

## ELECTRONIC EXCITATION DECAY IN $\text{LiNbO}_3\text{:Yb}^{3+}$ CRYSTALS IN THE PRESENCE OF PAIR IMPURITY CENTERS

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**Abstract:** The influence of phenomenon of electronic excitation trapping, caused by processes of nonradiative redistribution of electronic excitation energy (NREEE) in impurity subsystem, on kinetic characteristics of excitation decay in  $\text{LiNbO}_3\text{:Yb}^{3+}$  crystals is investigated. Probabilities of elementary acts of the NREEE are calculated. On the basis of the simplest model the kinetic characteristics of electronic excitation decay with taking into account the presence of both the single and pair impurity centers in crystalline matrix in the case of small concentration of impurity ions are calculated. The dependences of electronic excitation lifetime and luminescence quantum yield on the impurity concentration and pump power are determined.

**Keywords:**  $\text{LiNbO}_3\text{:Yb}^{3+}$  crystals, pair center, excitation trapping, kinetics of excitation decay

### 1. Introduction

The possibility of combining, within the same framework of one monocrystal, both diverse “active” (lasers, amplifiers, etc.) and “passive” (modulators, frequency converters, etc.) devices has increased the interest of many researchers in rare earth ( $\text{RE}^{3+}$ ) ion-doped  $\text{LiNbO}_3$  (LN) crystals [1-3]. It is known that the distribution of  $\text{RE}^{3+}$  ions in the matrix of LN crystals is considerably inhomogeneous therefore even at small  $\text{RE}^{3+}$  concentration ( $\sim 1\%$ ), along with single impurity centers (SC), it is quite possible the formation of some quantity of pair impurity centers (PC) consisted of two  $\text{RE}^{3+}$  ions separated each from other by a distance of  $\sim 3\text{--}4 \text{ \AA}$  [4-7]. In particular, in [5-7] on the basis of spectroscopic and electron paramagnetic resonance measurements it was established the presence of impurity PCs in matrix of LN crystal. Evidently, between the closely set of  $\text{RE}^{3+}$  ions in PC the effective nonradiative redistribution of electronic excitation energy (NREEE) takes place. In turn the NREEE process leads to “excitation trapping” phenomenon [8, 9]. Some models of distribution of impurity ions in the crystalline matrix of  $\text{LN:Yb}$  and influence of inhomogeneous distribution of impurity ions on kinetics of electronic excitation decay in impurity subsystem are discussed in [4]. Redistribution of electronic excitations energy in impurity subsystem of  $\text{LN:RE}$  crystals caused by NREEE processes, as well as kinetics of electronic excitation decay, inclusive of cooperative and up-conversion luminescence have been studied in many works (see, for example, [10-14]). In particular, kinetic characteristics of electronic excitation decay in  $\text{LiNbO}_3\text{:Yb}^{3+}$  crystals with taking into account the presence of both the SCs and PCs in crystalline matrix in the case of high concentration of impurity ions (when it is possible to neglect

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the phenomenon of excitation depletion) have been calculated in [10].

In this paper on the basis of the simplest model proposed in [10] we calculate the kinetic characteristics of electronic excitation decay in the impurity subsystem of  $\text{Yb}^{3+}$ -doped LN crystal in the case of low concentration of impurity ions.

## 2. Modeling calculation

As a starting point we assume that both SCs and PCs are present in the matrix of LN crystal at any concentrations of impurity ions. Our calculation is based on the simplest model proposed in [10], within the framework of which the PCs and SCs are considered as “acceptors” and “donors” with excitation relaxation lifetime  $\tau_a$  and  $\tau_d$ , respectively. Moreover, it is assumed that NREEE processes between PCs, as well as the energy transfer from PC to single centers, can be neglected, so that the electronic excitation found oneself in a PC does not leave it before relaxation. The lifetimes  $\tau_a$  and  $\tau_d$  are coupled with each other by the expression [9]

$$\tau_a = \tau_d \left( 1 + \frac{1}{a \cdot (e^{\gamma/a} - 1)} \right), \quad (1)$$

where  $a = W/W_o$  ( $W$  and  $W_o$  are probabilities of elementary act of NREEE between  $\text{RE}^{3+}$  ions in PC and of intra-center transitions, respectively),  $\gamma$  is a positive parameter approximately equal to unity, its correct value can be determined by comparing calculated and experimental values of kinetic characteristics of excitation decay. It is seen that in the limit of fast NREEE process in PC, from (1) we have  $\lim_{W \rightarrow \infty} \tau_a = (1 + 1/\gamma) \cdot \tau_d \approx 2\tau_d$ .

The kinetic equations for the numbers of excited “donors”  $n_d$  and “acceptors”  $n_a$  can be written in the form [15]

$$\dot{n}_d = -\frac{1}{\tau_d} \cdot n_d - w_1 \cdot n_d; \quad \dot{n}_a = -\frac{1}{\tau_a} \cdot n_a + w_1 \cdot n_d \quad (2)$$

with the initial conditions  $n_d(0) = n_{d0}$ ;  $n_a(0) = n_{a0}$ . Here  $w_1$  is the rate of excitation transfer from SC to PC, which, within the framework of the Forster–Dexter’s theory (the case of small concentration of acceptor), when the excitation energy transfer from donor to acceptor is induced by interaction with dipole-dipole coupling, is determined by the following expression [15-18]:

$$w_1(t) = (\tau_d t)^{-\frac{1}{2}} \cdot q, \quad (3)$$

where  $q = 2\pi^{3/2} R_c^3 N_a / 3$  ( $N_a$  is the concentration of acceptor,  $R_c$  is the donor-acceptor critical distance determined from the condition  $W_1(R_c) \cdot \tau_d = 1$ ,  $W_1$  is the probability of elementary act of energy transfer from donor to acceptor). Solution of Eq. (2) leads to the following laws for

excitation decay [15]:

$$n_d(t) = n_{d0} \cdot \exp\left(-t/\tau_d - 2q\sqrt{t/\tau_d}\right), \quad (4)$$

$$n_a(t) = n_{a0} \cdot \exp(-t/\tau_a) + n_{d0} \cdot \sqrt{\pi} q_1 \exp\left(q_1^2 - \frac{t}{\tau_a}\right) \cdot \left\{ \Phi(q_1 + \sqrt{\alpha \cdot t}) - \Phi(q_1) \right\}, \quad (5)$$

where  $q_1 = q \cdot (1 - \tau_d \cdot \tau_a^{-1})^{-1/2}$ ,  $\alpha = \sqrt{\tau_d^{-1} - \tau_a^{-1}}$ ,  $\Phi(z) = \frac{2}{\sqrt{\pi}} \int_0^z \exp(-x^2) dx$  is the error integral.

With taking into account Eq. (4) and Eq. (5) for the lifetime of excitation in the whole impurity system

$$\tau = \int_0^\infty t \cdot (n_a + n_d) dt \bigg/ \int_0^\infty (n_a + n_d) dt \quad (6)$$

and for the quantum yield of luminescence

$$\eta = \frac{1}{(n_{a0} + n_{d0})} \left\{ \frac{1}{\tau_{ar}} \int_0^\infty n_a(t) dt + \frac{1}{\tau_{dr}} \int_0^\infty n_d(t) dt \right\}, \quad (7)$$

it is easy to obtain the following expressions:

$$\tau = (n_{a0} \tau_a^2 + n_{d0} \tau_d^2 A) / (n_{a0} \tau_a + n_{d0} \tau_d B) \quad (8)$$

and

$$\eta = \frac{n_{a0} \eta_a + n_{d0} \eta_d B_\eta}{n_{a0} + n_{d0}}, \quad (9)$$

where

$$A = 1 + q^2 - \frac{1}{2} \sqrt{\pi} q e^{q^2} (3 + 2q^2) \cdot (1 - \Phi(q)) - \sqrt{\pi} q_1 e^{q_1^2} \left[ \frac{\tau_a^2}{\tau_d^2} \cdot \Phi(q_1) - \frac{1}{\tau_d^2} \cdot \int_0^\infty t \exp(-t/\tau_a) \Phi(q_1 + \alpha \sqrt{t}) dt \right], \quad (10)$$

$$B = 1 - \sqrt{\pi} q e^{q^2} (1 - \Phi(q)) - \sqrt{\pi} q_1 e^{q_1^2} \left[ \frac{\tau_a}{\tau_d} \cdot \Phi(q_1) - \frac{1}{\tau_d} \cdot \int_0^\infty \exp(-t/\tau_a) \cdot \Phi(q_1 + \alpha \sqrt{t}) dt \right], \quad (11)$$

$\eta_d = \tau_d / \tau_{dr}$  and  $\eta_a = \tau_a / \tau_{ar}$  are quantum yields of donor and acceptor luminescence, respectively ( $\tau_{ar}$  and  $\tau_{dr}$  are radiative lifetime of the excited levels of donor and acceptor ions); expression for  $B_\eta$  will coincide with the one for B, if the square bracket on the right side of Eq. (11) will be multiplied by  $\tau_{dr} / \tau_{ar}$ . Note, when the excitation trapping is absent ( $\tau_a = \tau_d$ ), we have  $q_1 = 0$  and  $\alpha = 0$ , so that Eq. (10) and Eq. (11) transform into the known expressions [15]:

$$A = 1 + q^2 - \frac{1}{2} \sqrt{\pi} q e^{q^2} (3 + 2q^2) \cdot (1 - \Phi(q)), \quad B = 1 - \sqrt{\pi} q e^{q^2} (1 - \Phi(q)). \quad (12)$$

As seen from Eq.(8) and Eq.(9), when initially there are only excited PCs ( $n_{d0} = 0$ ,  $n_{a0} \neq 0$ ), then

$\tau = \tau_a$  and  $\eta = \eta_a$ . On the other hand, when initially only SCs are excited ( $n_{d0} \neq 0$ ,  $n_{a0} = 0$ ), then  $\tau = (A/B)\tau_d$  and  $\eta = \eta_d B_\eta$ , i.e. in this case excitation lifetime and luminescence quantum yield are not similarly varied.

Thereby, in order to find dependences of kinetic characteristics of excitation decay in the whole impurity system on the impurity concentration, pump power, and so on, one needs to determine elementary act probabilities of NREEE between impurity ions within a PC, as well as the rates of excitation energy transfer from SC to PC.

### 3. Nonradiative transfer of electronic excitation energy in LN:Yb<sup>3+</sup> crystals

Energy levels scheme of Yb<sup>3+</sup> ion in LN crystal, plotted on the basis of absorption and emission spectra obtained in [19,20], is shown in Fig.1. Energy spectrum of Yb<sup>3+</sup> ion consists of only two manifolds, <sup>2</sup>F<sub>7/2</sub> and <sup>2</sup>F<sub>5/2</sub>, separated from each other by  $\sim 10000 \text{ cm}^{-1}$ , values of their Stark splitting are in the region of  $200\text{--}400 \text{ cm}^{-1}$ . Note that Stark levels  $\nu_2$ ,  $\nu_3$ ,  $\nu_4$  and  $\nu_6$ ,  $\nu_7$  are sufficiently separated from the corresponding ground Stark levels  $\nu_1$  and  $\nu_5$  of <sup>2</sup>F<sub>7/2</sub> and <sup>2</sup>F<sub>5/2</sub> manifolds (Fig. 1). It makes possible to neglect the Boltzmann distribution of excitation over Stark states, i.e. to suppose that in initial moment donor ion was in the  $\nu_5$  state and acceptor ion was in  $\nu_1$  state. On the other hand, the energy of Debye phonon is equal to  $350 \text{ cm}^{-1}$  for LiNbO<sub>3</sub> crystal [3], so NREEE processes can be efficiently realized by three channels: one resonance

$$\nu_5(^2F_{5/2}) \rightarrow \nu_1(^2F_{7/2}): \nu_1(^2F_{7/2}) \rightarrow \nu_5(^2F_{5/2}) \quad (13)$$

and two nonresonance (with participation of one crystal phonon)

$$A: \nu_5(^2F_{5/2}) \rightarrow \nu_1(^2F_{7/2}): \nu_1(^2F_{7/2}) \rightarrow \nu_6(^2F_{5/2}) \quad (14)$$

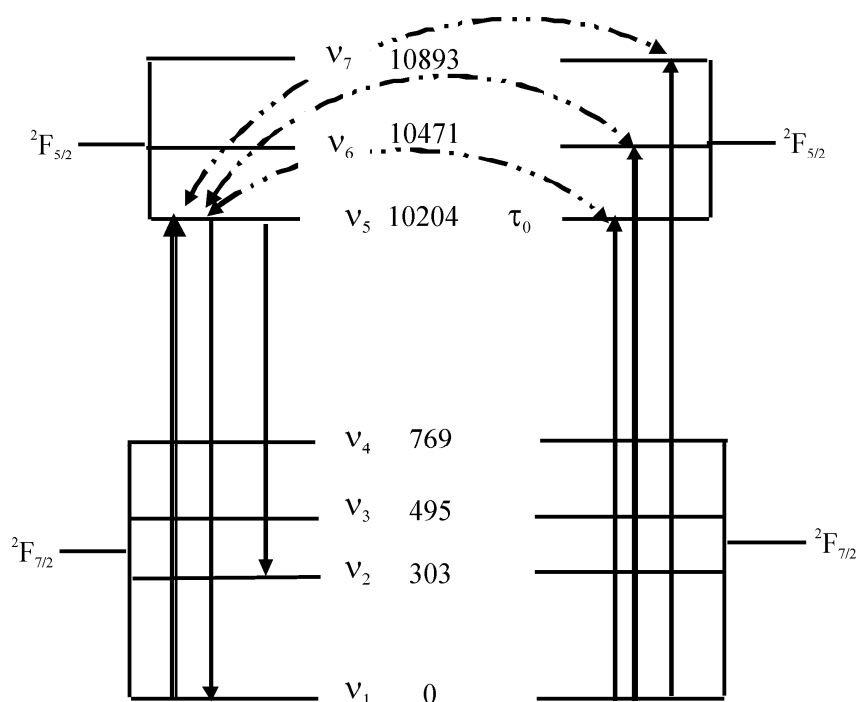
$$B: \nu_5(^2F_{5/2}) \rightarrow \nu_2(^2F_{7/2}): \nu_1(^2F_{7/2}) \rightarrow \nu_5(^2F_{5/2}). \quad (15)$$

We will suppose that NREEE processes are induced by multipole-multipole (including forced dipole-dipole) resonance and nonresonance interactions of Yb<sup>3+</sup> ions, each of which leads to two mechanisms of energy transfer: direct and exchange [15-18, 21]. (Note that NREEE can be also induced by electron-phonon interaction of impurity ions with crystal phonons [15, 22-25]. But we will leave out of account the contribution of this mechanism because of it, as our estimations have revealed, is negligibly small [10]). So, the probability of elementary act of NREEE between Yb<sup>3+</sup> ions separated from each other by a distance R can be schematically presented in the form

$$W(R, T) = W_{\mu\mu}^{(r)}(R, T) + W_{fdd}^{(r)}(R, T) + W_{exc}^{(r)}(R, T) + \\ + W_{\mu\mu}^{(nr)}(R, T) + W_{fdd}^{(nr)}(R, T) + W_{exc}^{(nr)}(R, T), \quad (16)$$

where the first three terms are related to resonance and the last three terms to nonresonance multipole-multipole, forced dipole-dipole and exchange mechanisms of NREEE, respectively, T is

temperature. Evidently, the probabilities of exchange mechanisms are proportional to the overlap integral of radial wave functions of impurity ions, therefore these mechanisms, in fact, are efficient when the donor-acceptor distance is small enough (such as in the case of larger concentrations of impurity ions, or for  $\text{Yb}^{3+}$  ions in PC, the distance between of which is equal to  $\sim 3\text{--}4 \text{ \AA}$ ). Evidently, in the case of small concentration of impurity ions only forced dipole-dipole mechanism leads to a considerable contribution in the rate of energy transfer from SC to PC. At the same time, all terms in Eq. (16) must be taken into account to describe the NREEE within PC. Quantitative calculations of the elementary act probabilities of the resonance and nonresonance energy transfer passed in compliance with the schemes (13)–(15), have been carried out in [10]. The results of these calculations for the above mentioned mechanisms, as well as the values of the corresponding critical distances determined from the condition  $W(R_c) \cdot \tau_d = 1$  (where  $\tau_d = 728 \text{ \mu s}$  is the lifetime of the excitation in SC [20]) for each mechanism under consideration are given in table. It is seen that resonance mechanisms are more efficient and the total probability of NREEE within PC is equal to  $1.9 \cdot 10^6 \text{ s}^{-1}$ , that is considerably larger than the probability of intra-center transition,  $1.373 \cdot 10^3 \text{ s}^{-1}$ . So, according to (1), the lifetime of excitation in PC is equal to  $\tau_a = 1452.2 \text{ \mu s}$  which is approximately two times larger than  $\tau_d$ . In addition, using the analytical expressions of the elementary act probabilities of NREEE given in [10] we found that the critical distance of energy transfer caused by total, resonance and nonresonance, forced dipole-dipole transitions is largest,  $R_c = 11.2 \text{ \AA}$ . That is what we were limited by taking into account of only forced dipole-dipole mechanism when we used Eq. (3) for the rate of energy transfer from SC to PC.



**Fig. 1.** Scheme of Stark levels of  $\text{Yb}^{3+}$  in  $\text{LiNbO}_3$  crystal.

Probabilities of elementary acts of nonradiative energy transfer [10].

Energy transfer mechanisms		Mechanisms of NREEE			
		Resonance	Non resonance		
			A	B	Total
<sup>c</sup> μμ	<sup>a</sup> R <sub>c</sub> , Å	8.2	4.12	3.97	4.34
	<sup>b</sup> W, s <sup>-1</sup>	1.5·10 <sup>6</sup>	1.85·10 <sup>3</sup>	1.27·10 <sup>3</sup>	3.12·10 <sup>3</sup>
<sup>d</sup> fdd	<sup>a</sup> R <sub>c</sub> , Å	7.9	8.34	9.06	9.81
	<sup>b</sup> W, s <sup>-1</sup>	8.4·10 <sup>4</sup>	1.13·10 <sup>5</sup>	1.86·10 <sup>5</sup>	2.98·10 <sup>5</sup>
<sup>e</sup> exch.	<sup>a</sup> R <sub>c</sub> , Å	4.1	3.29	3.44	3.45
	<sup>b</sup> W, s <sup>-1</sup>	3.3·10 <sup>3</sup>	7·10 <sup>-3</sup>	0.1	~ 0.1

<sup>a</sup> critical distance; <sup>b</sup> probability of elementary act of NREEE at T = 300K and R = 4 Å;  
<sup>c, d, e</sup> multipole-multipole, forced dipole-dipole and exchange mechanisms of NREEE.

#### 4. Electronic excitation decay in LiNbO<sub>3</sub>:Yb<sup>3+</sup> crystals

Now we determine the kinetic characteristics of electronic excitation decay in LiNbO<sub>3</sub>:Yb<sup>3+</sup> crystals when the system under consideration is excited by short squared excitation pulse with duration  $\tau_p$ . So, using the values  $\tau_d = 728.2 \mu s$ ,  $\tau_a \approx 2\tau_d$ ,  $R_c = 11.2 \text{ Å}$ , at first, we find  $q = 0.522 \times 10^{20} N_a$ ,  $q_1 = 0.738 \times 10^{20} N_a$  and  $\alpha = 26.204$ , and then by Eq.(10) and Eq.(11) we obtain the expressions for the parameters A and B depended on acceptor concentration  $N_a$ . Dependences of the excitation lifetime in the whole impurity subsystem and the quantum yield of the luminescence on the wavelength  $\lambda = 1010 \text{ nm}$ , corresponding to  $v_5 \rightarrow v_2$  transition, on the relative concentration of initial excited PCs ( $n_{a0}/N_{yb}$ ) are determined by Eq. (8) and Eq. (9). The analytical expressions, obtained in this way, are lengthy therefore here we present the plots of these dependences for several values of the number of initially excited SCs, assumed that  $N_{yb} = 10^{20} \text{ cm}^{-3}$  and  $N_a = 10^{19} \text{ cm}^{-3}$  (Fig.2). In doing so we use the calculated values of probabilities of radiative transitions ( $\tau_{dr}^{-1} = \tau_{ar}^{-1} = 568.8 s^{-1}$ ) on the wavelength  $\lambda = 1010 \text{ nm}$  [20]. As is obvious from Fig.2 when initially excited PC's concentration increases the lifetime of excitation and the quantum yield of luminescence on the considered wavelength are increased beginning from the values  $\tau(0) = 1.61 \cdot \tau_d$  and  $\eta(0) = 1.04 \cdot \eta_d$ , respectively. Thus, even through PCs have not been excited at initial moment (i.e. when  $n_{a0} = 0$ ) there is a substantial delay of excitation decay. Of course, it can be explained by transfer of excitation energy from SC to PC.

In order to determine the corresponding dependences on PC's and SC's concentrations for several values of pump intensity we assume that the concentrations of PCs and SCs, which were

excited at initial time, can be described by the expressions:

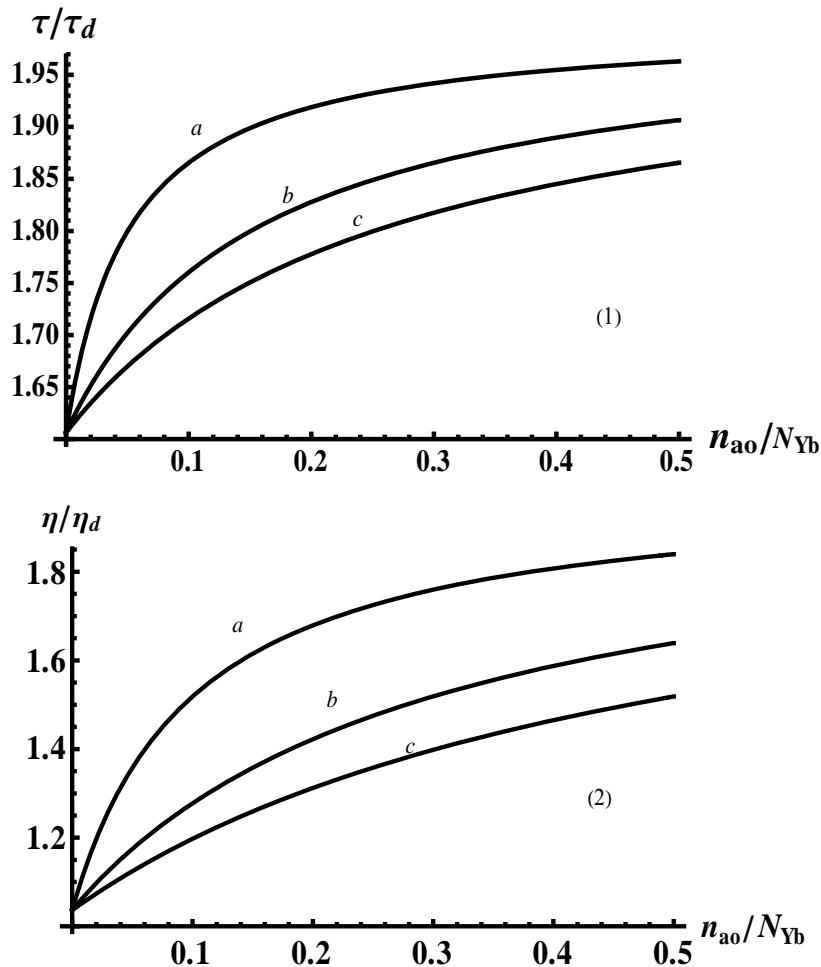
$$n_{a0} = 2N_{pc} \cdot \exp(-\sigma_p F \tau_p) \cdot [1 - \exp(-\sigma_p F \tau_p)], \quad (17)$$

$$n_{d0} = (N_{yb} - 2N_{pc}) \cdot [1 - \exp(-\sigma_p F \tau_p)]. \quad (18)$$

Note that the concentration of acceptors depends on the pump intensity because of the PCs, both ions of which are initially excited, cannot be considered as acceptors, under used assumptions. Thereby, the concentration of acceptors is determined by the expression:

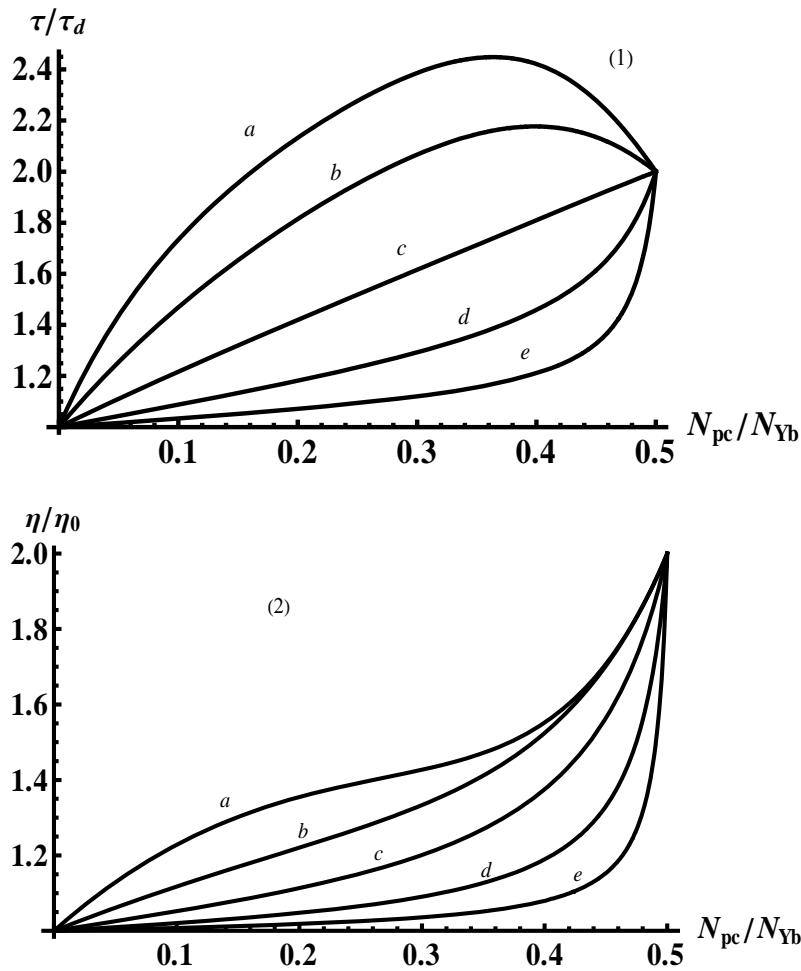
$$N_a = N_{pc} \times [2 \exp(-\sigma_p F \tau_p) - \exp(-2\sigma_p F \tau_p)]. \quad (19)$$

Here  $\sigma_p$  is the absorption cross-section of  $\text{Yb}^{3+}$  ion,  $F$  is the photon flux density in pulse of the incident radiation,  $N_{pc}$  and  $N_{yb}$  are the concentrations of PC and  $\text{Yb}^{3+}$  ions, respectively. We use Eq. (19) in order to find the value of the basic parameter ( $q$ ) of excitation energy transfer from donor to acceptor. Evidently, if the PCs are absent ( $N_{pc} = 0$ ), then  $q = q_1 = 0$ , and so  $A = B = 1$ , and Eq. (8) and Eq. (9) lead to equations  $\tau = \tau_d$  and  $\eta = \eta_d$ , respectively. Note that, as expected, Eq. (19) leads to the exactly same result in the case of very intense pumping ( $\sigma_p F \tau_p \rightarrow \infty$ ).



**Fig. 2.** Dependences of excitation lifetime (1) and of quantum yield (2) on the relative concentration of excited PC, when  $N_a = 0.1 \times N_{Yb} = 10^{19} \text{ cm}^{-3}$ : a)  $n_{d0} = 0.5 \times N_{Yb}$ ; b)  $n_{d0} = 0.3 \times N_{Yb}$ ; c)  $n_{d0} = 0.1 \times N_{Yb}$ .

Substituting Eq. (17) and Eq. (18) into Eq. (8) and Eq. (9), we obtain the dependences of excitation lifetime in the whole impurity system and of luminescence quantum yield on the concentration of PCs. Plots of these dependences for several values of the pump power are presented in Fig.3. As shown from Fig.3, the excitation lifetime and the luminescence quantum yield on the wavelength  $\lambda = 1010$  nm are more slowly increased at more large values of the pump power. In fact, it is explained by decreasing the acceptor concentration with increasing the pump power. Really, at the strong pumping the number of the full excited PCs, which cannot be considered as acceptors, becomes larger, and so the concentration of acceptors decreases. Thus, these dependences permit to determine the concentration of PCs via the experimental values of the excitation lifetime and the luminescence quantum yield.



**Fig. 3.** Dependences of excitation lifetime (1) and quantum yield of luminescence at 1010 nm wavelength (2) on the relative PC concentration at  $N_{Yb} = 10^{20} \text{ cm}^{-3}$ : a)  $\sigma_p F \tau_p = 0.01$ ; b)  $\sigma_p F \tau_p = 1$ ; c)  $\sigma_p F \tau_p = 2$ ; d)  $\sigma_p F \tau_p = 3$ ; e)  $\sigma_p F \tau_p = 4$ .

## 5. Conclusions

Thereby, the proposed calculation model enables to determine the concentration of PC via the experimental data of some kinetic characteristics of excitation decay measured at several pump power. But it is needed to emphasize that we limited ourselves to consideration of the case of



enough small concentrations of impurity ions and the obtained analytical expressions are corrected for only such crystal systems. Of course, in the more general case we must take into account some additional NREEE processes, such as energy migration within donor subsystem, excitation energy transfer from PC to SC and from PC to PC, etc. Nevertheless we believe that for doped crystal systems, including some quantity of PCs, the main idea promoted in this paper (namely, partition of the whole impurity system on the donor and acceptor subsystems with the variable concentrations) is also suitable, even when the concentration of PC is not small.

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