaF_2 crystals, where a minimum donor damping time equal to the acceptor damping time $\tau_1 = \tau_2$ (for $n_2 \gg n_1$) has been observed, and 2) observation of a small increase (by 10%) in the damping time of infrared luminescence in the resonance interaction of Er^{3+} and Yb^{3+} ions in LaF_3 (Ref. 28).

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