Gain limiting processes in Er-doped Si nanocrystal waveguides in SiO_2

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Erbium-doped Si nanocrystal based optical waveguides were formed by Er and Si ion implantation into SiO_2. Optical images of the waveguide output facet show a single, well-confined optical mode. Transmission measurements reveal a clear Er related absorption of 2.7 dB/cm at 1.532 μm, corresponding to a cross section of $8\times10^{-20}\,\mathrm{cm}^2$. The Si nanocrystals act as sensitizers for Er but under high doping conditions ($\sim$50 Er ions per nanocrystals) no pump-induced change in the Er related absorption is observed under optical pumping ($\lambda=458$ nm), which is ascribed to an Auger quenching effect. For very high pump powers, a broad absorption feature is observed, attributed to free carrier absorption. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1418417]

The rare earth ion Er^{3+} is widely used for optical amplification. The transition from the first excited state ($^4I_{13/2}$) to the ground state ($^4I_{15/2}$) within the Er $4f$ shell occurs around 1.53 μm, which is one of the standard wavelengths in optical telecommunication. To achieve the population inversion required for optical amplification, rather large pump powers are needed, because of the small absorption cross sections ($\sim10^{-21}\,\mathrm{cm}^2$) of the intra-$4f$ transitions.[1] This problem can be solved by using a sensitizer that more efficiently absorbs pump light and subsequently transfers the absorbed energy to the Er^{3+} ion. As has been shown before,[2–5] silicon nanocrystals can serve as an efficient sensitizer for Er. Their absorption cross section is in the range 10^{-14}–$10^{-16}\,\mathrm{cm}^2$ depending on nanocrystal size and excitation wavelength,[6] and as we have demonstrated,[4] optically generated excitons inside Si nanocrystals can recombine and transfer energy to Er at a rate $>10^6\,\mathrm{s}^{-1}$ and an efficiency of at least 60%. Due to the broad absorption bands of Si nanocrystals, this sensitization scheme would allow for optical pumping of an Er-doped optical amplifier using a broadband light source rather than an expensive pump laser. In addition, electrical pumping of the Er may be possible.

Given these great benefits, it is important to study the gain perspective of this material. We have therefore fabricated Er-doped channel waveguides composed of a Si nanocrystal doped core. We will show that the increased refractive index due to the nanocrystals causes excellent confinement of an optical mode at 1.5 μm centered on the Er and nanocrystal doped region. Thus, this nanocomposite material is not only attractive as a sensitizer system, but also as a waveguide material. All experiments in this article are performed under high doping conditions ($\sim$50 Er ions per Si nanocrystal). Optical transmission measurements are made to study the mode confinement and to derive the absorption cross section at 1.53 μm. Gain measurements performed under intense optical excitation of the Si nanocrystals support our earlier finding[4,5] that there is an upper limit of $\sim$1 Er ion that can be excited by a single nanocrystal.

Silicon ions were implanted at 165 keV to a fluence of $1.7\times10^{17}\,\mathrm{cm}^{-2}$ into a 5 μm thick layer of SiO_2 that was grown by wet thermal oxidation of Si(100). The material was annealed at 1100 °C for 10 min. in flowing Ar in order to induce nucleation and growth of Si nanocrystals. The sample was then implanted with Er at 700 keV to a fluence of $1.2\times10^{16}\,\mathrm{cm}^{-2}$ and annealed at 1000 °C for 10 min. in flowing Ar to remove implantation induced damage. Subsequently it was annealed at 800 °C for 10 min. in forming gas (H_2:N_2 at 1:9) to optimize the nanocrystal and Er photoluminescence (PL) intensity.

Figure 1(a) shows the Er and Si concentration depth profiles, determined using Rutherford backscattering spectrometry (RBS) using a 2 MeV He^+ beam at a scattering angle of 165°. The Si concentration profile peaks at a depth of 240 nm at a peak concentration of 42 at. %, corresponding to an excess Si concentration of 13 at. %. Assuming an average nanocrystal diameter of 3 nm,[7,8] the nanocrystal concentration at the peak of the Si profile is estimated to be $1.3\times10^{19}\,\mathrm{cm}^{-3}$. The Er concentration profile peaks at a depth of 240 nm at a peak concentration of $N_{\mathrm{Er}}=8.3\times10^{20}\,\mathrm{cm}^{-3}$ (i.e. $\sim$50 Er ions per Si nanocrystal).

A series of 3.5 μm wide ridge waveguides was formed in the implanted and annealed SiO_2 film using standard photolithography and Ar beam etching to a depth of 0.5 μm, i.e., well beyond the Si-doped region. The excess Si inside the ridges locally raises the index of refraction, providing the index contrast required for optical mode confinement. To reduce scattering losses, the waveguides were covered with a 1.25 μm thick SiO_2 cladding layer using microwave sputtering. The samples were subsequently cut to a length of 8 mm, and the waveguide input and output facets were mechanically polished. A sketch of the final structure is drawn in Fig. 1(b), where $y=0$ corresponds to the position of the SiO_2 surface before the etching process. The light gray area represents the remaining SiO_2 after etching of the ridge, and the location of the implanted Er and Si is indicated by the dark gray line at $y=0.24\,\mu$m. The smooth solid line at $y\approx1\,\mu$m corresponds to the sample surface after deposition of the SiO_2 cladding layer.

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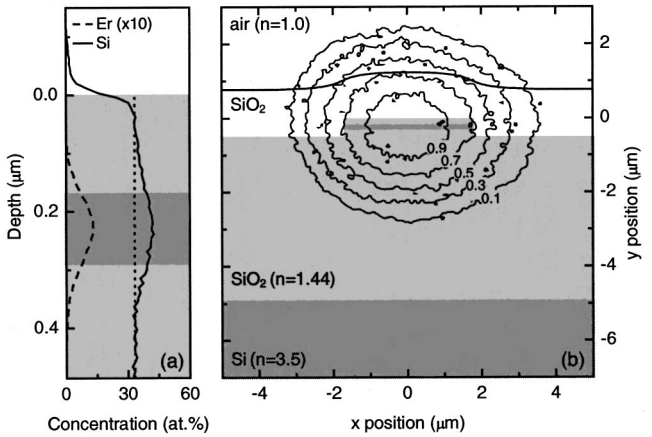

FIG. 1. (a) Er and Si concentration profiles in an ion implanted planar waveguide sample as determined by RBS. (b) Optical mode image at a wavelength of 1.49 μm taken at the output facet of an Er-doped Si nanocrystal based ridge waveguide. The contour lines indicate constant intensity. A sketch of the waveguide structure is included.

Figure 1(b) shows isointensity contours of the optical mode image at 1.49 μm light from an InGaAsP laser guided through the waveguide. The emission from the output facet was projected onto an infrared camera using a microscope objective. At this wavelength, the waveguide only supports the fundamental mode. The mode is slightly elliptical, with a full width at half maximum (FWHM) of 3.8 μm in the x direction, and 3.1 μm in the y direction. In this particular sample structure, with only a small section of the waveguide doped with Er, the Er related absorption and gain will be small, as the effective overlap Γ of the optical mode with the Er profile is only 1%. This low value can be easily increased by changing the size of the nanocrystal doped core by performing multiple Er and Si implants, or by depositing a layer of Er- and Si-doped SiO_2, e.g., using sputter deposition[2] or chemical vapor deposition.[3]

Figure 2(a) shows normalized transmission spectra of Si nanocrystal waveguides containing no Er (dotted line) and 1.3 at. % Er (solid line), measured using a broadband light source butt coupled to the nanocrystal waveguide using a single-mode tapered fiber. In the Er-doped sample, a clear dip is observed around 1.53 μm, due to the $^4I_{15/2} \rightarrow {}^4I_{13/2}$ absorption transition of Er^{3+}. The Er related absorption obtained by dividing the normalized curves in Fig. 2(a) is shown in Fig. 2(b) (solid line). The peak absorption at 1.532 μm is found to be 2.7 dB/cm. From this value and the measured values for Γ and N_{Er}, we obtain an absorption cross section $\sigma_{Er}(1.532\,\mu\text{m}) = 8 \times 10^{-20}\,\text{cm}^2$. This value is rather large compared to values found in Er-doped Si ($2 \times 10^{-20}\,\text{cm}^2$)[8] and SiO_2 ($4 \times 10^{-21}\,\text{cm}^2$).[1] The large cross section may be due to the strong asymmetry of the local dielectric environment of the excitable Er^{3+} ions (note that in a perfectly symmetrical geometry the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition is parity forbidden). This may also explain the relatively short luminescence lifetimes typically observed in this type of material. The high absorption cross section also translates into a high emission cross section,[9,10] which implies that high gain could be achieved in this material over a short length.

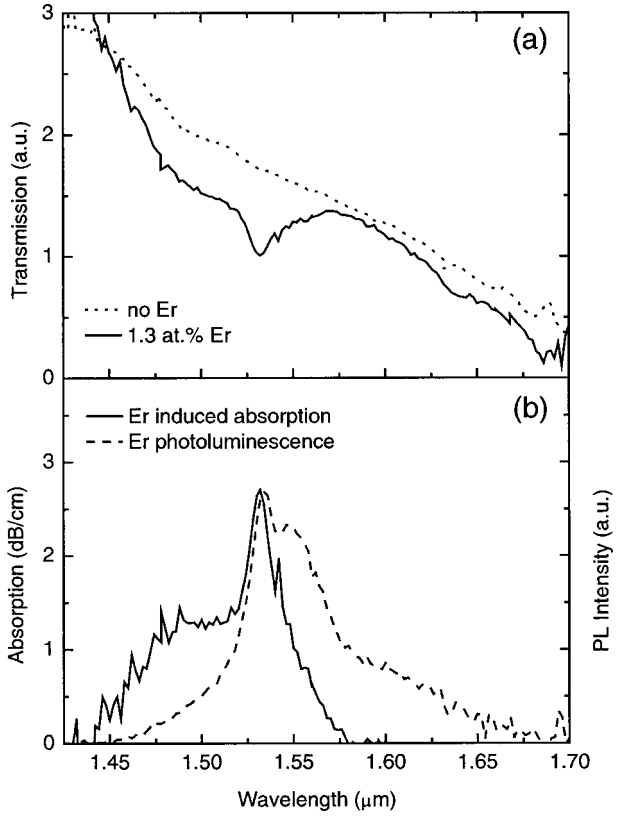

FIG. 2. (a) Normalized transmission spectra of Si nanocrystal based waveguides containing no Er (dotted line) and 1.3 at. % Er (solid line) (b) Er related absorption spectrum (solid line) derived from the data in (a) and Er PL spectrum (dashed line) collected at the waveguide output facet (pump power 180 mW at 458 nm).

Optical pumping was achieved by projecting a 458 nm laser beam onto the top of the waveguide using a cylindrical lens. The elongated spot was aligned with the waveguide, and evenly covered the full waveguide length (see inset in Fig. 3). Figure 2(b) shows the spectrum of guided Er photoluminescence (PL) collected at the output facet. The spectral shape was found to be independent of the applied pump power in the range 25–180 mW. From PL rise and decay time measurements at 1.535 μm (see inset in Fig. 3), it follows that at the lowest applied pump power (25 mW), the Er excitation rate R_{Er} is at least 570 s^{-1}, which would be sufficient to keep 70% of the excitable Er in the first excited state.

Next, optical gain measurements were performed by measuring transmission spectra as a function of pump power. These measurements were done using a chopped white light source as a signal source under continuous pumping at 458 nm. The transmitted signal light was measured using lock-in detection, which prevents the accidental detection of any spontaneous emission induced by the pump. It was found that even at a pump power as high as 250 mW, no change in the Er related absorption could be detected within the measurement accuracy. This puts an upper limit to the amount of Er in the waveguide that is excited through Si nanocrystals. From the experimentally obtained values for σ_{Er} and Γ, and the known noise in our transmission spectra of 1% we conclude that the concentration of excitable Er in the Si nanocrystal-doped region is less than $3 \times 10^{19}\,\text{cm}^{-3}$. Note

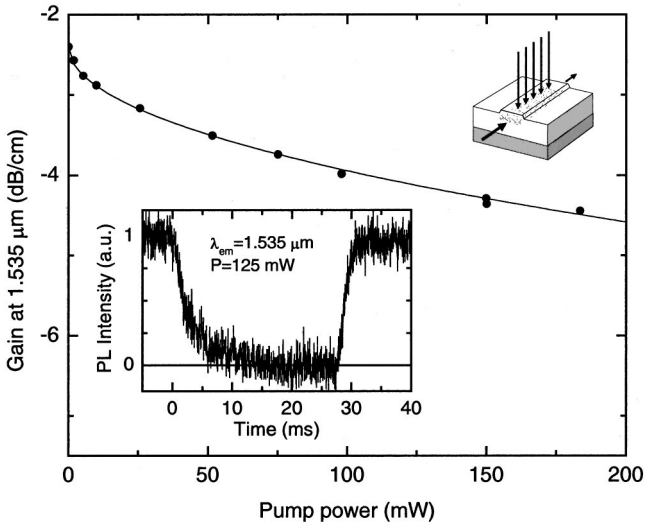

FIG. 3. Gain at 1.535 μm as a function of the applied continuous-wave 458 nm pump power as obtained from transmission measurements. The gain at zero pump power corresponds to the measured Er ground state absorption. A sketch of the measurement configuration is included. The inset shows the time-dependent PL signal at 1.535 μm collected at the output facet during pump modulation.

that the peak Er concentration in the waveguides was $8.2\times10^{20}\,\mathrm{cm}^{-3}$. The fact that the maximum excitable Er concentration is close to the estimated nanocrystal peak concentration of $1.3\times10^{19}\,\mathrm{cm}^{-3}$ in our samples provides strong support for our earlier finding[4,5] that the amount of excitable Er in Si nanocrystal-doped SiO_2 is limited to $\sim$1 Er ion per nanocrystal. Any Er in excess of this limit can not be excited and will introduce an unbleachable absorption at 1.53 μm. This fundamental limit can be explained in terms of an Auger quenching mechanism, in which strong coupling occurs between an excited Er ion and an exciton in a nearby nanocrystal.[5] At high power, when the exciton generation rate exceeds the Er decay rate, this coupling may limit the excited Er fraction to $\sim$1 per nanocrystal.

Although no change in the Er related absorption is observed, the overall transmission of the waveguides is reduced during optical pumping, corresponding to negative gain. Figure 3 shows the gain at 1.535 μm as a function of applied pump power at 458 nm. The gain at zero pump power was taken to be -2.4 dB/cm, corresponding to the measured Er ground state absorption at 1.535 μm [see Fig. 2(b)]. The same pump power dependence of the gain was observed at all wavelengths in the range 1.4–1.7 μm. Apparently, optical pumping at 458 nm induces an absorption in the waveguide material. After pump switch off, the signal transmission converges to its original value over a period of several minutes in a strongly nonsingle exponential fashion. This behavior is unlikely to be a thermal effect, since the sample temperature was found to increase by less than a degree even at the highest applied pump power.

The observed transmission change could be due to free carrier absorption, a well-known effect in bulk silicon, in which free carriers absorb subbandgap radiation due to intraband transitions.[11] The transmission change can not be due to absorption by quantum confined electron–hole pairs, since these recombine at a rate $>10^5\,\mathrm{s}^{-1}$ at room temperature.[12,13] However, if the charge carriers are physically separated, their lifetime can increase by orders of magnitude. Such separation can occur when optical excitation leads to the ejection of a carrier into a deep trap state near the nanocrystal surface. This nanocrystal photoionization process is also thought to be responsible for the observed ''blinking'' of luminescent nanocrystals.[14,15] In this case, the lifetime of the induced absorption is determined by the time it takes the trapped carrier to tunnel back into the nanocrystal, which can indeed be on the order of minutes.[16] We note that in our measurements the pump-induced absorption is a relatively small effect, that occurs at pump powers that are well above those required to invert Si nanocrystal-sensitized erbium.

In conclusion, Er-doped Si nanocrystal based waveguides in SiO_2 show excellent mode confinement at 1.5 μm. The Er absorption and emission cross section is found to be $8\times10^{-20}\,\mathrm{cm}^2$. For high Er concentration ($\sim$50 Er ions per nanocrystal), no change in the Er related absorption is observed under optical pumping at 458 nm. This confirms our previous model that predicts that a single nanocrystal can only excite $\sim$1 Er ion at a time. Under high density optical pumping (much higher than required for inversion) a broadband absorption feature is observed which is attributed to free carrier absorption inside the nanocrystals. We conclude that an Er-doped Si nanocrystal sensitized optical waveguide amplifier pumped using a broadband source may show net gain, provided the Si nanocrystal and Er concentration are properly chosen.

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