# Numerical and experimental investigation of upconversion in Er doped sol-gel $SiO_2$

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*Abstract*-**The upconversion processes in erbium doped sol-gel $SiO_2$ are studied. The photoluminesce was measured for selected sol-gel samples and compared with the results of the rate equation model. The results show that the presence of cooperative upconversion processes in erbium doped materials can be easily verified experimentally by measuring the photoluminescence spectrum.**

## I. INTRODUCTION

Lanthanides are widely used in photonics for light generation and amplification. Erbium is an element in the lanthanide series that is particularly useful for optical telecoms. This is due to the radiative $^4I_{13/2}$ - $^4I_{15/2}$ transition, the optical wavelength of which happens to coincide with the 3rd optical window of silica optical fibres.

Although, low phonon energy glasses (e.g. fluoride or chalcogenide) are extensively studied because they offer better efficiency of the radiative transitions, there is nonetheless a substantial interest in the development of silica glass based hosts. This is mainly due to the silica glass stability and robustness. Further, lanthanide doped silica glass can be fabricated using a simple, low cost technology e.g. the sol-gel method.

Of particular interest are silica glass materials with large lanthanide dopant concentrations. This is because the large lanthanide concentration allows for the reduction of the dimensions of the fabricated photonic devices. Unfortunately, at large lanthanide dopant concentration the lanthanide ions cluster together. This in turn has an impact on the photoluminescence, which is usually considered deleterious though not always, e.g. thulium doped fibre laser [1].

In this paper we investigate the upconversion processes in sol-gel silica glass samples that are highly doped with erbium. We show that the presence of upconversion can be verified experimentally by measuring the photoluminescence spectrum within the range of wavelengths covered by a low cost Si detector.

## II. PHOTOLUMINESCENCE MODEL

As the pump source used in the experiments is and Ar ion laser operating at the 488 nm we limit the model to 8 levels (Fig.1). This assumption is further supported by the fact that in silica hosts radiative transitions between levels that are lying above $^4S_{3/2}$ level are quenched through phonon assisted transitions. The erbium ion model corresponding to the set of levels given in Figure 1 is described by the following set of 8 equations:

$$\frac{dN_1}{dt} = -\frac{N_1}{\tau_{r1}} - \frac{N_1}{\tau_{nr1}} + \frac{\beta_{21}N_2}{\tau_{r2}} + \frac{N_2}{\tau_{nr2}} + \frac{\beta_{31}N_3}{\tau_{r3}} + \frac{\beta_{41}N_4}{\tau_{r4}} + \frac{\beta_{51}N_5}{\tau_{r5}} + \frac{\beta_{61}N_6}{\tau_{r6}} + \frac{\beta_{71}N_7}{\tau_{r7}}$$

$$\frac{dN_2}{dt} = -\frac{N_2}{\tau_{r2}} - \frac{N_2}{\tau_{nr2}} + \frac{\beta_{32}N_3}{\tau_{r3}} + \frac{N_3}{\tau_{nr3}} + \frac{\beta_{42}N_4}{\tau_{r4}} + \frac{\beta_{52}N_5}{\tau_{r5}} + \frac{\beta_{62}N_6}{\tau_{r6}} + \frac{\beta_{72}N_7}{\tau_{r7}}$$

$$\frac{dN_3}{dt} = -\frac{N_3}{\tau_{r3}} - \frac{N_3}{\tau_{nr3}} + \frac{\beta_{43}N_4}{\tau_{r4}} + \frac{N_4}{\tau_{nr4}} + \frac{\beta_{53}N_5}{\tau_{r5}} + \frac{\beta_{63}N_6}{\tau_{r6}} + \frac{\beta_{73}N_7}{\tau_{r7}}$$

$$\frac{dN_4}{dt} = -\frac{N_4}{\tau_{r4}} - \frac{N_4}{\tau_{nr4}} + \frac{\beta_{54}N_5}{\tau_{r5}} + \frac{N_5}{\tau_{nr5}} + \frac{\beta_{64}N_6}{\tau_{r6}} + \frac{\beta_{74}N_7}{\tau_{r7}}$$

$$\frac{dN_5}{dt} = -\frac{N_5}{\tau_{r5}} - \frac{N_5}{\tau_{nr5}} + \frac{\beta_{65}N_6}{\tau_{r6}} + \frac{N_6}{\tau_{nr6}} + \frac{\beta_{75}N_7}{\tau_{r7}}$$

$$\frac{dN_6}{dt} = -\frac{N_6}{\tau_{r6}} - \frac{N_6}{\tau_{nr6}} + \frac{\beta_{76}N_7}{\tau_{r7}} + \frac{N_7}{\tau_{nr7}}$$

$$\frac{dN_7}{dt} = N_0R_{07} - N_7R_{70} - \frac{N_7}{\tau_{r7}} - \frac{N_7}{\tau_{nr7}} \qquad ------(i)$$

$$N_d = \sum_{i=0}^{7} N_i \qquad ----------(ii)$$

$N_d$ is the total erbium dopant concentration in the host material; Sol-gel $SiO_2$ and by conservation law, equates to the sum of erbium distribution in all the excited states. $\tau_{ri}$ are the radiative lifetimes of an excited state and were obtained from Judd-Ofelt analysis [2]. $R_{07}$ & $R_{70}$ represents the pump rate and rate of stimulated emission of the pump, respectively

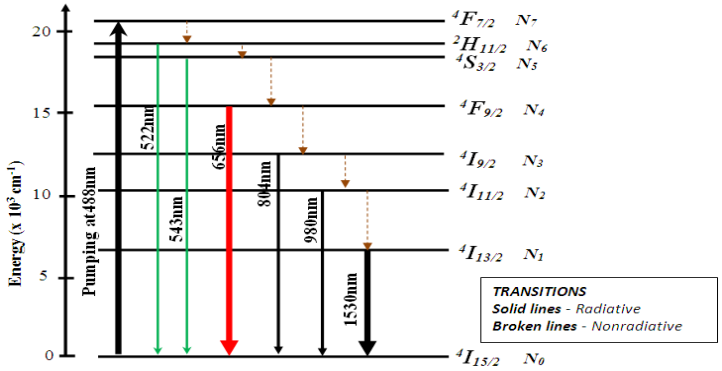


*Fig.1. Atomic level model of erbium*

$\boldsymbol{\beta_{ij}}$ represents the radiative branching ratio for an excited states i to a lower-lying state j as obtained from the Judd-Ofelt model with parameters $\Omega_2$=1.9,$\Omega_4$=0.7,$\Omega_6$ =0.9 ($10^{-20}$cm$^2$) [3]. The rates $\boldsymbol{R_{ij}} = \frac{I\sigma_{ij}}{h\vartheta}$ with I the pump intensity, $\sigma_{ij}$ the cross section and hϑ, the pump photon energy

## III. RESULTS

In this section he results obtained by modeling are compared with experimental ones. The remaining parameters used in modeling are given in Table I.

Table I

| *Parameter* | *Value* |
|---|---|
| Sample ($SiO_2$:$Er_2O_3$), 1 mol% of $Er^{3+}$ ions erbium concentration | $0.52*10^{21}$ cm$^{-3}$ |
| Absorption cross section at 488 nm excitation wavelength | $\sigma_{07}$ =1 x $10^{-21}$ cm$^2$ [4] |
| Pump Intensity | $10^3 - 10^{10}$ W/cm$^2$ |

The measurements were carried out using an Ar ion laser operating at 488 nm as the pump source. The photoluminescence was collected from the sample and spectrally resolved using a Bentham TMC600 spectrograph coupled with an Andor iDus DU440A CCD camera. The emission spectra were corrected for the overall spectral response of spectrograph and CCD detection system. The wavelength calibration was performed using a Ne pen-type calibration lamp.

Figure 2 shows the results obtained from the measured photoluminescence spectrum. Each transition is represented by a line that height of which is proportional to the total integrated (over all Stark split sub-levels) radiative transition rate between the respective atomic levels. The dominant lines correspond to $^4F_{9/2} - {}^4I_{15/2}$ transition (red) and $^4I_{11/2}$ (and $^4I_{13/2}$ – not included in Fig.2) - $^4I_{15/2}$ (infrared) transitions.

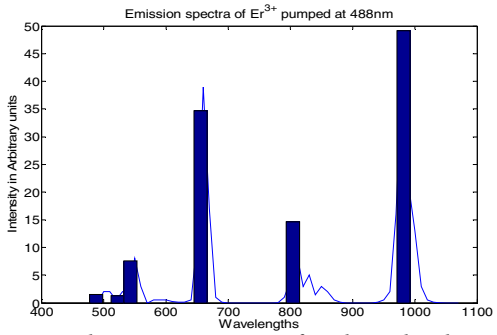


*Fig.2. Measured emission spectra for the sol-gel sample, the bars give the integrated area under each curve corresponding to a multiplet.*

Figure 3 shows the numerical results obtained by solving equations (i) and (ii). Contrary to the experimentally observed results the dominant photoluminescence line corresponds to $^4S_{3/2} - {}^4I_{15/2}$ transition (green). Accumulation of population in $^4S_{3/2}$ due to low multiphonon relaxation gives an intense 543nm green emission.

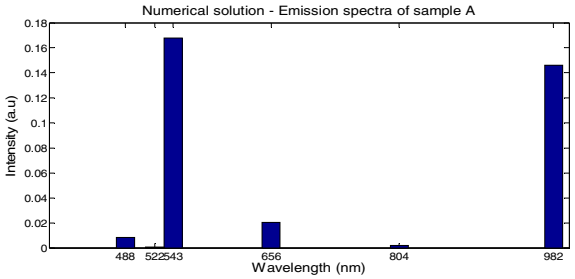


*Fig.3. Calculated emission spectra of the sol-gel sample obtained by solution of equations (i) and (ii).*

The dominance of red upconversion fluorescence from the $^4F_{9/2}$ can be predicted however, by including the upconversion processes (Fig.4). This is done phenomenlogically [5] by modifying the first 5 rate equations in (i):

$$\left(\frac{dN_1}{dt}\right)_{\text{with UP}} = \left(\frac{dN_1}{dt}\right)_{\text{without UP}} - 2C_{up}N_1^2 - W_{CR}N_1N_2$$

$$\left(\frac{dN_2}{dt}\right)_{\text{with UP}} = \left(\frac{dN_2}{dt}\right)_{\text{without UP}} - 2C_{up}N_2^2 - W_{CR}N_1N_2$$

$$\left(\frac{dN_3}{dt}\right)_{\text{with UP}} = \left(\frac{dN_3}{dt}\right)_{\text{without UP}} + C_{up}N_1^2\,, \qquad \left(\frac{dN_4}{dt}\right)_{\text{with UP}} = \left(\frac{dN_4}{dt}\right)_{\text{without UP}} + W_{CR}N_1N_2$$

$$\left(\frac{dN_5}{dt}\right)_{\text{with UP}} = \left(\frac{dN_5}{dt}\right)_{\text{without UP}} + C_{up}N_2^2 \qquad \text{----------(iii)}$$

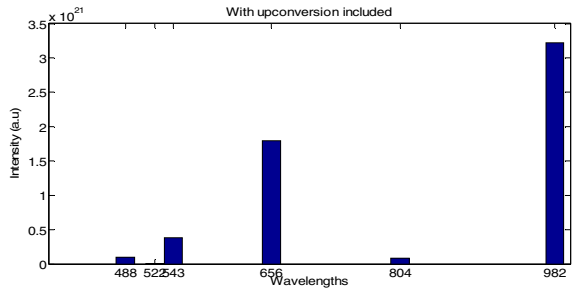


*Fig.4. Calculated emission spectra of the sol-gel sample obtained by solution of rate equations including the upconversion terms.*

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