

Towards population inversion of electrically pumped Er ions sensitized by Si nanoclusters

O. Jambois^{1,*}, F. Gourbilleau², A. J. Kenyon³,
J. Montserrat⁴, R. Rizk² and B. Garrido¹

¹ MIND-IN2UB, Dept. Electrònica, Universitat de Barcelona, Martí i Fanquès 1, 08028, Barcelona, CAT, Spain

² CIMAP, UMR CNRS 6252, 6 Boulevard Marchal Juin, 14050 CAEN, France

³ Department of Electronic and Electrical Engineering, University College London, Torrington Place, London WC1E 7JE, United Kingdom

⁴ Instituto de Microelectrónica de Barcelona (IMB-CNM, CSIC), Bellaterra 08193, Barcelona, Spain
*ojambois@el.ub.es

Abstract: This study reports the estimation of the inverted Er fraction in a system of Er doped silicon oxide sensitized by Si nanoclusters, made by magnetron sputtering. Electroluminescence was obtained from the sensitized erbium, with a power efficiency of $10^{-2}\%$. By estimating the density of Er ions that are in the first excited state, we find that up to 20% of the total Er concentration is inverted in the best device, which is one order of magnitude higher than that achieved by optical pumping of similar materials.

©2009 Optical Society of America

OCIS codes: (250.5980) Semiconductor optical amplifiers; (160.5690) Rare-earth-doped materials

References and links

1. R. J. Walters, G. I. Bourianoff, and H. A. Atwater, "Field-effect electroluminescence in silicon nanocrystals," *Nat. Mater.* **4**(2), 143–146 (2005).
2. J. Carreras, J. Arbiol, B. Garrido, C. Bonafos, and J. Montserrat, "Direct modulation of electroluminescence from silicon nanocrystals beyond radiative recombination rates," *Appl. Phys. Lett.* **92**(9), 091103 (2008).
3. J. Wurga, R. Li, S. N. Basu, and L. Dal Negro, "Electroluminescence from silicon-rich nitride/silicon superlattice structures," *Appl. Phys. Lett.* **93**(15), 151116 (2008).
4. O. Jambois, J. Carreras, A. Pérez-Rodríguez, B. Garrido, C. Bonafos, S. Schamm, and G. B. Assayag, "Field effect white and tunable electroluminescence from ion beam synthesized Si- and C-rich SiO₂ layers," *Appl. Phys. Lett.* **91**(21), 211105 (2007).
5. F. Iacona, D. Pacifici, A. Irrera, M. Miritello, G. Franzò, F. Priolo, D. Sanfilippo, G. Di Stefano, and P. G. Fallica, "Electroluminescence at 1.54 μm in Er-doped Si nanocluster-based devices," *Appl. Phys. Lett.* **81**(17), 3242–3244 (2002).
6. B. Garrido, C. García, S.-Y. Seo, P. Pellegrino, D. Navarro-Urrios, N. Daldosso, L. Pavesi, F. Gourbilleau, and R. Rizk, "Excitable Er fraction and quenching phenomena in Er-doped SiO₂ layers containing Si nanoclusters," *Phys. Rev. B* **76**(24), 245308 (2007).
7. G. Franzò, E. Pecora, F. Priolo, and F. Iacona, "Role of the Si excess on the excitation of Er doped SiO_x," *Appl. Phys. Lett.* **90**(18), 183102–183104 (2007).
8. A. Irrera, D. Pacifici, M. Miritello, G. Franzò, F. Priolo, F. Iacona, D. Sanfilippo, G. Di Stefano, and P. G. Fallica, "Excitation and de-excitation properties of silicon quantum dots under electrical pumping," *Appl. Phys. Lett.* **81**(10), 1866–1868 (2002).
9. J. Valenta, N. Lalic, and J. Linnros, "Electroluminescence of single silicon nanocrystals," *Appl. Phys. Lett.* **84**(9), 1459–1461 (2004).
10. O. Jambois, Y. Berencen, K. Hijazi, M. Wojdak, A. J. Kenyon, F. Gourbilleau, R. Rizk, and B. Garrido, "Current transport and electroluminescence mechanisms in thin SiO₂ films containing Si nanocluster-sensitized erbium ions," *J. Appl. Phys.* **106**(6), 063526 (2009).
11. P. Horak, W. H. Loh, and A. J. Kenyon, "Modification of the Er³⁺ radiative lifetime from proximity to silicon nanoclusters in silicon-rich silicon oxide," *Opt. Express* **17**(2), 906–911 (2009).
12. M. Wojdak, M. Klik, M. Forcales, O. B. Gusev, T. Gregorkiewicz, D. Pacifici, G. Franzò, F. Priolo, and F. Iacona, "Sensitization of Er luminescence by Si nanoclusters," *Phys. Rev. B* **69**(23), 233315 (2004).
13. A. J. Kenyon, M. Wojdak, I. Ahmad, W. H. Loh, and C. J. Oton, "Generalized rate-equation analysis of excitation exchange between silicon nanoclusters and erbium ions," *Phys. Rev. B* **77**(3), 035318 (2008).
14. K. Hijazi, R. Rizk, J. Cardin, L. Khomenkova, and F. Gourbilleau, "Towards an optimum coupling between Er ions and Si-based sensitizers for integrated active photonics," *J. Appl. Phys.* **106**(2), 024311 (2009).

15. D. Navarro-Urrios, Y. Lebour, O. Jambois, B. Garrido, A. Pitanti, N. Daldosso, L. Pavesi, J. Cardin, K. Hijazi, L. Khomenkova, F. Gourbilleau, and R. Rizk, "Optically active Er[sup 3+] ions in SiO[sub 2] codoped with Si nanoclusters," J. Appl. Phys. **106**(9), 093107 (2009).

1. Introduction

One of the key challenges of Si photonics is the realization of an efficient Si-based light source. Various Si nanocluster (Si-ncl)-based materials using quantum confinement effects in silicon have been reported to address this issue, with the aim of realizing light emitting diodes: Si nanoclusters or nanocrystals inside a dielectric such as SiO₂ [1,2] or Si₃N₄ [3], as well as the integration of carbon [4] or rare earths [5]. Progress is still needed to increase the power efficiency of such systems. Another challenging goal is the realization of a Si-based injection laser. One of the most promising candidates is the system of Er-doped silica sensitized by Si-ncl, principally because the emission of Er ions at 1.55 μm is important for telecom applications as it overlaps the absorption minimum of silica-based fibers. Moreover, the improvement in Er excitation thanks to Si-ncl sensitization is twofold: i) advantage can be taken of the broadband absorption spectrum of the Si-ncl; ii) the effective cross section of the system is increased three or four orders of magnitude (though, as the effective cross section is dependent on the pump photon flux, caution is advised when using this as a figure of merit). Some studies report that a principal limitation of the material is that only a small proportion of Er ions are coupled to Si-ncls [6,7]. Moreover, material studies made by optical pumping show that high fluxes are required to achieve population inversion, leading to fast non-radiative processes such as up-conversion or Auger interactions. However, it has been reported that by pumping the Si-ncl electrically the excitation cross section is increased by two orders of magnitude from that achieved using optical pumping [8,9]. Electrically pumping the Er-doped system sensitized by Si-ncl may thus be a way to achieve population inversion. In this work, we study the fraction of inverted Er in the electrically-pumped Er doped SRSO system.

Active layers of Er-doped SRSO were prepared by magnetron co-sputtering of three confocal cathodes, SiO₂, Er₂O₃ and Si, under a pure Ar plasma. Samples were deposited on a B-doped p-type <100> Si wafer. The active layers were annealed at 900°C for 30 minutes. Electroluminescence was measured using conventional MOS structures fabricated by standard photolithographic processes. The gate electrode of the structure consisted of 200 nm thick semitransparent n-type polycrystalline silicon with an area of $2.56 \times 10^{-4} \text{cm}^2$. Details of structural characterisation can be found in Ref. 10, the results of which are summarized in Table 1. Electrical transport was characterized with an Agilent B1500A semiconductor parameter analyser. EL was detected using an Hamamatsu near infrared photomultiplier (PMT) H10330-75. For EL dynamic studies, a pulse generator was used connected to a digital GHz oscilloscope.

Table 1. Si excess, Er concentration and thickness of the active layer

Material	Si excess (%)	Er concentration (at.cm ⁻³)	Thickness (nm)
C350	8	9×10^{19}	52
C352	18	4.8×10^{20}	45

2. Conduction mechanisms and power efficiency

Current density-electric field characteristics of the two devices are shown in Fig. 1. Applied voltages were small enough to avoid breakdown. The strong dependence of current on applied voltage is characteristic of dielectrics, and we have previously shown that current transport in this kind of layer is by a Poole-Frenkel-type mechanism [10]. As can be seen in the inset of Fig. 1, where the data have been plotted in the Poole-Frenkel representation ($\log(I/V) \propto V^{1/2}$, see Ref. 10 for details), good qualitative agreement is found. Permittivities between 4 and 12 can be extracted from the fit for both samples, which suggests that charge flows from Si-ncl

to Si-ncI through a mechanism that is essentially thermal ionisation, assisted by the strong applied electric field.

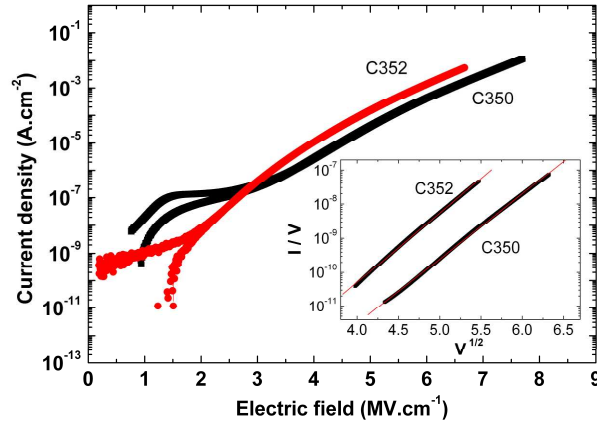


Fig. 1. J-E characteristics of the two layers. Inset shows the I-V characteristics in the Poole Frenkel representation.

Electroluminescence at 1.54 μm was observed for both devices. Figure 2(a) shows the EL spectrum of sample C352 at -30 V, corresponding to a carrier flux of $3.4 \times 10^{16} \text{ q.cm}^{-2}.\text{s}^{-1}$. The observed EL band is characteristic of the $^4\text{I}_{13/2}-^4\text{I}_{15/2}$ transition in the internal 4f-shell of the Er^{3+} ions. For comparison, a photoluminescence (PL) spectrum of the same layer is presented, which shows close correspondence between the bands. Note that the PL was pumped with the 476 nm line of an Ar laser; this wavelength, which is not resonant with any Er absorption band, allows us to verify that the Er ions are excited by the Si-ncI through a sensitizing mechanism.

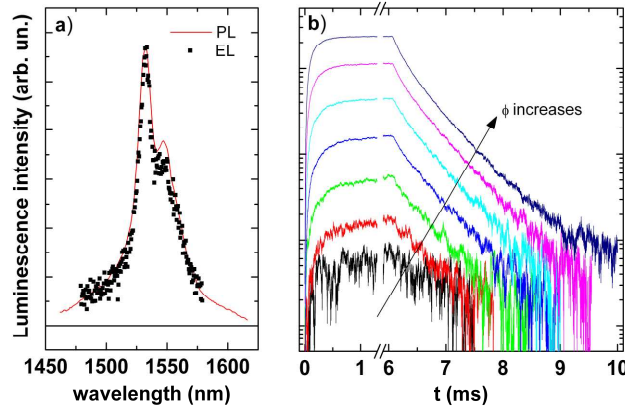


Fig. 2. a) Electroluminescence spectra of layer C352 (squares). A photoluminescence spectrum has been superimposed (red line) for comparison. b) Electroluminescence versus time for increasing flux (as symbolised by the arrow) from $7 \times 10^{13} \text{ cm}^{-2}.\text{s}^{-1}$ to $4 \times 10^{16} \text{ cm}^{-2}.\text{s}^{-1}$.

By precisely calibrating the experimental setup, the power efficiency η_{PE} (defined as the ratio between emitted optical power and electrical power input), has been estimated. Device C352 shows the highest power efficiency: about $1.3 \times 10^{-2}\%$. To our knowledge, this is the highest reported for this kind of device, and is one order of magnitude higher than our previous reported on a different device [10]. Converted into external quantum efficiency η_{EQE} ,

knowing that $\eta_{\text{EQE}} = \eta_{\text{PE}} \times eV/\hbar\omega$, with V the applied voltage and $\hbar\omega$ the energy of the emitted photons, we find an external quantum efficiency of 0.4% - similar to that reported by Iacona et al. using a similar MOS structure [5]. This shows that our calibration of optical power is good.

3. Inverted fraction of Er ions

From the estimation of the optical power emitted by the device, we can estimate the number of Er ions in the first excited state [6]. Let us call this number N_2 .

$$N_2 = \frac{W_{\text{opt}} \times \tau_{\text{rad}}}{\hbar\omega \times S \times d} \quad (1)$$

with τ_{rad} the Er radiative lifetime, S the emission area and d the thickness of the active layer. It is difficult to estimate the radiative lifetime of the Er ions, because it is affected by the presence of the Si-ncl due to the Purcell effect, and will depend strongly on nanocluster size and the Er-to-nanocluster separation [11]. In order to estimate a value, we performed Energy Filtered Transmission Electron Microscopy to characterize nanocluster size distribution for sample C350, and no Si-ncls could be observed. As sensitization of the Er ions is observed, this suggests that the clusters have a diameter smaller than the resolution of this technique which is in our case about 1 nm. Sample C352 has a higher Si excess: we expect thus that the Si-ncl size and/or density are higher, leading to a shorter radiative lifetime. Following Ref. 11, we estimate that τ_{rad} for sample C350 is about 10 ms because it contains very small clusters, whereas that of sample C352 is about 5 ms. By measuring the EL decay time of both samples (more details are presented below), we observe that the lifetime of C352 is shorter, as expected if the radiative lifetime is shorter. Another important value to estimate is the fraction of light that can be extracted from the device. By taking into account both total internal reflection inside the active layer due to the index contrasts and back reflection from the back electrode, we calculate that 12% of the emitted light is able to leave the top electrode. Absorption measurements made on 200 nm thick doped polycrystalline Si show that 53% of the light is transmitted at 1.55 μm . In expression (1), the optical power W_{opt} emitted by the device takes into account those corrections. The electron flux is calculated by dividing the current density J by the electron charge q . By dividing N_2 by the total Er concentration for a given flux, we obtain the percentage of Er in the excited state at steady state.

Figure 3 shows the evolution of the inverted Er fraction with electron flux calculated by this method for both devices. We observe an increase of about four orders of magnitude in N_2 over the range studied. At low flux the population of the first excited state increases linearly with electron flux. At higher fluxes, saturation is observed for both devices. From Fig. 3 we see that, for layer C350, about 20% of the total Er population is inverted, whereas it is about 3% for layer C352. This is, to our knowledge, the first time that the inversion level has been estimated for electrical pumping. Notably, values reached by optical pumping are never higher than few percents [6,12], and high fluxes are necessary to reach them.

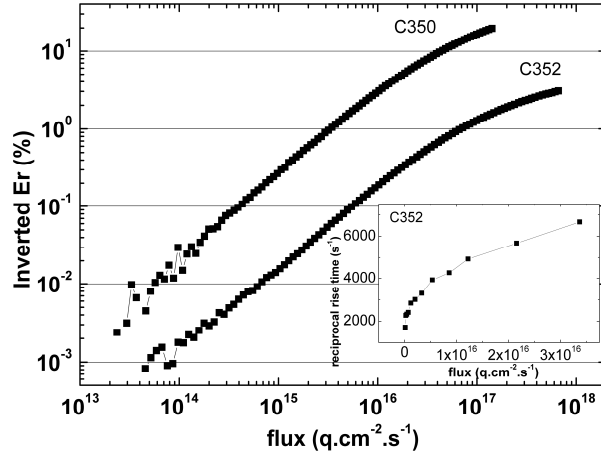


Fig. 3. Estimation of the inverted Er fraction for both layers. Inset shows the non linear behaviour of the reciprocal rise rate with flux.

Figure 2(b) shows the time-resolved EL for C352 with increasing charge flux: EL rise and decay times are observed to be non-exponential (for a stretched exponential fit-to-the-data we find a dispersion value β above 0.8), indicating a distribution of time constants [13]. In the following, we define the rise (decay) time as the time the EL needs to increase (decrease) a factor $1/e$ when the voltage is switched on (off). Decay times of about 870 and 470 μ s are found for device C350 and device C352, respectively - roughly constant with flux. Inset of Fig. 3 shows the evolution of the reciprocal rise time with flux. Assuming a two level system, a linear relation between the reciprocal rise time and the flux should be found: $1/\tau_{\text{rise}} = \sigma\phi + 1/\tau_{\text{decay}}$ with σ the excitation cross section of the luminescent center and ϕ the excitation flux. We observe a sublinear evolution of the reciprocal rise time with flux (see inset of Fig. 3), suggesting that the main mechanism for Er excitation is through Si-ncl, as in the case of optical pumping [13]. This result is supported by the analysis of the conduction mechanism, showing that Si-ncl play a dominant role in charge transport. By means of photoluminescence measurements, we have shown that about 22% of the total Er concentration is coupled to the Si-ncl in layer C350 [14]. This shows that electrical pumping allows the excitation of almost all the coupled Er. Note that in view of the high inversion level we report here, we don't exclude the possibility that a fraction of Er ions - coupled to Si-ncl or not - is directly excited through impact excitation. If we compare now the inversion level of both devices, it seems that high concentrations of Si and Er are prejudicial, although a high Si excess favors charge transport. For layer C352, which contains five times more Er than C350, no improvement of inverted fraction is seen, suggesting either that some of the Er has clustered at this concentration [14], or that cooperative non-radiative processes are more likely to occur.

Further work is ongoing to optimize such thin layers for electrical pumping. In particular, a detailed analysis of the dynamics of the system is underway. Nevertheless, the inversion fractions reported are promising, as they are approaching the critical value of 50% that is required for optical gain. Moreover, the optical activity of the non-coupled Er has also to be measured [15] as, if they are unable to absorb the light signal, they will not have a detrimental effect on the achievement of optical gain. Thus, the values of inversion fraction we report here should be taken as a lower limit.

4. Conclusion

In conclusion, we report a significant development in Si photonics for the realisation of a Si-based optical source by demonstrating an increased fraction of inverted Er ions. We demonstrate the benefits of using electrical pumping to reach high values of inversion. Finally

a power efficiency of $10^{-2}\%$ is reported, corresponding to an external quantum efficiency of 0.4%.

Acknowledgments

This work was partially supported by the European Community through the LANCER Project (FP6-IST 033574). O.J. acknowledges Spanish Ministry of Science for financial support (Juan de la Cierva program) and for sample fabrication in the Integrated Nano and Microelectronics Clean Room facilities at IMB-CNM (ICTS access program).