

Microscopic theory for excitation of erbium ions via silicon nanocrystals in silicon dioxide

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Abstract

In this work, we present the results of microscopic calculation for the probability of excitation of Er ions by electron–hole pairs confined in Si nanocrystals surrounded by silicon dioxide. For simplicity we consider the case of low pumping of nanocrystals when we may take into account only one electron–hole pair inside of a single nanocrystal. We have found the probability of Er ion excitation in three principally different cases for the location of the Er ion relatively to the nanocrystal: (i) Er ion is situated inside of the nanocrystal, (ii) Er ion is situated at the boundary of the nanocrystal or very close to the boundary, and (iii) Er ion is situated at the considerable distance from the nanocrystal so that the tunneling of electrons and holes may be neglected.

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1. Introduction

Since a decade the material composed of Si nanocrystals dispersed in SiO₂ doped by Er is a subject of big interest for creation of amplifiers or lasers on the basis of silicon technology [1,2]. The investigations have got an additional stimulus after an observation of the effective gain at wavelength 1.5 μm in waveguides based on such a material [3]. The gain is a consequence of an effective transfer of the excitation generated in nanocrystals in form of electron–hole pairs (excitons) to Er³⁺ ions [4]. Although till now there is a lot of experimental results concerning this material, the nanocrystals properties and the mechanism of the excitation transfer are not sufficiently understood. Therefore a theoretical investigation of excitons in nanocrystals and the transfer process is a very important issue for improvement of the material characteristics.

2. Theoretical model

In order to understand physical processes under participation of electrons and holes confined in nanocrystals one

has to know their energy spectrum and wave functions. For modelling properties of the nanocrystals we use the envelope function method, which should be a good approximation for not too small nanocrystals with radius $R_{nc} > 2$ nm (they represent a considerable fraction of all nanocrystals). Thus it should be pointed out that we are capable to apply our results directly to describe properties of nanocrystals with the size larger than the average in the experiments [4–6]. However the results obtained can be also used for qualitative understanding of properties of smaller nanocrystals.

2.1. Electron and hole states

The conduction band of bulk Si has six equivalent minima in the first Brillouin zone at positions $\pm\vec{k}_{0,z} = (0, 0, \pm 0.85)k_X$, $\pm\vec{k}_{0,y} = (0, \pm 0.85, 0)k_X$, and $\pm\vec{k}_{0,x} = (\pm 0.85, 0, 0)k_X$, where $k_X = 2\pi/a$ and $a = 0.357$ nm is the lattice constant of Si [7]. The minima are situated in the neighborhood of the six X -points (there are three non-equivalent X -points). The conduction band is doubly degenerate at each of the X -points, which is a consequence of the fact that Si lattice has two atoms in the elementary unit cell and the origin can be chosen at the center of any of them.

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Assuming that the Bloch amplitudes do not change much in the neighborhood of the X -points and taking their symmetry properties [8,9] into account, we can write one of the three equivalent ground state wave functions $(\psi_z^c, \psi_y^c, \psi_x^c)$ of an electron in the nanocrystal as

$$\psi_z^c = \zeta^c(\vec{r}) u_c \frac{1}{\sqrt{2}} (e^{i\vec{k}_{0,z}\vec{r}} e^{-ik_0 a/8} + e^{-i\vec{k}_{0,z}\vec{r}} e^{ik_0 a/8}), \quad (1)$$

where u_c is one of two Bloch amplitudes of a bulk electron at the $(0,0,1)k_X$ point in the Brillouin zone such that u_c gives zero overlap integral with Bloch amplitudes of the bottom of the valence band and corresponds to the lower conduction band at the $\vec{k}_{0,z}$ point. This overlap integral with the second Bloch amplitude u_s is not equal to zero and u_s corresponds to the upper conduction band at $\vec{k}_{0,z}$. There is a very important connection between these amplitudes, $u_c = u_s e^{i2k_X z}$, which follows from the symmetry. The envelope function ζ^c in Eq. (1) satisfies the following equation:

$$\frac{\hbar^2}{2m_{\parallel}} \frac{\partial^2}{\partial z^2} \zeta^c(x, y, z) + \frac{\hbar^2}{2m_{\perp}} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \zeta^c(x, y, z) + E \zeta^c(x, y, z) = 0, \quad (2)$$

where $m_{\parallel} = 0.916m_0$, $m_{\perp} = 0.19m_0$ with m_0 being the free electron mass. Eq. (2) has been solved numerically assuming infinitely high energy barrier at the boundary of the nanocrystal and we have found energies of several lowest states, $E_i^c = \tilde{E}_i \hbar^2 / (2m_{\parallel} R_{nc}^2)$ with $\tilde{E}_0 = 34.30$, $\tilde{E}_1 = 49.00$, $\tilde{E}_2 = 67.49$, $\tilde{E}_3 = 80.32$, and corresponding eigenfunctions. More details to the calculation can be found in [10].

If we take into account the finite energy barrier for electrons $U_e = 3.2$ eV [11] and use Bastard boundary conditions [8,12] in the first order of the perturbation theory we get following expression for the electron envelope function of the ground state at $r \geq R_{nc}$

$$\tilde{\zeta}_0^c(r) = -\frac{1}{\kappa_c} \left[\frac{m_0}{m_{\perp}} \frac{\rho}{R_{nc}} \frac{\partial \zeta_0^c}{\partial \rho} + \frac{m_0}{m_{\parallel}} \frac{z}{R_{nc}} \frac{\partial \zeta_0^c}{\partial z} \right]_{r=R_{nc}} e^{-\kappa_c(r-R_{nc})}, \quad (3)$$

where $\rho = \sqrt{x^2 + y^2}$ and $\kappa_c = \sqrt{2m_0(U_e - E_c^0)}/\hbar$.

For description of the valence band structure in Si we use generalization of the Luttinger Hamiltonian [13] in the limit of vanishing spin–orbit coupling, which is justified for Si

$$\hat{H} = (A + 2B)\hbar^2 k^2 - 3B\hbar^2 (\vec{k} \cdot \hat{J})^2, \quad (4)$$

where \hat{J} is the unitary angular momentum operator acting in the space of Bloch amplitudes,

$$A = -\frac{1}{4} \frac{m_h + m_1}{m_h m_1}, B = -\frac{1}{4} \frac{m_h - m_1}{m_h m_1}, \quad (5)$$

$$m_h = \frac{m_0}{\gamma_1 - 2\gamma}, \quad m_1 = \frac{m_0}{\gamma_1 + 2\gamma}, \quad \gamma = \frac{1}{5}(3\gamma_3 + 2\gamma_2). \quad (6)$$

Values of the constants γ_1 , γ_2 , and γ_3 for Si are 4.22, 0.53, and 1.38, respectively [7]. Eigenfunctions of the Hamiltonian (4) can be found as eigenfunctions of the square \hat{F}^2 of the full angular momentum operator $\hat{F} = \hat{L} + \hat{J}$

($\hat{L} = -i\vec{r} \times \partial_{\vec{r}}$) and its projection \hat{F}_z onto the axis z . Eigenvalues of \hat{F}^2 and \hat{F}_z are $F(F+1)$ and M , respectively, where F is non-negative integer and M is integer, which absolute value is not larger than F . The basis of the Bloch amplitudes space can be chosen in the form of spherical components $u_0 = Z$, $u_{\pm} = \mp \sqrt{1/2}(X \pm iY)$ of the corresponding functions $X = yz$, $Y = xz$, and $Z = xy$, of the representation $\Gamma_{25'}$. In order to find hole states with the lowest quantization energies it is sufficient to look at states with $F=0$ and $F=1$. In such a case we have following solutions of the Schrödinger equation assuming infinitely high energy barriers at $r = R_{nc}$, written using spherical coordinate system (r, θ, ϕ)

$$\psi_{00}^P(r, \theta, \phi) = A_n j_1(\lambda_n \beta r / R_{nc}) \sum_{m_1, m_2} C_{1m_1 m_2}^{00} Y_{1m_1}(\theta, \phi) u_{m_2}, \quad (7)$$

$$\psi_{1M}^{SD}(r, \theta, \phi) = A'_n \left[R_n^S(r) Y_{00}(\theta, \phi) u_M + R_n^D(r) \sum_{m_1, m_2} C_{2m_1 m_2}^{1M} Y_{2m_1}(\theta, \phi) u_{m_2} \right], \quad (8)$$

$$\psi_{1M}^P(r, \theta, \phi) = A''_n j_1(\lambda''_n r / R_{nc}) \sum_{m_1, m_2} C_{1m_1 m_2}^{1M} Y_{1m_1}(\theta, \phi) u_{m_2}, \quad (9)$$

where

$$R_n^S(r) = \frac{j_0(\lambda'_n r / R_{nc}) - j_0(\lambda'_n \beta r / R_{nc})}{j_0(\lambda'_n)}, \quad (10)$$

$$R_n^D(r) = -\frac{1}{\sqrt{2}} \frac{j_2(\lambda'_n r / R_{nc})}{j_0(\lambda'_n)} - \sqrt{2} \frac{j_2(\lambda'_n \beta r / R_{nc})}{j_0(\lambda'_n \beta)}, \quad (11)$$

A_n , A'_n , A''_n are normalization constants, $j_l(x)$ are spherical Bessel functions of the order l , $Y_{lm}(\theta, \phi)$ are spherical harmonics, $C_{j_1 m_1 j_2 m_2}^{jm}$ are Clebsch–Gordan coefficients, $\beta = \sqrt{(\gamma_1 - 2\gamma)/(2\gamma_1 + 4\gamma)} \approx 0.505$, coefficients λ_n , λ'_n , λ''_n ($n = 0, 1, 2, \dots$) are found from the equations

$$j_1(\lambda_n \beta) = 0, \quad j_2(\lambda'_n) j_0(\lambda'_n \beta) + j_2(\lambda'_n \beta) j_0(\lambda'_n) = 0, \quad (12)$$

$$j_1(\lambda''_n \beta) = 0,$$

defining hole energies

$$E_h = -\frac{\hbar^2}{2m_h R_{nc}^2} \lambda^2. \quad (13)$$

The lowest hole energies correspond to $\lambda'_0 = 4.286$, $\lambda''_0 = 4.493$, $\lambda'_1 = 6.276$, $\lambda''_1 = 7.725$, $\lambda_0 = 8.892$.

In Ref. [10], the correction to the energy of an electron–hole pair confined in the nanocrystal introduced by Coulomb interaction was calculated taking into account discontinuity of the dielectric constant at the boundary of the nanocrystal. Using values of the dielectric constant of Si $\kappa_{Si} = 12$ and of the effective dielectric constant of the surrounding medium $\kappa_m = 4$, the energy of the ground exciton state was found as

$$E_{ex}^0 = E_g + 34.3 \frac{\hbar^2}{2m_{\parallel} R_{nc}^2} + 18.4 \frac{\hbar^2}{2m_h R_{nc}^2} - 2.3 \frac{e^2}{\kappa_1 R_{nc}}. \quad (14)$$

If we take into account finite height of the energy barriers at the boundary of nanocrystals, we have to formulate correct boundary conditions that is not a trivial task. In order to simplify the problem, we recall that as it was pointed out in [10,11] the states of the lowest hole level in the quantum dot can be approximated by the triply degenerate states of the spherically symmetrical single-band Hamiltonian with an effective hole mass given by

$$m^* = \frac{3m_h m_l}{2m_l + m_h}. \quad (15)$$

Energy of such states is

$$E_h^0 = -\frac{\hbar^2 \pi^2}{2m^* R_{nc}^2}, \quad (16)$$

and normalized wave functions are given by

$$\psi_M^0(r) = \zeta_0^h(r) u_M, \quad \zeta_0^h(r) = \frac{1}{\sqrt{4\pi}} \sqrt{\frac{2}{R_{nc}}} \frac{\sin(\pi r/R_{nc})}{r}. \quad (17)$$

Such approximation overestimates the energy value by 13%. For the energy barrier $U_h = 4.3$ eV [11] using Bastard boundary conditions [8,12] in the first order of the perturbation theory we get following expression for the envelope function at $r \geq R_{nc}$

$$\zeta_0^h(r) = -\frac{1}{\kappa_h} \frac{m_0}{m^*} \frac{\partial \zeta_0^h}{\partial r} \Big|_{r=R_{nc}} e^{-\kappa_h(r-R_{nc})}, \quad (18)$$

where $\kappa_h = \sqrt{2m_0(U_h + E_h^0)}/\hbar$.

2.2. Calculation of Auger excitation probability

We consider the process of an excitation transfer from an electron–hole pair confined in a nanocrystal to strongly localized 4f-electrons of an Er^{3+} ion that is mediated by Coulomb interaction. The energy conservation by recombination of an exciton in the nanocrystal and excitation of an Er^{3+} ion can be provided by local phonons. Thus the Auger excitation is assisted by a multiphonon transition.

In accordance with Eq. (14) and with experimental data [14] the ground energy of an exciton in the nanocrystal is in the range 1.2–1.7 eV for the nanocrystals with $R_{nc} > 2$ nm. Therefore such excitons can effectively transfer energy to Er^{3+} ions being in the second ${}^4\text{I}_{11/2}$ excited state (energy of transition to the ground ${}^4\text{I}_{15/2}$ state $\Delta_{02} = 1.24$ eV) or in the third ${}^4\text{I}_{9/2}$ excited state (transition energy $\Delta_{03} = 1.55$ eV). For excitation of an Er^{3+} ion into the fourth ${}^4\text{F}_{9/2}$ excited state (transition energy $\Delta_{04} = 1.9$ eV) several phonons with a total energy by an order of magnitude exceeding kT at room temperature should be absorbed that strongly decreases the probability of such a process. Nevertheless we will also take into account a possibility of the excitation of this state. It is known from experiment [15] that in contrast to transitions between the ground ${}^4\text{I}_{15/2}$ multiplet of an Er^{3+} ion and the upper excited states, ${}^4\text{I}_{11/2}$ and ${}^4\text{I}_{9/2}$, the Huang–Rhys factor S [13,16] is very small for the transition between the ground state

and the first excited ${}^4\text{I}_{13/2}$ state (transition energy $\Delta_{01} = 0.8$ eV). Therefore phonons can not compensate the energy excess of 0.4 eV.

Then we can write the probability in the following way

$$W_{tr} = \frac{2\pi}{\hbar} \sum_{f'} \overline{|M_{ff',vM}|^2} \sum_N J_T(N) \delta(E_{ex}^0 - \Delta_{ff'} - N\hbar\omega), \quad (19)$$

where $\hbar\omega$ is phonon energy, N is number of emitted phonons, $\Delta_{ff'}$ is energy of transition between the initial state in the ground ${}^4\text{I}_{15/2}$ multiplet of the Er^{3+} ion and the excited state in one of the upper multiplets. $J_T(N)$ is the phonon factor calculated in the model of two shifted parabolic potentials [16] and depends on the Huang–Rhys factor S , the phonon energy $\hbar\omega$ and temperature. Index f' in Eq. (19) numerates all possible final states of the 4f-electron. The bar over the absolute value square of the electron transition matrix element $M_{ff',vM}$ means averaging over all initial states f of the 4f-electrons of the Er^{3+} ion being in the ground state, all three ground states of the confined electron numerated by $v = x, y, z$ and all three ground states of the confined holes numerated by $M = -1, 0, 1$.

General expression for the electron transition matrix element is given by

$$M_{ff',vM} = \frac{4\pi e^2}{V} \int d^3 r_1 \int d^3 \Delta r_2 \psi_{e,v}^*(\vec{r}_1) \psi_{h,M}(\vec{r}_1) \times \sum_{\vec{q}} \frac{1}{\kappa(q, \omega) q^2} e^{i\vec{q}(\vec{r}_1 - \vec{R} - \Delta\vec{r}_2)} \psi_f^*(\Delta\vec{r}_2) \psi_{f'}(\Delta\vec{r}_2), \quad (20)$$

where V is normalization volume, \vec{R} is the position of the center of the Er^{3+} ion. \vec{R} , \vec{r}_1 are counted from the center of the nanocrystal, $\Delta\vec{r}_2$ is the position of the 4f-electron relative to the center of the Er^{3+} ion, and ψ_e , $\psi_{f'}$ are wave functions of the 4f-electron of the ion being in the ground and excited states, respectively. In Eq. (20) the time and spatial dispersion of the dielectric constant $\kappa(q, \omega)$ is taken into account.

Crucial for the evaluation of the electron matrix element are the calculation of the overlap integral between electron and hole Bloch amplitudes with the factor $e^{i\vec{q}\vec{r}_1}$ and the calculation of the integral $\langle f | e^{-i\vec{q}\Delta\vec{r}_2} | f' \rangle$ taking into account restrictions, which these integrals impose on the of the transferred wave vector q . Depending on the position the Er^{3+} ion relative to the nanocrystal different contributions can play the leading role.

3. Results

3.1. Excitation of an Er ion inside a nanocrystal

It occurs that for the Er^{3+} ion situated inside the nanocrystal or at a very small distance away from the nanocrystal, the absolute value of the transferred wave vector is around $k_{0s} = 1.15k_x$ ($1/k_{0s} = 0.075$ nm). So the interaction has a contact character, i.e. it is determined by values of the electron and hole wave functions at position where Er^{3+} ion is situated.

The probability of excitation transfer averaged over the orientation of the Er^{3+} ion and its position inside the nanocrystal is given by the following expression

$$W_{\text{tr}}^{\text{inside}} = \frac{3\pi}{2} \frac{1}{\hbar^2 \omega} \left(\frac{e^2}{\kappa R_{\text{nc}}} \right)^2 Q |\langle u_0 | u^s \rangle|^2 \frac{\gamma_f r_f^4}{R_{\text{nc}}^4} J_{\text{T}}^{\text{all}}(R_{\text{nc}}), \quad (21)$$

where

$$Q = \frac{A_0^2}{3} R_{\text{nc}}^3 \int d^3 R |\xi_0^c(\vec{R})|^2 [R_0^s(R)]^2 \approx 3.31, \quad (22)$$

$|\langle u_0 | u^s \rangle| \approx 0.25$ is the overlap integral between the bottom of the valence band $\Gamma_{2s'}$ and the second conduction band A_2^c with k at the position in the first Brillouin zone where the first conduction band has its minimum [17]. We have neglected small contribution of the D-states to the hole ground state wave function. The factor $\gamma_f r_f^4$ comes from summation over f and averaging over f' of the absolute value square of the matrix element $\frac{1}{3} \langle f | r^2 | f' \rangle$, where $r_f \approx 0.43$ nm is the radius of the 4f-shell of the Er^{3+} ion and the unknown factor γ_f is of the order of 1. $J_{\text{T}}^{\text{all}}(R_{\text{nc}})$ gives contributions to the phonon factor from transitions to all possible excited states of the Er^{3+} ion at given nanocrystal radius R_{nc} . The exact value of the Huang-Rhys factor S is not known for the material considered here. We use value $S = 0.1$, which is in accordance with experimental values from [15] obtained for Er^{3+} ions in fluorozirconate glass. The result of Eq. (23) is illustrated in Fig. 1, where we have used $\gamma_f = 1$, $S = 0.1$ and demonstrated how the result changes if one changes the value of the phonon energy, which we do not know. One should notice that in the experiment the dependence of the excitation transfer on the radius of the nanocrystal will be strongly smoothed comparing to Fig. 1 due to the inhomogeneous distribution of the nanocrystals size.

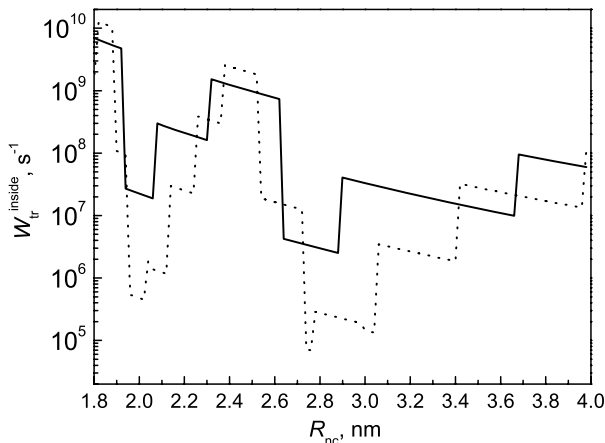


Fig. 1. Dependence of the probability of the excitation transfer to an Er^{3+} ion inside the nanocrystal $W_{\text{tr}}^{\text{inside}}$ on the nanocrystal radius R_{nc} after Eq. (21) for the phonon energy equal to 120 meV (—) and 60 meV (⋯), $S = 0.1$ and room temperature.

3.2. Excitation of an Er ion at the boundary or near the boundary of a nanocrystal

Using electron and wave function at the boundary of a nanocrystal calculated in Section 2.1 in the first order of the perturbation theory we can write the result for the excitation probability in the same way as Eq. (21) but now with the radius dependent factor

$$Q_{\text{surf}}(R_{\text{nc}}) = \frac{1}{2} \left(\frac{4\pi}{3} R_{\text{nc}}^3 \right)^2 |\xi_0^h|^2 \int_0^\pi d\theta \sin \theta |\xi_0^c(\theta)|^2, \quad (23)$$

where $\theta = \arccos(z/R_{\text{nc}})$. At distance d away from the boundary of the nanocrystal, where the interaction occurs due to tunneling, the result should be multiplied with factor $F(d, R_{\text{nc}}) = \exp(-2(\kappa_e + \kappa_h)d)$, which for nanocrystals considered here only very weakly depends on R_{nc} . Dependence of the transfer probability to erbium at the boundary of the nanocrystal on R_{nc} is presented in Fig. 2. In inset to this figure we show dependence of the tunnelling factor $F(d, R_{\text{nc}})$ on the distance from the nanocrystal d for $R_{\text{nc}} = 2$ nm. One can conclude that Auger-excitation due to tunnelling is considerable for $d \lesssim 1$ nm only.

3.3. Excitation of an Er ion at some distance from a nanocrystal

Increasing the distance d between the Er^{3+} ion and the nanocrystal it becomes more difficult for the confined carriers to transfer large momentum to the Er^{3+} ion. So it may become preferable to divide the transferred momentum between f-electrons of erbium and the nanocrystal boundary. We have estimated corresponding contributions, among which there is also a contribution corresponding to the dipole–dipole interaction. It occurs that they give negligibly small values of the transfer probability (less than

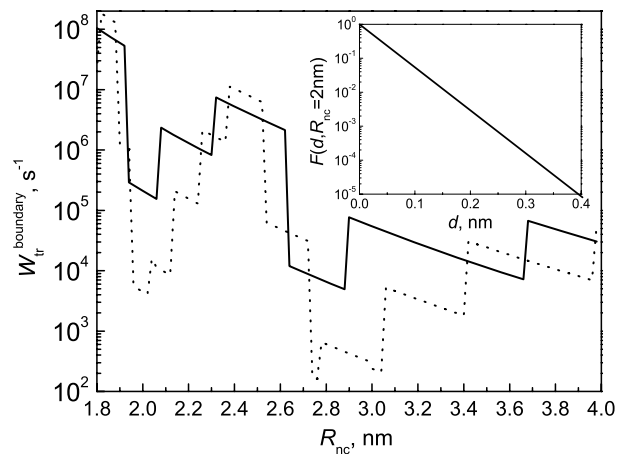


Fig. 2. Dependence of the probability of the excitation transfer to an Er^{3+} ion at the boundary of the nanocrystal $W_{\text{tr}}^{\text{boundary}}$ on the nanocrystal radius R_{nc} for the phonon energy equal to 120 meV (—) and 60 meV (⋯), $S = 0.1$ and room temperature. In the inset dependence of the tunnelling factor $F(d, R_{\text{nc}})$ on the distance d of the Er^{3+} ion from the nanocrystal is shown.

10^2 s^{-1}) so that they should play no role in the transfer process.

4. Discussion and conclusion

For the transfer process to be effective the transfer probability should be larger than the probabilities of other exciton recombination processes. For not too high pumping of the nanocrystals such process is the radiative recombination which can be phonon-assisted or direct. The direct recombination becomes possible because of the breakdown of the momentum conservation rule due to confinement.

The probability of the direct recombination of the exciton being in the ground state in the nanocrystal with radius around 2 nm was found to be of the order 10^2 s^{-1} and it increases to values around 10^4 s^{-1} for nanocrystals having radius 1 nm. The probability of the phonon-assisted recombination for nanocrystals with the radius around 2 nm is of the order 10^4 s^{-1} increasing by an order of magnitude for nanocrystals with the radius of 1 nm [18,19]. These values are comparable to the measured exciton recombination rates [6]. Thus we can conclude that under low pumping conditions the Auger-excitation of erbium is the most effective exciton recombination process if Er^{3+} ion is situated inside nanocrystal or at the boundary of nanocrystal. For high pumping it would be important to estimate also the probability of the Auger recombination of two excitons created in the same nanocrystal.

In conclusion, we have obtained the probability of the Auger excitation of the Er^{3+} ion by an electron–hole pair confined in the nanocrystal being in the nanosecond range if the ion is situated inside the nanocrystal. It is in the microsecond range if the ion is at the boundary or very close the boundary of the nanocrystal. If the distance between the ion and the nanocrystal exceeds 1 nm the Auger excitation transfer is not effective.

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