Erbium-doped slot waveguides containing size-controlled silicon nanocrystals

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Silicon based slot waveguides with a slot containing Si nanocrystals (Si-nc) and Erbium ions (Er$^{3+}$) inside a silica matrix were prepared using sputter deposition and low-energy ion implantation. This sequence enabled independent optimization of nanocrystal formation and Er$^{3+}$ incorporation parameters. Using a superlattice approach, the size of the Si-nc inside the slot could be controlled and optimized for maximum Er$^{3+}$ luminescence yield at 1.54 $\mu$m. Er$^{3+}$ is found to be efficiently pumped by Si-nc of sizes around 3 to 4 nm. Increasing Er$^{3+}$ photoluminescence at 1.54 $\mu$m with increasing post-implantation annealing temperatures up to 1000°C is attributed to annealing of matrix or Si-nc interface defects mainly. Additionally, a dependence of the Er$^{3+}$ luminescence intensity on both the excitation and emission linear polarization orientation is shown, which demonstrates efficient field enhancement in sputtered slot waveguide structures. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4919420]

I. INTRODUCTION

The ever-growing need for fast and powerful integrated circuits currently leads to a great interest in photonics and optoelectronics. Si-based devices that operate at 1.54 $\mu$m are highly desirable as they are easily integrated into existing manufacturing processes and data transmission schemes. Waveguides (WG) are an essential part of such integrated circuits and are used as connections, resonators, polarization splitters, or signal amplifiers. For Si-based integrated WG, crystalline Si is well suited as a high-refractive index material. A high-index material (n = 3.48) and silica as a low-index material (n = 1.44). The slot WG structure was introduced in 2004 where a thin low-index slot was integrated into the guiding high-index material. Due to the continuity of the electric flux density at their interfaces, the electric field component normal to the structure plane is enhanced in the low-index material.

For telecommunication applications, an active gain medium like erbium ions (Er$^{3+}$), emitting at 1.54 $\mu$m, is required. However, Er$^{3+}$ exhibits a poor absorption cross section and thus must be sensitized in order to be pumped efficiently at low excitation powers. Such sensitizer can be achieved with Si-nc. The combination of slot WG with Si-nc-sensitized Er$^{3+}$ all based on Si-compatible technology has therefore received some attention. However, so far no optical gain could be obtained with this structure, mostly due to absorption losses caused by Si-nc. To be able to optimise these structures, it is important to improve the understanding of the interaction of Si-nc and Er$^{3+}$ in the slot WG structure.

II. SAMPLES AND METHODS

Two sample sets have been prepared. Set 1 consists of a 20 period SiO$_x$/SiO$_2$ SL deposited on a Si wafer using radio frequency magnetron cosputtering of Si and SiO$_2$. Rutherford backscattering spectrometry indicated a relative atomic amount of oxygen in the SiO$_x$ layers of $x = 0.93$. The SiO$_2$ layer thicknesses were varied between 3 and 5 nm. The thickness of the SiO$_2$ layers was kept constant at 5 nm. At elevated temperatures, the SiO$_2$ layers undergo phase separation into SiO$_2$ and Si clusters resulting eventually in Si-nc with sizes dominated by the initial SiO$_2$ layer thickness. Thus, after applying rapid thermal annealing (RTA) at 1050°C for 330 s in a nitrogen atmosphere, three samples of different Si-nc sizes were obtained. High resolution...
transmission electron microscopy (HRTEM) images confirmed the presence of Si-nc of roughly the sizes 3, 4, and 5 nm. Subsequently, Er\(^{3+}\) was implanted in the Si-nc SL stack at an energy of 350 keV at a fluence of about 2 \(\times\) 10\(^{14}\) cm\(^{-2}\). Implantation parameters had been optimized using SRIM simulations\(^{23}\) to result in a broad implantation depth distribution with its peak in the center of the SL and covering most of the Si-nc layers.

Post-implantation annealing was carried out at temperatures between 700 and 1000 °C for 60 min in a conventional furnace under a nitrogen atmosphere. Annealing in N\(_2\) atmosphere has been shown to result in a submonolayer nitrogen coverage of the Si-nc, which reduces on the one hand the polarity at the Si-nc to matrix interface, thereby increasing the Si-nc bandgap, and on the other hand to saturate dangling bonds at the Si-nc interface, thereby improving PL yield.\(^{24,25}\) As both RTP and furnace annealing treatments were performed under N\(_2\) atmosphere, these N-related effects were active for all of our samples. From a comparison of Si-nc PL spectra after RTA and after Er\(^{3+}\) implantation and the corresponding furnace anneal,\(^{26}\) no relative PL peak blueshift could be found for annealing temperatures up to 950 °C. This indicates rather complete N\(_2\) diffusion already during the initial RTA treatment and no further specific N-related changes during the longer furnace anneal.

Sample set 2 consists of slot WG structures as schematically shown in Fig. 1. An 800 nm SiO\(_2\) layer was grown on a Si wafer using wet oxidation. Subsequently, a 100 nm Si layer (Si slab 1) was sputtered on top, as well as a 30 nm slot consisting of a SiO\(_2\)/SiO\(_2\) SL (3 periods) terminated by SiO\(_2\) on either side. Again Si-nc sizes of 3, 4, and 5 nm were prepared. To limit overall slot thickness, the SiO\(_2\) layers were reduced to 3 nm without affecting size control of the resulting Si-nc. The samples were then annealed similarly to set 1 to form three layers of Si-nc. Subsequently, Er\(^{3+}\) was implanted specifically inside the slot using a low implantation energy of 17 keV and a similar fluence as for set 1. Implantation parameters had been determined by SRIM simulations to result in a narrow ion implantation distribution centered around the middle of the slot. After the top Si layer of 100 nm (Si slab 2) was deposited by sputtering, implantation damage was annealed at 900 °C for 60 min in a nitrogen atmosphere. For both the SL and WG set, one Si-nc-free sample was prepared by depositing stoichiometric SiO\(_2\) instead of SiO\(_x\). These reference samples were processed identically to the Si-nc containing samples.

To characterize the emission properties of Er\(^{3+}\) in a Si-nc containing SiO\(_2\) host, PL spectroscopy was used. Most measurements were performed with a HeCd laser emitting at 325 nm focussed to a power density of 0.28 W/cm\(^2\) to prevent Auger effects. Light emitted from recombining charge carriers was dispersed by a monochromator and detected either by a liquid nitrogen cooled CCD camera (visible and near infrared range) or a Ge detector cooled by liquid nitrogen (1.54 μm range). μ-PL measurements were conducted using a Renishaw Invia Reflex μ-Raman spectrometer where a Nd:YAG laser emitting at 532 nm was focussed to a power density of about 20 000 W/cm\(^2\). All PL spectra except those from the Renishaw setup (Fig. 6) were corrected for the spectral response of the detection system. Transmission electron microscopy of the sample cross sections was performed on a Jeol Electron Microscope 2020FS at an acceleration voltage of 200 kV.

### III. RESULTS AND DISCUSSION

#### A. Superlattices

In Fig. 2, the PL spectra of samples of set 1 are presented. For each sample, two signals were recorded: The PL of the Si-nc around 870 nm (taken before Er\(^{3+}\) implantation) and the Er\(^{3+}\) signal around 1.54 μm. From the Si-nc PL spectra (left panel), it is obvious that the smallest Si-nc (3 nm) result in the most intense emission spectrum with the peak maximum at highest energy. This is mainly due to the stronger quantum confinement and thus the enlargement of the bandgap and transition probability in smaller Si-nc. Additionally, an increased density in the same number of SL periods may lead to a higher overall Si-nc density. For the Si-nc-free reference sample, no peak in this spectral range was found, supporting the assignment of this PL peak to the Si-nc.

![Figure 2](image-url)
FIG. 3. Post-implantation annealing on sample SL-3 nm shows increasing Si-nc and Er\(^{3+}\) PL intensity. PL peak area values are scaled to match at the lowest annealing temperature to emphasize relative changes.

The intensity of the Er\(^{3+}\) PL signal (right panel of Fig. 2) correlates clearly with the optical efficiency of the Si-nc. The strongest Er\(^{3+}\) PL was found in the samples containing 3 and 4 nm Si-nc. Judging from this saturation, Si-nc sizes between 3 and 4 nm seem to present an optimum for efficient energy transfer to Er\(^{3+}\). The existence of both a general size-dependence and an optimum Si-nc size for Er\(^{3+}\) luminescence sensitization has been reported before\(^{10,27–29}\), although typically in differently prepared systems. Also for Er-integration via co-sputtering, the Er\(^{3+}\) PL showed a maximum for Si-nc sizes in the range of 3–4 nm.\(^{29}\) For smaller Si-nc, the decreasing Si-nc absorption cross section leads to a decrease in Er\(^{3+}\) PL, whereas for larger Si-nc the weaker quantum confinement shifts the Si-nc energy levels below the upper Er\(^{3+}\) levels, thereby strongly decreasing energy transfer efficiency. On the other hand, the Er\(^{3+}\)-doped Si-nc-free sample (SL-SiO\(_2\)) shows no PL peak around 1.54 μm at all. Although Er\(^{3+}\) in SiO\(_2\) should be optically active as well,\(^{12}\) applied Er fluence and excitation power density are too low for Er\(^{3+}\) to be excited directly due to its low absorption cross section. Indeed, only using the more powerful micro-spot laser of the Renishaw system a very small Er\(^{3+}\) signal was also found for this sample. This confirms the greatly enhanced Er\(^{3+}\) emission by pumping via small Si-nc.

When the material is implanted with ions its crystalline structure is damaged. Er\(^{3+}\) implantation into the SL samples (set 1) lead to a complete loss of the Si-nc PL. Annealing this damage at different temperatures, a rise in both the Si-nc and Er\(^{3+}\) PL could be observed, as is shown for the 3 nm SL sample in Fig. 3. A clear tendency of both signals to increase with increasing temperature can be seen. Predominantly, this is attributable to the annealing of defects in the silica matrix or at the interface between Si-nc and matrix, which increasingly favors radiative transitions both in the Si-nc and the Er\(^{3+}\) ions. Annealing of optically active defects in the silica matrix could be confirmed by PL investigations on the reference sample of Er\(^{3+}\)-implanted pure silica where corresponding PL bands in the 600–800 nm wavelength range gradually vanish up to an annealing temperature of 950°C.

However, while the Si-nc PL increases steadily between 850 and 1000°C the Er\(^{3+}\) PL saturates at 950°C. This might be due to the fact that all Er\(^{3+}\) next to a Si-nc are already activated and saturated. It is to be expected that with a higher Er\(^{3+}\) concentration the PL signal would increase further until Er\(^{3+}\) ions start to cluster or the maximum number of Er\(^{3+}\) which can be sensitized by one Si-nc is reached.

B. Waveguides

The evolution of sample set 2 was followed using TEM and Scanning TEM (STEM) analyses. We will here focus on the sample subset grown with the previously determined optimum SiO\(_2\) thickness of 3 nm. Fig. 4(a) shows the TEM image in Fresnel defocus of a cross section after depositing the bottom Si slab and the slot and after RTA. Due to the temperature treatment, the bottom Si slab crystallized and Si-nc formed out of the SiO\(_2\) layers in the slot. The three dark horizontal lines in the TEM image are an indication of the higher density of the Si-nc layers. HRTEM images revealed that the Si-nc were in a crystalline state, which can be seen from the lattice fringes in Fig. 5. Fig. 4(b) shows a TEM image after Er\(^{3+}\)-implantation into the slot and sputtering of the top Si slab. It is noticeable that the uppermost Si-nc row barely shows scattering contrast anymore. Presumably, the upper Si-nc layer was at least partly destroyed by beam damage. Even after annealing, no reformation of Si-nc in the upper layer could be observed. Looking at a STEM image of the slot, Fig. 4(c), areas of elements with different atomic numbers can be identified. The dark line in the center of the slot indicates the peak concentration of Er\(^{3+}\) overlapping with the center Si-nc layer. This corresponds very well to the implantation depth.

FIG. 4. Brightfield TEM micrographs of the WG sample with 3 nm SiO\(_2\) and 3 nm SiO\(_2\) layer thickness. (a) Taken in defocus mode after deposition of the slot and RTA. Three layers of Si-nc are visible in the slot; the bottom slab is poly-crystalline Si. (b) Taken after Er\(^{3+}\)-implantation and sputtering of the top Si slab. A reduced scattering contrast of the upper part of the slot indicates severe damage to the uppermost Si-nc layer. (c) STEM image of the slot showing a high Er\(^{3+}\)-concentration (dark line) exactly at the center Si-nc layer.
simulated using the SRIM software. In both the center and lower Si-nc layer of the implanted sample, crystalline areas could be identified in HRTEM indicating that there, Si-nc were not completely destroyed by the Er$^{3+}$ implantation. With the Renishaw setup, it was possible to excite and detect luminescence from the capped WG sample from the cleaved edge. This geometry allowed us to evaluate the electric field enhancement of our slot waveguide structure by studying the Er$^{3+}$ luminescence for different linear polarization orientations of both excitation and emission light. Appropriate polarizers and λ/2 plates in both excitation and detection paths were used to either rotate the polarization axis of the exciting laser light or determine the detected Er$^{3+}$ luminescence polarization orientation. As expected from previous works, a preference for Er$^{3+}$ luminescence polarized vertically to the slot plane (y-polarized), i.e., the TM waveguide mode, could be observed, see Fig. 6(a). Interestingly, a similar effect was found when observing the Er$^{3+}$ luminescence and varying the excitation polarization orientation. This is shown in Figs. 6(b) and 6(c) for observing the total Er$^{3+}$ luminescence, i.e., with the polarization optics in the detection path removed. Please note that the same dependence for detecting at a fixed orientation, i.e., tracking either TM or TE mode, has been observed. When the E-field of the incoming laser beam is polarized exactly perpendicular to the slot structure (y-polarized), the most intense Er$^{3+}$ PL peak is observed. With the beam polarized in-plane (x-polarized), the intensity decreases by a factor of 2.5. This is remarkable as waveguiding should be poor due to absorption of this high energy light and the generally low quality of polycrystalline waveguides. We suggest that the penetration depth of the green light into the waveguide structure is still sufficient to cause an enhancement of the vertically polarized electric field component also for this wavelength, which results in more efficient excitation of the Si-nc and thus also the Er$^{3+}$ ions in the slot.

The good signal-to-noise ratio of the PL spectra shown in Fig. 6 also indicates that despite the destruction of the upper Si-nc layer during Er implantation still a rather intense Er$^{3+}$ signal can be detected, even when exciting and detecting from the cleaved edge. From the STEM image in Fig. 4(c), it can furthermore be seen that the main concentration of Er$^{3+}$ is situated around the center Si-nc layer. Therefore, mainly one single layer should be responsible for the observed Er$^{3+}$ PL signal. Optimizing the structure to incorporate only one Si-nc layer while still having sufficient luminescence intensity thus seems feasible. This should be advantageous with respect to reducing carrier absorption losses in the Si-nc that currently seem to limit the performance of Si-nc-based waveguide architectures.

IV. SUMMARY AND CONCLUSION

In conclusion, SL and slot WG structures containing size-controlled Si-nc have been prepared by sputter deposition. Strongest sensitization of implanted Er$^{3+}$ ions has been achieved with Si-nc sizes between 3 and 4 nm. Therefore, Si-nc are an important component in Er-based waveguides and amplifiers. With the SL approach, an effective way of achieving well-defined and controllable Si-nc sizes in the slot waveguide geometry is demonstrated. This in turn provides an efficient tool to enhance the Er$^{3+}$ luminescence. Post-implantation annealing temperatures of 1000 °C have been found to yield the highest Er$^{3+}$ PL intensities in the studied system. Considering the impact of implantation on the WG slot, it is suggested that even a single layer of well-defined Si-nc may provide sufficient sensitization for obtaining optical signals from Er$^{3+}$-doped slot waveguide structures, while at the same time reducing the related carrier absorption losses to a minimum. The significant polarization dependence observed for both emission and excitation light demonstrates efficient field enhancement even in sputtered slot waveguide structures.

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20Some Si-nc did not couple to Er3+ and continued to show luminescence also after implantation.

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