Design of active devices based on rare-earth-doped glass/glass ceramic: from the material characterization to the device parameter refinement

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ABSTRACT

The accurate knowledge of the rare-earth spectroscopic parameters is fundamental for the design of both fiber and integrated active devices. The lifetimes, the branching ratios, the up-conversion, the cross-relaxation, the energy transfer coefficients of the rare-earths must be preliminarily identified before the design. The particle swarm optimization (PSO) is an efficient global search approach; when applied to rare-earth-doped host materials and devices, it permits the rare-earth spectroscopic characterization starting from optical gain measurements. The model for the peculiar case of a SiO2 - SnO2 : Er3+ glass ceramic system is illustrated. Two different, direct and indirect, pumping schemes are considered for the rare-earth spectroscopic characterization. In the direct pumping scheme, a pump at 378 nm wavelength is used to excite the erbium ions. The SnO2 does not take part in the excitation process. On the contrary, in the indirect pumping scheme the SnO2 is involved by exploiting the absorption band around 307 nm wavelength via a proper pump. In this case, the energy transfer between the SnO2 and the Er3+ ions occurs during the amplification process. The fabricated SiO2 - SnO2 : Er3+ glass ceramic slab waveguide is simulated via a finite element method (FEM) code and a homemade code is used to solve the rate equations. In order to identify the value of the SnO2:Er3+ energy transfer coefficient, the ratio between the two simulated optical gains at 1533 nm wavelength, with the direct and indirect pumping schemes, is compared with the ratio between the two emission intensity measurements.

Keywords: energy transfer, erbium, glass ceramic, rare-earth, tin dioxide.

1. INTRODUCTION

The design of rare-earth-doped fiber and integrated active optical devices generally requires an accurate knowledge of several spectroscopic parameters, whose measurement is often costly and time-consuming. The process is even more complicated when dealing with materials doped with multiple rare-earths, whose energy transfer mechanisms often show a nonlinear behavior. One way to solve this problem is to develop a comprehensive mathematical model of the system, which can be used to recover some of its internal parameters through reverse fitting approaches. As an example, in 1 an erbium-doped microsphere excited via resonant whispering gallery modes (WGMs) was considered. Both the geometrical and spectroscopic parameters were recovered by means of a global search procedure, the particle swarm optimization (PSO) algorithm 2,3,4,5,6,7. A double-step approach was used. In the first step, transmittance measurements performed with the optical pump switched off were fitted through the PSO algorithm and the geometrical parameters of the microsphere were recovered. Conversely, in the second step, optical gain measurements/simulations were performed by exciting the erbium-doped microsphere with an input pump power. The PSO algorithm allowed the recovery of the up-conversion coefficients of erbium with a maximum error of 0.75%. In 8, an erbium-doped chalcogenide glass
An exhaustive mathematical model was developed with the aim of recovering the erbium lifetimes and ion-ion interaction parameters. Simple optical gain measurements were used as input data for the PSO algorithm, which allowed the recovery of the spectroscopic parameters $C_3$, $C_{up}$, $C_{14}$, $\tau_{31}$, $\tau_{11}$ and $\tau_{14}$ with a maximum error of 3.5%.

In this work, a SiO$_2$ - SnO$_2$ : Er$^{3+}$ glass ceramic (GC) system is considered. A mathematical model based on the rate equations is proposed with the aim of investigating the SnO$_2$ - Er$^{3+}$ energy transfer mechanism and recovering its transfer coefficient $K_e$. The system is excited through two different pumping schemes, the direct pumping at 378 nm and the indirect pumping at 307 nm, and emission intensity measurements at 1533 nm are used as input data for the PSO algorithm.

2. THEORY

Glass ceramics are composite materials which contain one or more crystalline phases, with sizes ranging from few nanometers to several micrometers, evenly distributed within a glass phase. They have several interesting properties: (i) chemical durability, (ii) high hardness, (iii) biocompatibility, (iv) low thermal expansion coefficients. Furthermore, different GC structural configurations can be obtained by varying the chemical composition. In this work, the developed model regards a (100-x)SiO$_2$ - xSnO$_2$ : yEr$^{3+}$ GC system, in which SnO$_2$ nanocrystals are embedded into silica, with x = 30 mol% and y = 0.5 mol%. Tin dioxide can act as rare-earth luminescence sensitizer thanks to the high absorption cross section. It also improves the luminescence efficiency, by lowering the multiphonon nonradiative decay rates. The 70SiO$_2$ - 30SnO$_2$ : 0.5Er$^{3+}$ GC system, when suitably excited, enables light emission in the 1520-1560 nm wavelength range, which corresponds to the $^4I_{15/2} \rightarrow ^4I_{15/2}$ transition of erbium ions. Two different pumping schemes, the direct pumping at $\lambda = 378$ nm and the indirect pumping at $\lambda = 307$ nm, are studied. Fig. 1 shows the energy levels diagram for the case of direct pumping. In this pumping scheme, tin dioxide is not involved and the erbium ions are directly excited into the ($^4G_{11/2}$, $^4G_{9/2}$, $^2K_{15/2}$, $^2G_{7/2}$) energy levels. By considering a total of ten erbium energy levels, the following rate equations system is written:

$$\frac{\partial N_{10}}{\partial t} = W_{110}N_{1} - W_{101}N_{10} - \left( A_{109}N_{9} + A_{108}N_{8} + A_{107} + A_{106} + A_{105} + A_{104} + A_{103} + A_{102} + A_{101} \right) N_{10}$$

$$\frac{\partial N_9}{\partial t} = A_{109}N_{10} - \left( A_{98} + A_{97} + A_{96} + A_{95} + A_{94} + A_{93} + A_{92} + A_{91} \right) N_9$$

$$\frac{\partial N_8}{\partial t} = A_{108}N_{10} + A_{98}N_{9} - \left( A_{87} + A_{86} + A_{85} + A_{84} + A_{83} + A_{82} + A_{81} \right) N_8$$

$$\frac{\partial N_7}{\partial t} = A_{107}N_{10} + A_{97}N_{9} + A_{87}N_{8} - \left( A_{76} + A_{75} + A_{74} + A_{73} + A_{72} + A_{71} \right) N_7$$

$$\frac{\partial N_6}{\partial t} = A_{106}N_{10} + A_{96}N_{9} + A_{86}N_{8} + A_{76}N_{7} - \left( A_{65} + A_{64} + A_{63} + A_{62} + A_{61} \right) N_6$$

Figure 1. Energy levels diagram of the SiO$_2$ - SnO$_2$ : Er$^{3+}$ GC system for the case of direct pumping at $\lambda = 378$ nm. Tin dioxide is not excited and its energy levels are neglected.
Figure 2. Energy levels diagram of the SiO₂ - SnO₂ : Er³⁺ GC system for the case of indirect pumping at λ = 307 nm. Tin dioxide is excited and its interaction with the erbium ions is indicated by the grey box.

\[
\frac{\partial N_5}{\partial t} = A_{10,5}N_{10} + A_{9,5}N_9 + A_{8,5}N_8 + A_{7,5}N_7 + A_{6,5}N_6 - (A_{5,4} + A_{5,3} + A_{5,2} + A_{5,1})N_5
\]  

(1f)

\[
\frac{\partial N_4}{\partial t} = A_{10,4}N_{10} + A_{9,4}N_9 + A_{8,4}N_8 + A_{7,4}N_7 + A_{6,4}N_6 + A_{5,4}N_5 - (A_{4,3} + A_{4,2} + A_{4,1})N_4
\]  

(1g)

\[
\frac{\partial N_3}{\partial t} = A_{10,3}N_{10} + A_{9,3}N_9 + A_{8,3}N_8 + A_{7,3}N_7 + A_{6,3}N_6 + A_{5,3}N_5 + A_{4,3}N_4 - (A_{3,2} + A_{3,1})N_3
\]  

(1h)

\[
\frac{\partial N_2}{\partial t} = A_{10,2}N_{10} + A_{9,2}N_9 + A_{8,2}N_8 + A_{7,2}N_7 + A_{6,2}N_6 + A_{5,2}N_5 + A_{4,2}N_4 + A_{3,2}N_3 - A_{2,1}N_2
\]  

(1i)

\[
\frac{\partial N_4}{\partial t} = -W_{1,10}N_1 + (W_{10,1} + A_{10,1})N_{10} + A_{9,1}N_9 + A_{8,1}N_8 + A_{7,1}N_7 + A_{6,1}N_6 + A_{5,1}N_5 + A_{4,1}N_4 + A_{3,1}N_3 + A_{2,1}N_2
\]  

(1j)

The radiative decay rates are given by \( A_{ij} = \beta_{ij}/\tau_i \), where \( \tau_i \) is the lifetime of the \( i \)-th energy level and \( \beta_{ij} \) is the branching ratio of the \( i \rightarrow j \) transition. The transition rates are given by \( W_{ij} = \sigma_{ij}P_{pump}\Gamma_{p}(h\nu_{p}A_{d}) \), where \( h \) is the Planck constant, \( \sigma_{ij} \) is the cross section of the \( i \rightarrow j \) transition, \( P_{pump} \) is the coupled pump power, \( \nu_p \) is the pump frequency, \( \Gamma_p \) is the overlap coefficient between the pump beam profile and the doped region, and \( A_d \) is the area of the GC waveguide. The sum of the populations of the ten erbium energy levels must be equal to the dopant concentration \( N_{Er} \), as follows:

\[
\sum_{k=1}^{10} N_{k} = N_{Er}
\]  

(2)

Fig. 2 shows the energy levels diagram for the case of indirect pumping. In this pumping scheme, tin dioxide is excited and acts as luminescence sensitizer. Electrons are promoted from the valence band (VB) to the conduction band (CB), from which the SnO₂-Er³⁺ energy transfer mechanism occurs. By considering two additional equations for the VB and the CB of tin dioxide, the following rate equations system for the indirect pumping is written:

\[
\frac{\partial N_{12}}{\partial t} = W_{11,12}N_{11} - W_{12,11}N_{12} - A_{12,11}N_{12} - K_{tr}N_1N_{12}
\]  

(3a)

\[
\frac{\partial N_{11}}{\partial t} = -W_{11,12}N_{11} + W_{12,11}N_{12} + A_{12,11}N_{12} + K_{tr}N_1N_{12}
\]  

(3b)

\[
\frac{\partial N_{10}}{\partial t} = -(A_{10,9} + A_{10,8} + A_{10,7} + A_{10,6} + A_{10,5} + A_{10,4} + A_{10,3} + A_{10,2} + A_{10,1})N_{10} + K_{tr}N_1N_{12}
\]  

(3c)

\[
\frac{\partial N_{9}}{\partial t} = A_{10,9}N_{10} - (A_{9,8} + A_{9,7} + A_{9,6} + A_{9,5} + A_{9,4} + A_{9,3} + A_{9,2} + A_{9,1})N_{9}
\]  

(3d)

\[
\frac{\partial N_{8}}{\partial t} = A_{10,8}N_{10} + A_{9,8}N_9 - (A_{8,7} + A_{8,6} + A_{8,5} + A_{8,4} + A_{8,3} + A_{8,2} + A_{8,1})N_{8}
\]  

(3e)

\[
\frac{\partial N_{7}}{\partial t} = A_{10,7}N_{10} + A_{9,7}N_9 + A_{8,7}N_8 - (A_{7,6} + A_{7,5} + A_{7,4} + A_{7,3} + A_{7,2} + A_{7,1})N_{7}
\]  

(3f)

\[
\frac{\partial N_{6}}{\partial t} = A_{10,6}N_{10} + A_{9,6}N_9 + A_{8,6}N_8 + A_{7,6}N_7 - (A_{6,5} + A_{6,4} + A_{6,3} + A_{6,2} + A_{6,1})N_{6}
\]  

(3g)
Table 1. PSO optimization: parameters and variables.

<table>
<thead>
<tr>
<th>Parameter/Variable</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Search space dimension D</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>Number of particles N</td>
<td>30</td>
<td></td>
</tr>
<tr>
<td>Cognitive parameter c₁</td>
<td>1.494</td>
<td></td>
</tr>
<tr>
<td>Social parameter c₂</td>
<td>1.494</td>
<td></td>
</tr>
<tr>
<td>Inertial weight w</td>
<td>0.9-0.4</td>
<td></td>
</tr>
<tr>
<td>McCumber zero-line wavelength λ₀</td>
<td>290-330</td>
<td>nm</td>
</tr>
<tr>
<td>Coupled pump power Pₚₚₚ</td>
<td>0.1-30</td>
<td>mW</td>
</tr>
<tr>
<td>SnO₂-Er³⁺ energy transfer coefficient Kᵣ</td>
<td>10⁻²⁵⁻¹⁰⁻²¹</td>
<td>m²/s</td>
</tr>
</tbody>
</table>

\[
\frac{\partial N_5}{\partial t} = A_{10.0}N_{1.0} + A_{9.5}N_9 + A_{8.5}N_{8} + A_{7.5}N_7 + A_{6.5}N_6 - (A_{5.4} + A_{5.3} + A_{5.2} + A_{5.1})N_5 \quad (3h)
\]
\[
\frac{\partial N_4}{\partial t} = A_{10.4}N_{1.0} + A_{9.4}N_9 + A_{8.4}N_{8} + A_{7.4}N_7 + A_{6.4}N_6 + A_{5.4}N_5 - (A_{4.3} + A_{4.2} + A_{4.1})N_4 \quad (3i)
\]
\[
\frac{\partial N_3}{\partial t} = A_{10.3}N_{1.0} + A_{9.3}N_9 + A_{8.3}N_8 + A_{7.3}N_7 + A_{6.3}N_6 + A_{5.3}N_5 + A_{4.3}N_4 - (A_{3.2} + A_{3.1})N_3 \quad (3j)
\]
\[
\frac{\partial N_2}{\partial t} = A_{10.2}N_{1.0} + A_{9.2}N_9 + A_{8.2}N_8 + A_{7.2}N_7 + A_{6.2}N_6 + A_{5.2}N_5 + A_{4.2}N_4 + A_{3.2}N_3 - A_{2.1}N_2 \quad (3k)
\]
\[
\frac{\partial N_1}{\partial t} = A_{10.1}N_{1.0} + A_{9.1}N_9 + A_{8.1}N_8 + A_{7.1}N_7 + A_{6.1}N_6 + A_{5.1}N_5 + A_{4.1}N_4 + A_{3.1}N_3 + A_{2.1}N_2 - K_{Er}N_1N_{12} \quad (3l)
\]

Like the previous case, the sum of the populations of the two tin dioxide energy levels must be equal to the total concentration $N_{SnO_2}$, as follows:

$$N_{11} + N_{12} = N_{SnO_2} \quad (4)$$

The systems (1a)-(1j) and (3a)-(3l) are solved by assuming steady-state conditions, i.e. all time derivatives vanish. The emission cross section for the CB-VB interband transition of the tin dioxide is calculated via the McCumber relation\(^2\), starting from the experimentally estimated absorption cross section, as follows:

$$\sigma_{e}^{SnO_2}(\lambda) = \sigma_{a}^{SnO_2}(\lambda) \cdot \frac{hc}{\lambda} \cdot \frac{(1 - \lambda)}{k_0} \quad (5)$$

where \(k_0\) is the Boltzmann constant, \(c\) is the speed of light in vacuum, \(\lambda_0\) is the zero-line wavelength and \(T\) is the temperature. The small signal optical gain per unit length for the signal at $\lambda_s = 1533$ nm is given by:

$$g = [\sigma_{e}^{Er}(\lambda_s)N_2 - \sigma_{a}^{Er}(\lambda_s)N_1]G_s \quad (6)$$

where $\sigma_{e}^{Er}$ are the erbium absorption/emission cross sections and $\Gamma_s$ is the signal overlap coefficient.

### 3. PARAMETERS RECOVERY VIA PARTICLE SWARM OPTIMIZATION

A full-vectorial FEM software is employed to perform the electromagnetic analysis of the GC waveguide, whose thickness is $t = 1.2 \ \mu m$. Silicon dioxide and air are used as substrate and coating materials, respectively\(^1\). The signal overlap coefficient is found to be equal to $\Gamma_s = 86.8\%$. The erbium concentration in the 70SiO₂ - 30SnO₂ : 0.5Er³⁺ compound is estimated to be about $N_{Er} \approx 10^{26} \text{ions/m}^3$. The PSO algorithm is employed for the parameters recovering. It is based on the social behavior of animal species like birds\(^1\) and exhibits advantages such as: (i) small number of tuning parameters, (ii) ability to avoid local maxima/minima, (iii) support for multicore processing. Table 1 reports the list of the PSO parameters and variables. The three-dimensional search space is constituted by the McCumber zero-line wavelength $\lambda_0$ of tin dioxide, which is not experimentally known, the coupled pump power $P_{pump}$ and the SnO₂-Er³⁺ energy transfer coefficient $K_r$. A total of $N = 30$ particles is considered.
Figure 3. Evolution of (a) fitness function $F$, (b) McCumber zero-line wavelength $\lambda_0$, (c) coupled pump power $P_{\text{pump}}$ and (d) SnO$_2$-Er$^{3+}$ energy transfer coefficient $K_{\text{tr}}$ as functions of the PSO iteration number for the $p = 2$ particle.

For each particle $p$, with $1 \leq p \leq N$, the optical gain coefficients $g^{\text{DP}}$ and $g^{\text{IP}}$ pertaining to the direct and indirect pumping configurations, respectively, are evaluated by means of Eq. 6. Their ratio is compared with the experimental ratio $R = 7.56$ between the emission intensities, measured by employing a Xenon lamp as excitation source$^{13}$, under the assumption that the two ratios have the same values. Therefore, the fitness function to be minimized is chosen as follows:

$$F = \left( \frac{g^{\text{IP}}}{g^{\text{DP}}} - R \right)^2$$

In other words, minimizing their squared distance allows for a ratio $g^{\text{IP}}/g^{\text{DP}}$ between the simulated optical gains as close as possible to the experimental ratio $R$. 

$$F = \left( \frac{g^{\text{IP}}}{g^{\text{DP}}} - R \right)^2$$
Table 2. PSO optimization: results.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Computation time $T_c$</td>
<td>2</td>
<td>h</td>
</tr>
<tr>
<td>Minimized fitness function value $F_{\text{min}}$</td>
<td>$6.0919 \times 10^{-17}$</td>
<td></td>
</tr>
<tr>
<td>McCumber zero-line wavelength $\lambda_0$</td>
<td>313.2</td>
<td>nm</td>
</tr>
<tr>
<td>Coupled pump power $P_{\text{pump}}$</td>
<td>2.4506</td>
<td>mW</td>
</tr>
<tr>
<td>SnO$<em>2$-Er$^{3+}$ energy transfer coefficient $K</em>{er}$</td>
<td>$8.4068 \times 10^{-22}$</td>
<td>m$^3$/s</td>
</tr>
</tbody>
</table>

After a computation time of about $T_c = 2$ h, the $p = 2$ particle is identified as the best particle yielding the smallest fitness function value. Fig. 3(a)-(d) show the evolution of the fitness function, the McCumber zero-line wavelength, the coupled pump power and the SnO$_2$-Er$^{3+}$ energy transfer coefficient, respectively, as functions of the PSO iteration number for the $p = 2$ particle. After $i = 45$ iterations, the fitness function value is already of the order of $10^{-9}$. About $i = 95$ iterations are enough to obtain a fitness function value less than $10^{-15}$, with negligible improvements given by further iterations. The successful convergence of the optimization procedure is thus confirmed. Table 2 reports the final values of the recovered parameters, with a minimized fitness as low as $F_{\text{min}} = 6.0919 \times 10^{-17}$. The recovered SnO$_2$-Er$^{3+}$ energy transfer coefficient is equal to $K_{er} = 8.4068 \times 10^{-22}$ m$^3$/s. The same mathematical model proposed here for the estimation of the SnO$_2$-Er$^{3+}$ energy transfer coefficient could be also exploited for the future design of GC-based lasers and optical amplifiers. An issue that will need to be addressed is the extremely high pump absorption exhibited by tin dioxide, which makes maintaining the population inversion for long lengths rather difficult.

4. CONCLUSIONS

A SiO$_2$ - SnO$_2$ : Er$^{3+}$ GC system was theoretically modeled in order to investigate the SnO$_2$-Er$^{3+}$ energy transfer mechanism. Direct pumping at 378 nm and indirect pumping at 307 nm were both considered to obtain light emission from erbium ions at 1533 nm. Preliminary photoluminescence measurements performed on a 70SiO$_2$ - 30SnO$_2$ : 0.5Er$^{3+}$ GC sample were employed and their ratio was compared to that calculated between the simulated optical gains per unit length at 1533 nm for both pumping schemes. In order to get a simulated ratio as close as possible to the experimental one, the PSO algorithm with a suitable fitness function was applied, yielding an error as small as $6.0919 \times 10^{-17}$. The recovered SnO$_2$-Er$^{3+}$ energy transfer coefficient was equal to $K_{er} = 8.4068 \times 10^{-22}$ m$^3$/s. The developed mathematical model could find further applications in the design of GC-based lasers and optical amplifiers, in addition to the spectroscopic characterization, even though the issue of extremely high pump absorption exhibited by tin dioxide will need to be addressed. Possible solutions may include optical microresonators, e.g. microrings and microdisks, distributed pump coupling structures and photonic band-gap cavities.

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