Pump-probe experiments on low loss silica waveguides containing Si nanocrystals

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ABSTRACT

Rib-loaded silica waveguides containing Si nanocrystals were grown by quadruple implantation of Si ions into a 2 μ m-thick thermally-grown SiO₂ layer. The thickness of the resulting flat-profile active region was about 300 nm, with a 9.5% Si excess (determined by X-ray photoelectron spectroscopy). Complete phase separation and nanocrystal formation was assured by annealing at 1100 °C, and studied by means of optical tools such as Raman and luminescence. The rib-loaded structure of the waveguides was fabricated by photolithographic and reactive ion etching processes, with patterned rib widths ranging from 1 to 8 μ m. Efficient light propagation was observed when end-fire coupling a probe signal both at 633 nm and 780 nm into the waveguides, with attenuation losses as low as 11 dB/cm. Signal amplification experiments, with pulsed and continuous wave (CW) top pumping, have shown increased signal absorption when the pump power is raised. This couples with the lack of any fast component in the time decay of the amplified spontaneous emissions as measured by ns pulsed pumping Variable Stripe Length (VSL) experiments. These two phenomena are interpreted as due to the lack of stimulated emission in these nanocrystalline systems.

INTRODUCTION

One of the major open issues in Si-based photonics is the achievement of an efficient and reliable light source, and thus the search for a low-loss optically active media that can be used for achieving optical gain, paving the way to a Si-based laser. Silicon nanocrystal (Si-nc) rich SiO₂ shows good promise of being such a media. Recently, optical gain has been observed in Si nanostructures, a highly significant step towards the realization of a coherent Si-based light source [1, 2]. But in order to take full advantage of all these very promising effects, low-loss waveguides should be fabricated from glass materials containing Si-nc in order to produce edge-emitting structures. Although the control and reduction of optical losses is a primary requirement to achieve net optical gain, so far few loss studies have been reported only on slab waveguides with Si-nc [3-5]. In this work we first present a thorough study on the physical and optical characteristics of rib-loaded waveguides containing Si-nc obtained by ion implantation of Si into silica. An accurate characterisation of the optical losses in the visible range was then performed by surface light scattering experiments and insertion losses measurement at 630 and 780 nm. A

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model is proposed which accounts for the measured 11 dB/cm loss values. Finally, pump & probe measurements were carried out, both in continuous and pulsed pumping configurations.

SAMPLE DESCRIPTION AND CHARACTERISATION

In order to introduce Si excess inside the oxide matrix, a multiple Si⁺ ion implantation was performed in both pure silica and a 2 μm thick SiO₂ film thermally grown on a Si substrate. Ion energies, ranging from 35 to 200 keV, and relative doses of the multiple implantations were chosen to give rise to a planar waveguide with constant Si content to depth of about 0.4 μm . Afterwards the samples were annealed at 1100 °C in N₂ atmosphere for different times, from 1 min up to 16 h.

The Si implanted profile was obtained by XPS analysis. It allowed us to evaluate a constant 9.5% Si excess in the implanted region.

Additional information about the microstructure of the layers has been obtained by means of Raman measurements on the sample prepared on the silica substrate. After annealing at 1100 °C for 20', the Raman spectrum presents a narrow asymmetric peak centred around 521 cm⁻¹, indicating the phase separation and rapid crystallization of the amorphous Si clusters. Further annealing at the same temperature (for durations up to 8 hours) does not modify appreciably neither the shape nor the intensity of the Si-nc signal, implying that full crystallisation has already been reached Additional infrared experiments corroborated that the matrix is stoichiometric SiO₂.

From the analysis of EFTEM images and the building of a statistical histogram of the nanoparticle size, a mean diameter of 4.1 nm is found, together with a rather small dispersion (0.3-0.5 nm typical). By assuming a perfect phase separation between excess Si and SiO₂ and by combining the values of both the Si excess and average particles size, we obtain an estimation of the density of of $\sim 4 \times 10^{18}$ Si-nc/cm³.

Finally, the photoluminescence spectrum showed a strong emission band in the 650-900 nm range, corresponding to excitonic recombination within the nanocrystals.

A more detailed report on these measurements will be published elsewhere [6].

WAVEGUIDE FABRICATION AND LOSSES

In order to determine the best combination of geometrical parameters for the design of silica rib-loaded waveguides for the visible–near infrared range (at 633 and 780 nm) a mode-solver software was used. The objective was achieved by making use of the m-line characterisation of the 1D waveguide, which provides the thickness and the refractive index of the active core (thickness of 340 nm and an index of 1.61 were found). Following the optimised design, a 1 μ m-thick PECVD SiO₂ layer was deposited over a 4" wafer with the 9.5% Si excess. A 0.7 μ m thick rib structure was then formed by using standard photolithography and reactive ion etching. The mask used in the photolithographic process was patterned with 1 cm² squares, each containing eight groups of 2 to 8 μ m-wide waveguides, 240 μ m apart to assure proper mode isolation. The squares were subsequently cut by a dicing saw to a length of 0.95 cm. AFM measurements of the rib widths are shown in Table I.

Efficient propagation of light was observed both at 780 nm and 633nm for all the waveguides. Propagation losses coefficient (α) measurements have been performed by looking at the surface scattered light intensity (fitting the exponential decay of the scattered light) and by insertion loss

measurements, which is a technique based on the measurement of the power transmitted from the waveguide output facet as a function of input signal power. Table I shows the loss coefficient measured for the different rib widths, techniques and wavelengths used. It is worth noting that the two independent techniques give the same (within the error bar) loss coefficients, thus demonstrating the reliability of both experimental procedures

Attenuation losses as low as 11 dB/cm (2.5cm⁻¹) were measured with no significant differences between the two wavelengths used. This is expected since the modal confinement is similar as it was observed in the waveguide simulations. An increment of the losses is obtained in narrow waveguides because the rib sidewall scattering increases and the modal confinement becomes worse.

| Rib width (μm) | | α (dB/cm) as obtained by light scattering | | α (dB/cm) as obtained by insertion losses | |
|----------------|----------------|---|--------------|--|---------|
| nominal | measured | @ 780nm | @ 633nm | @ 780nm | @ 633nm |
| 8 | 8.4±0.05 | 11 ± 1 | 11 ± 0.5 | ~11 | ~13 |
| 6 | 6.6 ± 0.05 | 12 ± 1 | 12 ± 0.5 | ~12 | ~13 |
| 4 | 4.45±0.05 | 12 ± 1 | 12 ± 0.5 | ~15 | ~13 |
| 2 | 2.45 ± 0.05 | 21 ± 1 | 19 ± 1 | ~21 | ~19 |

Table I. Rib widths and propagation losses.

The obtained loss value found in this work is comparable to the best results for planar waveguides measured by the shifting excitation spot (SES) technique, while is an order of magnitude better than the usual reported values [7,8].

Scattering losses due to the composite nature of the core layer have been calculated by Mie theory and result in about 2 dB/cm at 633 nm and 1 dB/cm at 780 nm [9]. Therefore the remaining contribution to the losses, i.e. around 9 dB/cm, is direct absorption of the Si nanocrystals. From this value, and using the already measured nanocrystal density, we can extract an absorption cross section at these wavelengths of around $5x10^{-19}$ cm², that is compatible with results that can be found in literature [10].

GAIN EXPERIMENTS

Internal gain measurements were performed in both CW and time resolved pump and probe setup. A signal beam was end-fire coupled in the rib-waveguide, while the pump beam was focused on a small stripe (approx 20µm wide and 1cm long) overlapping the rib width on the surface of the waveguide. The pumping wavelength in the CW experiment was the 488nm line of an Argon laser. We used two different probe beams, 633nm and 780nm, chopped in order to decorrelate effects due to the pump and allow small signal enhancement measurements through the lock-in acquisition. Within this configuration we define the signal enhancement as the ratio of the probe intensities leaving the sample when the pump is on and off.

In figure 1 it is shown the dependence of the signal enhancement with the pump power in the case of a 633nm probe (the result obtained when coupling 780nm light is similar). A decrease of the transmitted signal in presence of the pump is observed. It is clear that activated absorption channels cause the darkening of the sample. The temporal behaviour is shown in the inset of figure 1 for different pump powers: the signal takes several seconds to recover to the initial value after pump switching off. Recent works [11] reported similar data. The pump induced absorption

has been explained by confined carrier absorption, which is due to the spatial separation of carged carriers.

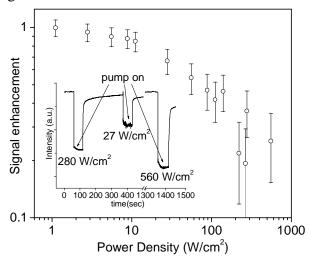


Figure 1. Signal enhancement at 633nm as a function of the 488 nm power density. The inset shows the time dependence of the transmitted signal for different density powers of the pump.

We also performed time resolved experiments using short optical pulses (6ns, 10 Hz repetition rate, 355nm wavelength) produced by a Nd:Yag laser, focusing the spot in a stripe (approx 15 μ m wide and 1cm long). In this configuration the probe is continuous. In figure 2 it is shown the difference between the transmitted probe beam intensity in presence of the pump beam ($I_{probe\ on}$) and the waveguided luminescence ($I_{probe\ off}$): the presence of the probe seems to quench the PL signal (about 9% at 3.2 kW/cm²), the larger quenching is observed at higher powers (about 13% at 6.9 kW/cm²). This behaviour is consistent with the presence of confined carrier absorption: when the probe is present excitons find a new non-radiative recombination channel (probe absorption) and, therefore, the luminescence signal is decreased. If one measures the time resolved and waveguided luminescence, a several μ s-long decay time is observed. No evidence of a ns fast recombination lifetime is measured.

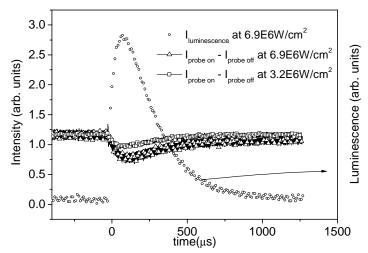


Figure 2. Time resolved intensity of the waveguided luminescence at high pump power (circles). Results of the difference between the signal with probe on and probe off for high power (triangles) and low power (squares) are also displayed.

In order to analyze more carefully the fast emission dynamics, time-resolved experiments were performed on a nanosecond time window in the 90 degree VSL configuration (TR-VSL), pumping in the same configuration as explained before [8]. This technique is used to measure the stimulated emission build-up time as the signal photons are amplified along the waveguide axis.

Emitted radiation was collected through a CCD Hamamatsu streak camera with picosecond time resolution and spectrally analyzed by means of a single grating spectrometer. Figure 3 shows the result for a wavelength window of 700-790nm and a collection time window of 200ns both for long and short stripe length when pumping with an average power of 2mW. Contrarily to other nanocrystalline silicon system where optical gain has been reported [8,12], no evidence of fast decay in the luminescence is observed. As the fast decay of the luminescence was associated with the onset of stimulated emission and gain, it seems that in this nanocrystalline system the balance among stimulated emission and confined carrier absorption is tipped towards confined carrier absorption. This is in agreement with the observation of no signal enhancement in pump and probe experiments.

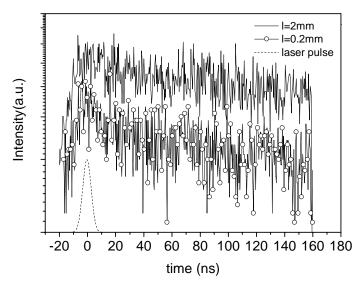


Figure 3 Pumping length dependence of time-resolved variable stripe length decay for a fixed average power of 2mW (corresponding to an instantaneous density power of 6.6 kW/cm²) plotted in a log scale. Two slit lengths have been used.

CONCLUSIONS

Rib-loaded waveguides were fabricated by photolithographic and reactive ion etching processes in Si nanocrystals rich SiO₂ sample. Light propagation in the waveguide was observed and very low losses of around 11 dB/cm at 633 and 780 nm were measured. No signal enhancement but confined carrier absorption was observed in the pump and probe measurements both in CW and in time resolved pumping. This couples with the lack of any fast decay in the time resolved luminescence measurements suggesting that no stimulated emission is present in these samples. This shows, once more, that the observation of optical gain in silicon nanocrystals is strongly dependent on the properties of the actual system used and that careful material optimisation is required to observe the effect.

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