

Excitation back transfer in a statistical model for upconversion in Er-doped fibres

Sergey Sergeev
s sergeev@wit.ie

Optics Research Group, Waterford Institute of Technology, Cork Road, Waterford, Ireland

Sergei Popov
sergey.sergeev@gmail.com

Department of Microelectronics and Applied Physics, Royal Institute of Technology, Electrum 229, SE-164 40 Kista, Sweden

We report a new analytical method to evaluate the accuracy of a statistical model of the migration assisted upconversion in Er-doped fibres. Unlike the mean-field approach to the excitation back transfer which was used in a previous statistical model, we use a new approximation accounting for the variance of population of the first excited level. Such an approach presents more realistic physical description of the excitation – emission processes in heavily-doped Er-based fibres. Implementing these results, we find that the accuracy of upconversion rate calculations is within 13% if the concentration of erbium ions is smaller than the critical one. [DOI: 10.2971/jeos.2007.07027]

Keywords: erbium, fibre amplifier, upconversion

1 INTRODUCTION

The study of the upconversion process in heavily-doped erbium-based materials is important for the development of efficient fibre optic amplifiers [1]-[8], lasers and sensors [9]-[14] because it effects on the efficiency of such devices. For example, increased upconversion rate leads to the degradation of the performance for high concentration erbium doped fiber/waveguide amplifiers (EDFAs/EDWAs) [1]-[8] and to complex dynamic regimes for lasers at 1.5 μm [9, 10]. However, for upconversion lasers emitting at 0.550 μm and 3 μm [11]-[13], and temperature sensors based on green luminescence [14], increased upconversion rate provides an increased efficiency.

To characterise the performance of high-concentration EDFAs/EDWAs and lasers, models accounting for the upconversion of excitation on homogeneously distributed (homogeneous upconversion, or HUC) and clustered erbium ions (pair-induced quenching, or PIQ) have been exploited [1, 2, 9, 10]. Detailed microscopic study of erbium-doped glasses by means of X-ray absorption fine structure spectroscopy (XAFS) has found no evidence of short-range pair-clustering of Er^{3+} ions [15]. Therefore, more accurate physical models have to be used for fitting experimental results. In our previous publications, we report a model satisfying such criteria [5]-[8]. Unlike HUC and PIQ models, the model takes into account the structure of the glass host matrix by means of pair-correlation function $h(R)$ (the probability density to find two erbium ions at the distance R) and critical distances of upconversion/migration. These distances are directly proportional to the spectra overlap: excited-state absorption - spontaneous emission and ground state absorption - spontaneous emission. To reduce the complex problem of upconversion and migration in an ensemble of uniformly distributed erbium ions, we applied a mean-field approximation to the excitation back transfer in which the variance in the first excited level popula-

tion was neglected [5]-[8]. It gave us an opportunity to derive an analytical expression for the upconversion coefficient as a function of the population of the first excited level and concentration of erbium ions [5, 6, 8].

At high concentration of erbium ions the distance between ions decreases and, therefore, the probabilities of upconversion and migration increase as well. As a result of reinforced migration, distribution of excitation tends to homogeneous for which variance takes the maximum value. Hence, for high concentration of erbium ions the mean-field approach to the excitation back transfer has to be changed for the other approximation accounting for variance in the first excited level population. We report in this paper such approach which gives us opportunity to find the correct analytical expression for upconversion coefficient as a function of the first excited level population and concentration of erbium ions. Additionally, we evaluate the validity of the statistical models of upconversion from [5] in the wide range of erbium doped ion concentrations.

2 MEAN-FIELD APPROXIMATION IN STATISTICAL MODEL OF MIGRATION ASSISTED UPCONVERSION

Dipole-dipole interactions between erbium ions randomly distributed in a host glass leads to two processes called excitation upconversion and migration [5]-[8]. Upconversion occurs between two erbium ions excited at the ${}^4\text{I}_{13/2}$ metastable level and results in the excitation energy transfer from one ion (donor) to the other (acceptor). The donor loses energy and goes to the ground state level ${}^4\text{I}_{15/2}$ whereas the acceptor receives energy and moves to the higher excited state ${}^4\text{I}_{9/2}$ (Figure 1). The acceptor returns back very quickly to the ${}^4\text{I}_{13/2}$

level through step-wise phonon-assisted relaxation processes (Figure 1). Additionally, the upconversion processes are assisted by excitation migration between excited and unexcited ions.

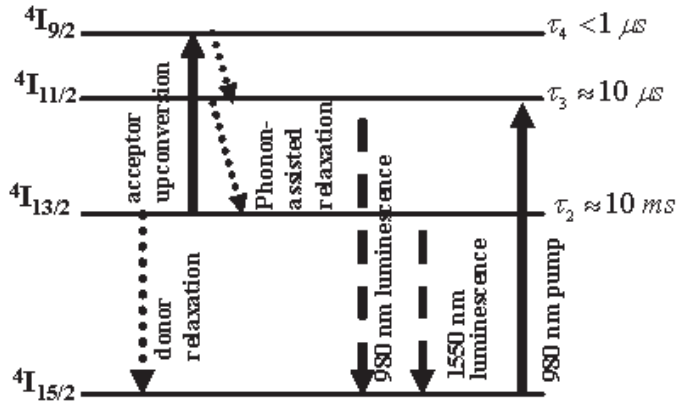


FIG. 1 Erbium ion transition diagram. Upconversion process on metastable ($4I_{13/2}$) level: the donor ion is deactivated whereas the acceptor is excited to $4I_{9/2}$ level. Relaxation of $4I_{9/2}$ level: non-radiative phonon-assisted, and radiative relaxation from the second excited level ($4I_{11/2}$) result in 980 nm fluorescence. Radiative relaxation from the metastable level causes 1550 nm fluorescence.

To describe upconversion and migration processes we use the following set of the rate equations [5, 6]

$$\frac{dn_{2k}}{dt} = (1 - n_{2k}\beta) \frac{I_p}{I_{ps}} - n_{2k} - n_{2k} \sum_{i=1, i \neq k}^N P_{ki} - n_{2k} \sum_{j=1, j \neq k}^N W_{kj} + \sum_{j=1, j \neq k}^N W_{kj} n_{2j}. \quad (1)$$

Here time t is normalised to the lifetime of the first excited level τ_2 ; $\beta = (\sigma_a + \sigma_e) / \sigma_a$ where σ_a and σ_e are the absorption and emission cross-sections, respectively; I_p , I_{ps} are power and saturation power for pump wave; n_{2k} is the probability to find an ion numbered k on the first excited level, N is the number of ions, and n_2 is the population of the first excited level ($n_2 = \lim_{N \rightarrow \infty} (\sum_{k=1}^N n_{2k} / N)$). The rates of upconversion P_{ki} and migration W_{kj} for the dipole-dipole mechanism of excitation energy transfer are given as $P_{ki} = (R_{up} / R_{ki})^6$, $W_{kj} = (R_m / R_{kj})^6$ (R_{up} and R_m are the critical distances for upconversion and migration respectively) [16]. Since $n_3 \sim 0.01n_2$, we neglect the population of the second excited level n_3 in Eq. (1) [5, 6]. As follows from [8], macroscopic equation that is used for experimental study of upconversion processes can be written as follows:

$$\frac{dn_2}{dt} = (1 - n_2\beta) \frac{I_p}{I_{ps}} - n_2 - C_{up} n_2^2. \quad (2)$$

Here C_{up} is the normalised upconversion rate, which can be found from Eq. (1) by averaging over distances between all ions and unexcited ions [5, 6]:

$$C_{up} = \lim_{N \rightarrow \infty} \frac{\sum_{k=1}^N n_{2k} \sum_{i \neq k}^N P_{ki} / N}{\left(\sum_{k=1}^N n_{2k} \right)^2 / N^2}. \quad (3)$$

To find the upconversion rate from Eq. (2) we have to find population n_2 by solving Eq. (1), with further averaging over the distances between erbium ions.

To simplify the problem, we apply the continuous wave excitation ($dn_2/dt = 0$) and mean-field approximation as follows [5, 6]

$$\sum_{j=1, j \neq k}^N W_{kj} n_{2j} \approx n_2 \sum_{j=1, j \neq k}^N W_{kj}. \quad (4)$$

In [6] was found that using the approximation Eq. (4) we lead to the following equation for n_{2k} in integral form

$$n_{2k} = \frac{I_p}{I_{ps}} \int_0^\infty \exp \left[-t \left(1 + \beta \frac{I_p}{I_{ps}} \right) \right] \exp \left(-t \sum_{i=1, i \neq k}^N P_{ki} \right) \times \exp \left(-t \sum_{j=1, j \neq k}^N W_{kj} \right) dt - n_2 \int_0^\infty \exp \left[-t \left(1 + \beta \frac{I_p}{I_{ps}} \right) \right] \times \exp \left(-t \sum_{i=1, i \neq k}^N P_{ki} \right) \frac{\partial}{\partial t} \exp \left(-t \sum_{j=1, j \neq k}^N W_{kj} \right) dt. \quad (5)$$

To find the population n_2 , an averaging over two ensembles should be performed – the first ensemble consists of excited ions and the second one contains both excited and unexcited ions:

$$n_2 = \langle n_{2k} \rangle_{R_{k,1}, \dots, R_{k,n_2N}, R_{k,1}, \dots, R_{k,N}} = \left(\frac{4\pi}{V} \right)^{n_2N+N} \prod_{i=1, i \neq k}^N \int_0^\infty h(R_{k,i}) R_{k,i}^2 dR_{k,i} \times \prod_{j=1, j \neq k}^N \int_0^\infty h(R_{k,j}) R_{k,j}^2 dR_{k,j} \cdot n_{2k}(R_{k,1}, \dots, R_{k,n_2N}, R_{k,1}, \dots, R_{k,N}). \quad (6)$$

Here $h(R)$ is the pair-correlation function to find two erbium ions at the distance R . Using the notation for erbium concentration $c_{Er} = N/V$, we find from Eqs. (5) and (6)

$$n_2 = \frac{I_p}{I_{ps}} \frac{\int_0^\infty \exp \left[-t \left(1 + \beta \frac{I_p}{I_{ps}} \right) \right] \cdot P(t) Q(t) dt}{1 + \int_0^\infty \exp \left[-t \left(1 + \beta \frac{I_p}{I_{ps}} \right) \right] \cdot P(t) \frac{\partial Q(t)}{\partial t} dt}, \quad (7)$$

where

$$Q(t) = \left\langle \exp \left(-t \sum_{i \neq k}^N W_{ki} \right) \right\rangle = \lim_{N \rightarrow \infty} \left(1 - \frac{4\pi c_{Er}}{N} \int_0^\infty h(R) f_m(R) R^2 dR \right)^N \equiv \exp \left(-4\pi c_{Er} \int_0^\infty h(R) f_m(R) R^2 dR \right), \quad (8a)$$

$$P(t) = \left\langle \exp \left(-\sum_{i \neq k}^N P_{ki} \right) \right\rangle = \lim_{N \rightarrow \infty} \left(1 - \frac{4\pi c_{Er} n_2}{N} \int_0^\infty h(R) f_{up}(R) R^2 dR \right)^N \equiv \exp \left(-4\pi c_{Er} n_2 \int_0^\infty h(R) f_{up}(R) R^2 dR \right). \quad (8b)$$

Here $h(R)$ is the pair-correlation function to find the erbium ions at distance R , function $f_{m(up)}(R) = 1 - \exp\left(-t\left(R_{m(up)}/R\right)^6\right)$ is the pair probability density for excitation to leave ion by the migration or upconversion. As follows from Eqs. (7) and (8), the problem of multi-particle interaction with the help of approximation Eq. (4) has been reduced to the pair interactions. On the other hand, for closely located unexcited and excited ions, the probability to find excitation localised on the pair will increase due to higher probability of the excitation migration between ions in pair than probability to leave the pair [16]. As a result, the function $f_m(R)$ will decrease as well. To find the correct form of this function we consider excitation migration between two ions located at distance R [16]. At the initial moment of time $t = 0$, one ion is excited and the other one is unexcited. The rate equation for probability density function for the excitation migration from initially excited ion to an unexcited one at the moment $t > 0$ takes the form [16]:

$$\frac{df_m(R)}{dt} = -f_m\left(\frac{R_m}{R}\right)^6 + (1 - f_m)\left(\frac{R_m}{R}\right)^6, \quad f_m(0) = 0. \quad (9)$$

As a result, we find

$$f_m(R) = \frac{1}{2} \left(1 - \exp\left[-2\left(\frac{R_m}{R}\right)^6 t\right] \right). \quad (10)$$

Applying pair-correlation function $h(R) \equiv 1$ for $R = [0, \infty]$, and substituting Eq. (10) to Eq. (8) we find from Eqs. (7) and (8) [5, 6]

$$Q(t) = \exp(-k_m \sqrt{t}), \quad P(t) = \exp(-k_{up} \sqrt{t}), \quad (11a)$$

$$n_2 = \frac{I_p/I_{ps}}{1 + \beta I_p/I_{ps}} \frac{(n_2 + \sqrt{r/2}) F\left(\frac{k_{up} + k_m}{2\sqrt{1 + \beta I_p/I_{ps}}}\right)}{n_2 + \sqrt{r/2} F\left(\frac{k_{up} + k_m}{2\sqrt{1 + \beta I_p/I_{ps}}}\right)}. \quad (11b)$$

Here $F(u) = 1 - \sqrt{\pi}u \exp(u^2) \operatorname{erfc}(u)$, $k_{up} = \sqrt{\pi}n_2\gamma$, $k_m = \sqrt{\pi r/2}\gamma$, $r = (R_m/R_{up})^6$, $\gamma = c_{Er}/c_0$, c_0 is the critical concentration: $c_0 = \left((4\pi/3)R_{up}^3\right)^{-1}$. The general form of Eqs. (11) accounting for short-range coordination order of erbium ions are given in details in [6].

In spite of the fact that the statistical model demonstrated good applicability for fitting experimental data for gain [7] and upconversion coefficient [8] in high concentration erbium doped fibres, the accuracy of the mean-field approximation Eq.(4) has to be justified to find the margins of parameters where statistical model of upconversion in the form of Eqs. (2) and (11) can provide reliable results.

3 VARIANCE IN EXCITATION BACK TRANSFER IN THE STATISTICAL MODEL OF MIGRATION ASSISTED UPCONVERSION

With the increased concentration of erbium ions, the distance between ions decreases and, therefore, the probabilities

of upconversion and migration increase as well. Migration smoothes out the distribution of excitation to homogeneous one for which the variance takes the maximum value. Hence, for high concentration of erbium ions the mean-field approach to the excitation back transfer has to be changed for an other approximation accounting for the variance in the first excited level population.

We find an appropriate approximation for the statistical model of upconversion accounting for the simplest form of pair-correlation function: $h(R) \equiv 1$ for $R = [0, \infty]$ [5]. This function and Eqs. (11) have been successfully used to fit experimental results for gain as a function of input signal power in [7]. It has been also found that parameters in [7] correspond to the condition when this approach is valid, i.e. $n_2 \leq 0.8$ [6].

We start the derivation of the correct approximation with derivation of the distribution function for stochastic variable $S_0 = \sum_{j=1, j \neq k}^N W_{kj} n_{2j} / n_2$ by using the results of [17]. The distribution function to find one ion in the centre of sphere and the other one at the distance R inside the sphere of the radius R_{max} is $\phi(R) = 3R^2/R_{max}^3$. Distribution function for variable $x_j = W_{kj} n_{2j} / n_2$ is

$$\phi(x_j) = -\frac{R_m^3}{2R_{max}^3} \left(\frac{n_{2j}}{n_2}\right)^{1/2} x_j^{-3/2}. \quad (12)$$

Fourier transform of the function Eq. (12) provides an expression for characteristic function [17]

$$\chi_j(q) = \int_0^\infty \exp(iqx_j) \phi(x_j) dx_j = 1 + i \frac{R_m^3}{R_{max}^3} \sqrt{i\pi q} \frac{n_{2j}}{n_2}. \quad (13)$$

Characteristic function of the sum of independent stochastic variables x_j equals to the product of characteristic functions for each variable, i.e.

$$\begin{aligned} \chi(q) &= \lim_{R_{max} \rightarrow \infty} \prod_{j=1}^N \left(1 + i \frac{R_m^3}{R_{max}^3} \sqrt{i\pi q} \frac{n_{2j}}{n_2} \right) = \\ &= \lim_{N \rightarrow \infty} \left(1 + i \frac{4\pi c_{Er} R_m^3}{3N} \sqrt{i\pi q} \right)^N \left(1 - \frac{\langle \sigma_m^2 \rangle}{8n_2^2} \right) = \\ &= \exp\left(\frac{4}{3} \pi^{3/2} c_{Er} R_m^3 i \sqrt{iq} \left(1 - \frac{\langle \sigma_m^2 \rangle}{8n_2^2} \right)\right), \end{aligned} \quad (14)$$

where $\langle \sigma_m^2 \rangle = \lim_{N \rightarrow \infty} \left(\sum_{k=1}^N (n_{2k} - n_2)^2 / N \right)$, $\langle \sigma_m^2 \rangle$ is the variance of the first excited level population n_2 . Using the Inverse Fourier Transform, we find the distribution function for $S_0 = \sum_{j=1, j \neq k}^N W_{kj} n_{2j} / n_2$:

$$f(S_0) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \exp(-iS_0q) \chi(q) dq = \frac{k_0}{2\sqrt{\pi}S_0^{3/2}} \exp\left(-\frac{k_0^2}{4S_0}\right). \quad (15)$$

Here $k_0 = \sqrt{\pi r/2}\gamma (1 - \langle \sigma_m^2 \rangle / (8n_2^2))$. By means of Eqs. (12)-(15) we can find distribution functions for stochastic variables $S_1 = \sum_{j=1, j \neq k}^N W_{kj}$, $S_2 = \sum_{i=1, i \neq k}^N P_{ki}$. These functions take the form of Eq. (15) with the $k_1 = \sqrt{\pi r/2}\gamma$, $k_2 = \sqrt{\pi}n_2\gamma$

[5, 17]. As follows from definition of variables and Eq. (15), the stochastic variables S_0 and S_1 follow the formula

$$S_0 = S_1 \left(1 - \frac{\langle \sigma_m^2 \rangle}{8n_2^2} \right)^{-2}. \quad (16)$$

Using Eq. (16) we rewrite Eq. (1) for continuous wave excitation (dn_{2k}/dt) as follows:

$$(1 - n_{2k}(S_1, S_2) \beta) \frac{I_p}{I_{ps}} - n_{2k}(S_1, S_2) (1 + S_1 + S_2) - n_2 \left(1 + \frac{\langle \sigma_m^2 \rangle}{4n_2^2} \right) S_1 = 0. \quad (17)$$

By solving the Eq. (17) with respect to $n_{2k}(S_1, S_2)$ and averaging over stochastic variables S_1 and S_2 with distribution functions similar to Eq. (15), we find the following equations for population of the first excited level n_2 and it's variance $\langle \sigma_m^2 \rangle$:

$$n_2 = \frac{(I_p/I_{ps}) (n_2 + \sqrt{r/2}) F(u) \times \left[n_2 - \frac{\sqrt{r/2} \langle \sigma_m^2 \rangle}{(4n_2^2)} \left[1 + \frac{\langle \sigma_m^2 \rangle}{(4n_2^2)} \right] F(u) \right]^{-1}, \quad (18a)$$

$$\langle \sigma_m^2 \rangle = \left(\frac{I_p/I_{ps}}{1 + \beta I_p/I_{ps}} \right)^2 \times \left[F(u) + \frac{\partial F(u)}{\partial u} \frac{u}{2} - \frac{2(I_p/I_{ps}) n_2}{(1 + \beta I_p/I_{ps})} \frac{\sqrt{r/2}}{n_2 + \sqrt{r/2}} \frac{\partial F(u)}{\partial u} \frac{u}{2} + n_2^2 (1 - F(u)) \left\{ \frac{\sqrt{r/2}}{n_2 + \sqrt{r/2}} \frac{\partial F(u)}{\partial u} \frac{\sqrt{\pi} \gamma \sqrt{r/2}}{4\sqrt{1 + \beta I_p/I_{ps}}} + \frac{\sqrt{r/2} (\sqrt{r/2} + n_2/2)}{(n_2 + \sqrt{r/2})^2} \right\} \right]. \quad (18b)$$

Here $u = \sqrt{\pi} \gamma (n_2 + \sqrt{r/2}) / (2\sqrt{1 + \beta I_p/I_{ps}})$.

4 RESULTS AND DISCUSSION

Using Eqs. (2) and (18) we find the upconversion coefficient C_{up} and population fluctuations $\delta = \sqrt{\langle \sigma_m^2 \rangle} / n_2$ as a function of population of the first excited state n_2 and normalised concentration of erbium ions γ . The results of calculations are shown in Figures 2 and 3. Approximation Eq. (4) corresponds to $\langle \sigma_m^2 \rangle = 0$ and results in a simplified model with excitation back transfer which neglects population variance [5].

For low and high population of the first excited state, the distribution of excitation is inhomogeneous with a low value of variance in population (Figure 2). For low population it is caused by small number of excited erbium ions. For high population, the upconversion is static, i.e. there is practically no excitation migration [5]. As a result, upconversion is depleting population more intensely in the regions where erbium ions are more closely located and is creating inhomogeneous distribution of excitation. For intermediate value of population, excitation migration smoothes out the inhomogeneity and, therefore, leads to increased variance in the population of the first excited state (Figure 2).

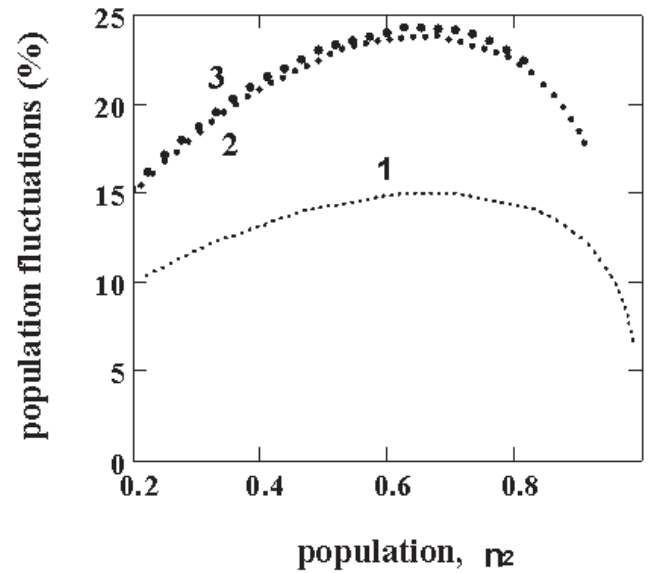


FIG. 2 Population fluctuations δ as a function of population of the first excited state n_2 and normalised concentration of erbium ions γ . Parameters: $r = 60$, $\beta = 1$ (pump at 980 nm [6]), $\gamma = 0.1$ (1), $\gamma = 1$ (2), $\gamma = 2$ (3).

As can be seen in Figure 3, accounting for the population variance leads to decreasing the upconversion coefficient. This effect intensifies with an increase in erbium ions' concentration and has to be taken into account for normalised concentrations $\gamma > 1$. Solvability of erbium ions in phosphate fibre is higher than in silica one and, therefore, only for erbium doped phosphate fibres concentration can exceed critical value without further performance degradation [1, 7].

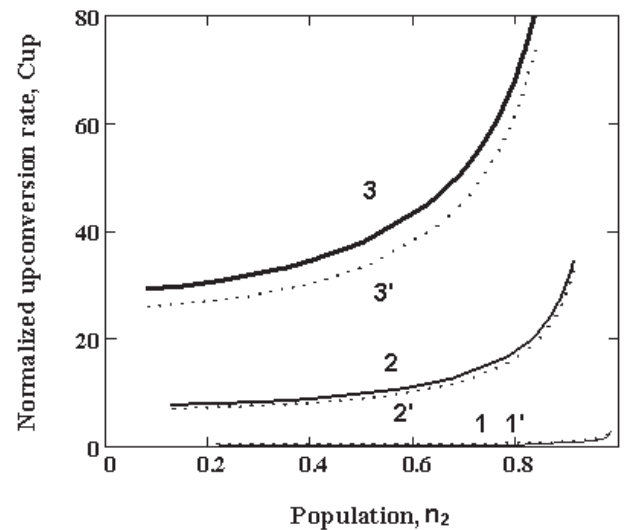


FIG. 3 Normalised upconversion coefficient C_{up} as a function of population of the first excited state n_2 and normalised concentration of erbium ions γ . Parameters: $r = 60$, $\beta = 1$ (pump at 980 nm [6]), $\gamma = 0.1$ (1, 1'), $\gamma = 1$ (2, 2'), $\gamma = 2$ (3, 3'). Simplified model (Eqs. (2) and (11): $\langle \sigma_m^2 \rangle = 0$) [5] (solid lines), generalised model (Eqs. (2) and (18): $\langle \sigma_m^2 \rangle \neq 0$) (dotted lines).

To quantify the precision of the simplified statistical model of

migration-assisted upconversion from [1] we use the variable

$$\epsilon = 2 \frac{[C(\langle \sigma_m^2 \rangle = 0) - C(\langle \sigma_m^2 \rangle \neq 0)]}{[C(\langle \sigma_m^2 \rangle = 0) + C(\langle \sigma_m^2 \rangle \neq 0)]}. \quad (19)$$

The results of calculations of the precision as a function of the first excited level population are shown in Figure 4. With the increased erbium ions concentration, the distance between ions decreases and more than one closely located ion can appear in vicinity of the excited ion. As a result, probability of excitation localisation within this cluster increases and probability of excitation delocalisation decreases. It leads to decreased contribution of migration into the acceleration of upconversion and, therefore, results in decreased value of the upconversion coefficient (Figure 4). For high population $n_2 \sim 1$ contribution of migration into the upconversion processes can be neglected which results in increased precision of the model considered in [5] (Figure 4).

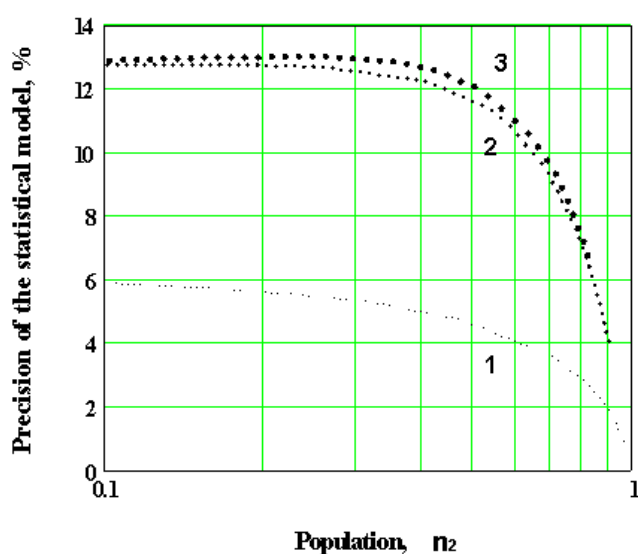


FIG. 4 Precision of the statistical model of upconversion ϵ as a function of population of the first excited state n_2 and normalised concentration of erbium ions γ . Parameters: $r = 60$, $\beta = 1$ (pump at 980 nm [6]), $\gamma = 0.1$ (1), $\gamma = 1$ (2), $\gamma = 2$ (3).

To sum up the theoretical consideration, we emphasise that decreased upconversion rate leads to improving the EDFA/EDWA characteristics and, vice versa, increased upconversion rate results in improved characteristics of the upconversion lasers and green luminescence based sensors [1]-[14]. It has been found, that control of the short-range order of the erbium ions in host matrix can be used to control the upconversion processes [1, 3, 6, 15]. Suppression of the short-range order and, therefore, upconversion processes can be realized by increasing the solubility of erbium in host matrix (co-doping by Al [3] or using phosphate glass [1]) or by modification of deposition process (Direct Nanoparticle Deposition [4]). Otherwise, enhancement of the short-range order of the erbium ions leads to increased upconversion and improved efficiency of the upconversion based devices. As follows from our consideration, for the case of enhanced upconversion, the model accounting for variance of the first excited level population (Eqs. (2) and (18)) will provide a

higher precision for the upconversion characterisation as to compare to the model considered in [5].

In conclusion, we report a new statistical model of migration assisted upconversion in erbium doped fibres. Unlike mean-field approach for the excitation back transfer that was used in the previous statistical model, in the present model we use a new approximation to excitation back transfer accounting for the variance of population of the first excited level. Furthermore, the range of validity of results for the upconversion coefficient, calculated from the simplified statistical model from [5], is evaluated. We find that the maximum deviation is less than 13% for normalised concentration of erbium ions $\gamma \leq 1$.

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