Silicon nanocrystals in erbium-doped silica for optical amplifiers

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Abstract : When optically pumped at room temperature, nanometer-sized silicon crystals embedded in erbium-doped silica can be used as efficient sensitisers for Er^{3+} ions, leading to enhanced photoluminescence from erbium at 1540 nm. This enhanced efficiency originates from the higher absorption cross section of Si nanocrystals (Si-nc), in combination with an efficient energy transfer to their neighbouring Er^{3+} ions, leading possibly to higher gains. The feasibility of this novel sensitisation technique can be further investigated by studying the main physical interaction processes in ErSRSO (erbium-doped silicon-rich silicon oxide). From our study, a theoretical model based on Förster-Dexter theory was suggested in order to gain a better understanding of the energy transfer between the activated Si-nc and the Er^{3+} ions. An experimental set-up was established for determining the interaction range between those two species. Their strong coupling was experimentally evidenced by time-resolved photoluminescence measurements.

1. Introduction

Advances in micro-photonics and materials science are driving optical integration even further towards the implementation of WDM (Wavelength Division Multiplexing) in FTTH (Fibre To The Home) systems. Several interacting optical components can now be monolithically placed on a single semiconductor substrate, thus delivering multiple optical functions and leading to more compact and cost-effective devices. The most popular platform for fibre-based functions is now silica-on-silicon technology. Current optical amplifiers based on EDFAs (Erbium Doped Fibre Amplifiers) are expensive and still too bulky and too complex for further integration.

Indirect pumping techniques via sensitisers such as rare-earth codopants (Yb^{3+}, Nd^{3+}) have already been successfully used in EDFAs to achieve higher gains. As a result, with a given concentration of Er^{3+} ions, the minimum required length of fibre for the optical amplification of signals at 1540 nm can be significantly reduced, thereby reducing the size of the device. A novel sensitisation technique of erbium in silica entails replacing those co-dopants by Si nanocrystals. Indeed, it was demonstrated that the quantum yield of the Er^{3+} ions at 1540 nm can be greatly enhanced by implanting Si nanocrystals into the silica host matrix. Such nanostructures (~3 nm in diameter) have already been used as light emitters [1]. They have the ability to emit light in the visible range due to quantum confinement effects. However most importantly, given their high absorption cross-section, they can effectively trap light in the form of confined excitons and subsequently pump the Er^{3+} ions by transferring their excitonic energy. This indirect pumping mechanism was demonstrated to be much more efficient than the direct excitation of Er^{3+} ions and enhances their effective cross section from 10^{-21} cm^2 in Er-doped silica to 10^{-17} to 10^{-18} cm^2 in ErSRSO [2]. Moreover, cheap broad-band light sources have been used to pump Si-nc [3]. In this study, the energy transfers that take place from the Si-nc to Er^{3+} ions within the silica host matrix were modelled using Förster-Dexter theory.

2. Theoretical model

2.1 General description of our model

The system under consideration consists of Si-ncs with a Gaussian distribution in diameters and randomly distributed erbium ions in a host silica matrix. Each Si-nc is approximated by a spherical volume whose core is made of crystalline Si (diamond-like structure as in bulk Si) surrounded by a transition layer of SiO_x (x<1) where the optically active erbium ions are most likely to be located. As optical pumping occurs, incident photons are efficiently absorbed by Si-nc, thereby generating confined excitons via an inter-band transition. At this stage, several radiative and non-radiative processes can occur. Two radiative channels were identified in annealed ErSRSO samples (i.e. samples devoid of defects that can act as traps and/or recombination centres for excitons). The most desired one is the stimulated emission of Er^{3+} ions at 1540 nm (~0.8 eV) corresponding to the relaxation of Er^{3+} from the first excited state $^4I_{13/2}$ to the ground state $^4I_{15/2}$. We assume that this indirect pumping of Er^{3+} ions via Si-nc is achieved by a non radiative energy transfer that can be modelled by a Förster-Dexter mechanism (section 2.1). The second radiative process that can occur is the radiative recombination of excitons within Si-nc, which gives rise to visible photoluminescence from Si-nc at ~1.6 eV. The presence of Er^{3+} ions quenches the 1.6 eV component and enhances the 0.8 eV luminescence. Non radiative decay channels reduce the quantum efficiency of the erbium photoluminescence. Such processes include upconversion (whereby an Er^{3+} in the metastable state is excited to higher energy states by energy transfer from an exciton) and inter Si-nc energy transfers.

2.1 Förster's theory and the determination of the interaction range

Some elements of Förster-Dexter theory have already been used to describe energy transfers that take within CdSe quantum dots, from Yb^{3+} to Er^{3+} ions in Yb:Er codoped silica and in Er^{3+}:Yb^{3+}-doped fluoride phosphate glasses [4], from Ce^{3+} to Tb^{3+} in $Y_3Si_2O_8Cl$ [5], from Mn^{2+} to Nd^{3+} in phosphate glasses [6] and more generally to describe ion-ion interactions [7] and the sensitisation of lanthanide ions by dye molecules and inorganic nanoparticles [8]. A Förster-Dexter process is a single-step nonradiative energy transfer between an activated sensitiser (Si-nc*) and an unexcited activator (Er^{3+}). The nature of this process is an electric multipole-multipole interaction and can be treated as a quantum-mechanical resonant interaction involving the exchange of a virtual photon. Förster [9] was the first to establish the electric dipole-dipole interaction model which was later extended by Dexter [10] to include higher-order multiple interactions and electronic exchange as the separation distance between the sensitiser and the acceptor becomes shorter (less than 10 Angstroms). We applied Förster's model to ErSRSO where the energy transfer rate between Si-nc and Er^{3+} can be characterized by the interaction range R_0 (or Förster radius) from which can also be determined the fraction of excitable erbium ions. Such data is essential for designing more efficient erbium-based optical waveguide amplifiers with Si-nc.

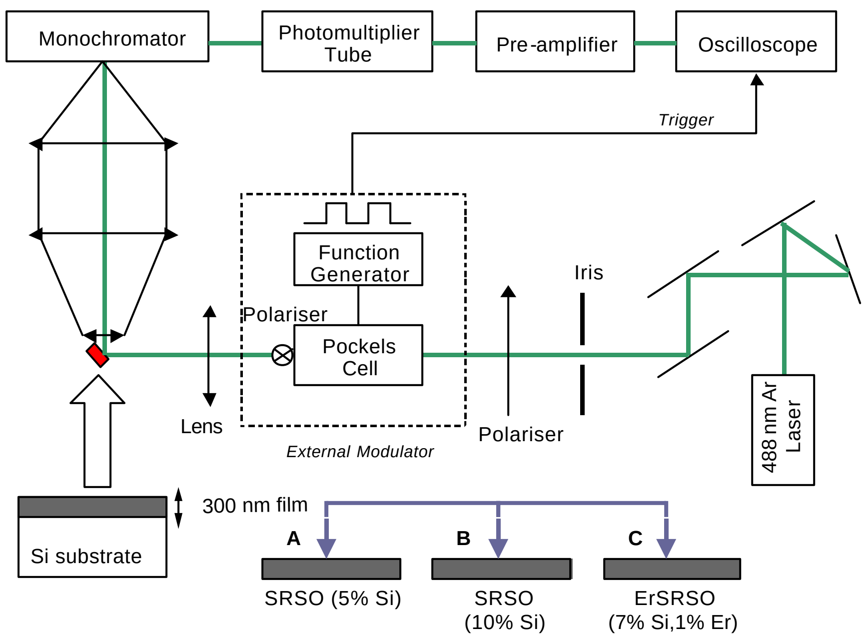
The interaction volume can be experimentally determined by studying the quenching of Si-nc photoluminescence due to the presence of Er^{3+}. To that purpose, the time-resolved photoluminescence originating from Si-nc was recorded in various samples (with and without erbium). After optically pumping the Si-nc for a short period of time, the photoluminescence decay were recorded for both undoped Si-nc (no erbium content, 5 at. % excess Si) U(t) and for doped Si-nc D(t). According to Förster's law, these intensities are related by the following expression [11]:

$$D(t) = U(t)\exp\left[-\gamma\sqrt{\frac{\pi t}{\tau}}\right] \quad (1) \text{ where } \gamma \text{ is directly related to } R_0 \text{ by } \gamma = N_{Er}\frac{4}{3}\pi R_0^3 \quad (2)$$

where N_{Er} is the concentration of erbium in the sample, τ is the effective radiative lifetime of Si-nc in the absence of erbium (in the SRSO sample) and can be obtained experimentally. γ is the only unknown parameter that can be determined by fitting Eq. (1) to the experimental data.

3. Experiment

Room-temperature photoluminescence measurements were carried out on several samples : (A) : 10 at. % excess Si annealed at 1050° C for 8 hours, (B) : 5 at. % excess Si annealed at 1050° C for 8 hours, (C) : 7 at. % excess Si , 1 at. % Er annealed at 1100° C for 8 hours. As shown in *Figure 1*, Si-nc were pumped by an argon-ion laser emitting at 488 nm. This laser beam was externally modulated by a square wave, by means of a Pockels cell driven by a function generator. A scanning single grating monochromator and a photomultiplier tube allowed the radiation emitted from excited silicon nanocrystals at around 1.6 eV to be detected. The emission spectra of these samples were also recorded using standard lock-in techniques.



ErSRSO = Erbium-doped silicon-rich silicon oxide, SRSO = Silicon-rich silicon oxide

Figure1. Experimental set-up used to measure the time-resolved photoluminescence of silicon nanocrystals in Er-doped (C)and undoped samples (A,B).

4. Results and discussion

Strong energy coupling between Si nanocrystals and Er^{3+} ions was evidenced by comparing the photoluminescence response of Si-nc in samples A (undoped) and C (doped) as shown in *Figure 2*. The quenching of the Si-nc PL (photoluminescence) characterized by a faster decay rate was observed in the presence of erbium. Those experimental PL decays were fitted by three types of exponential functions *(Fig. 2)*, namely :

single exponential	*bi-exponential*	*stretched exponential*
$A\exp\left(-\frac{t}{t}\right)$	$A_1\exp\left(-\frac{t}{t_1}\right)+A_2\exp\left(-\frac{t}{t_2}\right)$	$A\exp\left[-\left(\frac{t}{t}\right)^b\right]$

The D(t)/U(t) ratio expressed in Eq. (1) can be experimentally approximated by a stretched exponential accordingly Förster's theory. However, the precision of the fit was lowered by the presence of noise at the end tail of the decay recorded in ErSRSO samples. In such samples, the SNR (Signal to Noise Ratio) was too low, due to the effective quenching action of the erbium ions.

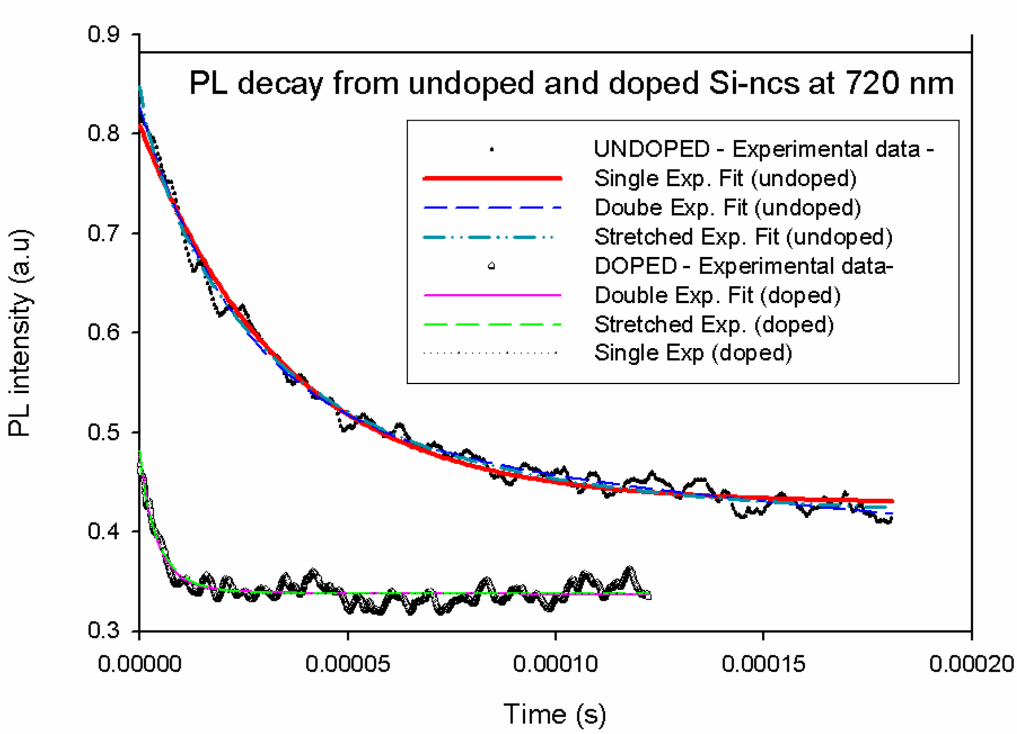


Figure 2. Room-temperature decay curves of photoluminescence from Si-nc measured at 720 nm with (DOPED : 1 at. % Er) and without erbium (UNDOPED). Both experimental curves were fitted with three types of exponentials.

5. Conclusions

To summarize, a strong coupling between Si-nc and Er^{3+} ions was studied and experimentally evidenced in erbium-doped silicon-rich silicon oxide (ErSRSO) thin films at room temperature. In this study, we have developed a theoretical model based on Förster's theory in order to account for the nature of those energy transfers leading to enhanced photoluminescence at 1540 nm from Er^{3+} ions. ErSRSO is a promising gain material that should be further studied for future micro-photonic applications. The theory is now rather well established, but further experimental investigation is still needed to determine the interaction range between Si-nc and Er^{3+} ions. Such knowledge may lead to the design of more efficient optical amplifiers.

6. References

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