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Stimulated emission in the active planar optical waveguide made of silicon nanocrystals

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Thin layers of silicon nanocrystals prepared by silicon-ion implantation into silica substrate form active planar optical waveguides. Testing experiments of optical gain have been performed on a sample implanted to a dose of 4×10^{17} cm^{-2} by using the variable stripe length (VSL) technique. The photoluminescence collected from the sample facet shows spectrally narrow, polarization-resolved transverse electric (TE) and transverse magnetic (TM) substrate modes. Continuous wave VSL revealed a reduction of optical losses in both modes only. On the contrary, a fast decay component due to amplified spontaneous emission is observed in time-resolved VSL experiments. This fast component shows positive net modal optical gain of ~12 cm^{-1}. The observed superlinear emission as a function of pump intensity, accompanied by shortening of the fast component lifetime, supports further the observation of stimulated emission.

1 Introduction

The first report on experimental observation of positive optical gain in silicon nanocrystals [1] (Si-nc) prepared by ion implantation has stimulated a novel wave of interest in silicon lasing, since a silicon laser would be a key device for the silicon optoelectronics. The presence of stimulated emission was later independently claimed in Si-nc prepared by various techniques such as plasma-enhanced chemical vapour deposition [2, 3], reactive Si deposition [4], magnetron sputtering [5], porous silicon in the sol-gel SiO$_2$ matrix [6] and colloidal Si-nc [7].

Crucial to gain observation is the formation of active planar waveguides where the active core layer is formed by Si-nc embedded in a SiO$_2$ matrix. However, this system shows under certain conditions unusual photoluminescence (PL) properties [8, 9], namely, polarization resolved narrow modes. Similar observations have been reported for luminescent thin films on transparent substrates which form asymmetric planar waveguides [10–12]. An explanation of this effect has been suggested in the form of Si-nc emission at an angle very near to the critical angle for total internal reflection at the core/substrate inter-

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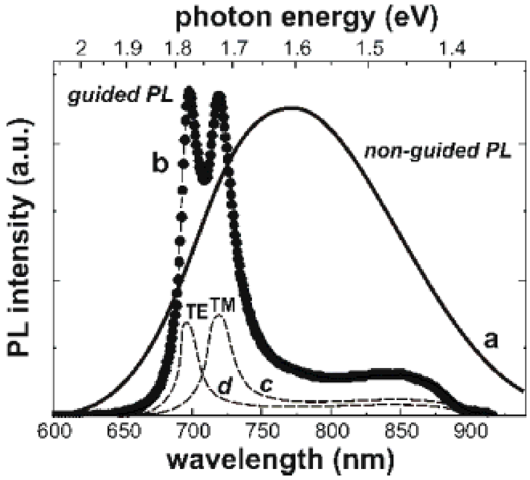

Fig. 1 Photoluminescence spectra of the sample excited by the 325 nm line of a He-Cd laser (I_{exc} = 0.26 W/cm^2). Curves *a* and *b* were detected in directions perpendicular (non-guided, common PL geometry) and parallel (guided, VSL/SES geometry) to the waveguide, respectively. Dashed lines (*c* and *d*) are the polarization resolved TM and TE modes.

face. Such an emission propagates in the substrate like a leaky mode and is localized near the core-substrate boundary [12, 13]. This phenomenon and its possible impact on stimulated emission has not been yet fully understood and further experimental and theoretical study are necessary.

Here we present a variable stripe length (VSL) [1–6, 8, 9] study of an active planar waveguide made of Si-nc in the SiO_2 matrix using both continuous wave (CW) and nanosecond optical pumping. A first attempt is done to clarify the role of the leaky modes in the measurements. While the stimulated emission is not present under CW excitation, the intense pulsed pumping leads to optical gain of ~12 cm^{-1}.

2 Results and discussion

The sample was prepared by 400 keV Si$^+$-ion implantation (a dose of 4×10^{17} Si/cm^2) into a fused silica slab (Infrasil) with optically polished surface and edges. Subsequent annealing was performed in N_2 atmosphere for 1 hour at 1100 °C and then for 1 hour in a forming gas (5% H_2 in N_2) at 500 °C. The optical gain was measured by the standard VSL technique using either the 365 nm line of a CW Ar$^+$-ion laser or the high-fluency pulses (6 ns, 10 Hz, 355 nm) of a third harmonic of a Nd-YAG laser. The emission from the sample edge was detected either by a double grating monochromator and a photomultiplier operating in photon counting regime (CW VSL) or by a single grating spectrometer connected to a streak camera (pulsed VSL). Signal from the streak camera was integrated over a 90 nm wavelength window. All the spectra were corrected for the spectral response of the detection systems. The experiments were performed at room temperature.

2.1 Waveguiding properties

The peculiar waveguiding properties of the sample are demonstrated in Fig. 1. While surface emission photoluminescence (PL) measurement (curve *a*) shows a wide emission band in the red spectral region 620-950 nm, the PL collected from the edge of the sample (curve *b*) reveals significant lineshape differences. The main features are narrow (~ 20 nm full-width-at-half-maximum), polarization-resolved TE (~ 695 nm) and TM (~ 720 nm) modes.

The development of the mode pattern in the waveguide emission can be followed by measuring the emission spectrum as a function of the distance of the excitation spot position from the sample edge (so

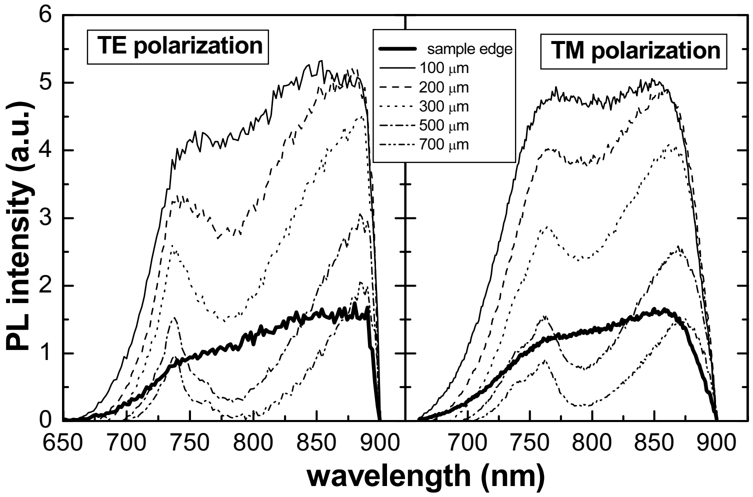

Fig. 2 Evolution of polarization resolved guided PL spectra as a function of the distance of the excited spot from the sample edge. PL was excited by the 365 nm line of a cw Ar^+-ion laser (pump intensity of 2 kW/cm^2). The decreasing edge at 870 nm is due to the cut-off of the measuring system.

called shifting-excitation-spot (SES) experiment [14]). Figure 2 shows the evolution of the edge emitted PL spectra in a SES experiment. For both polarizations, the broad featureless spectrum measured when the excitation is near the sample edge transforms progressively into a low wavelength peak due to the leaky modes, overlapped with an unpolarized broad emission band due to waveguide modes. Slightly shifted PL, TE and TM peak positions in comparison with the measurement in Fig. 1 are due to the different excitation and detection conditions.

It is obvious that analogous change of the emission spectra will show up in the VSL experiment (where instead of exciting small spot we deal with stripe-like excitation) even when the sample does not exhibit any optical gain [15]. This can cause serious difficulties in interpreting the VSL data [14], since the

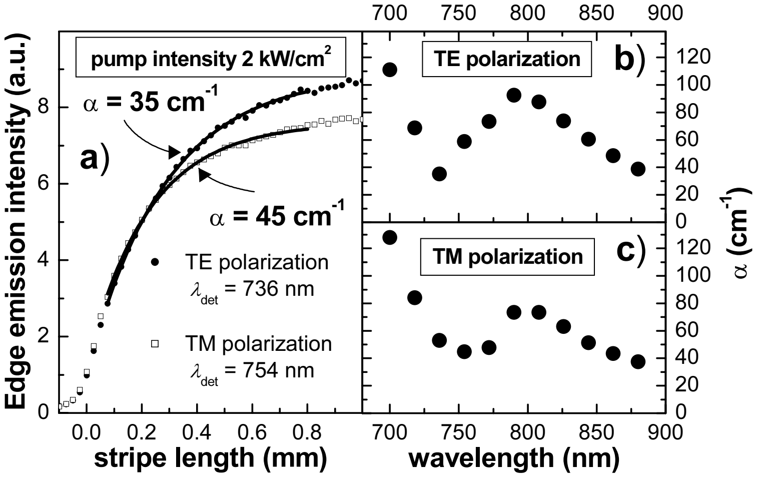

Fig. 3 a) Results of the CW VSL measurement at the peak of TE and TM modes. Theoretical fits (solid lines) based on one dimensional optical amplifier model give optical losses $\alpha = (35 \pm 2)$ cm^{-1} for TE and $\alpha = (45 \pm 4)$ cm^{-1} for TM mode, respectively. b), c) Spectra of the optical losses for both polarizations determined from the VSL curves recorded at different wavelengths.

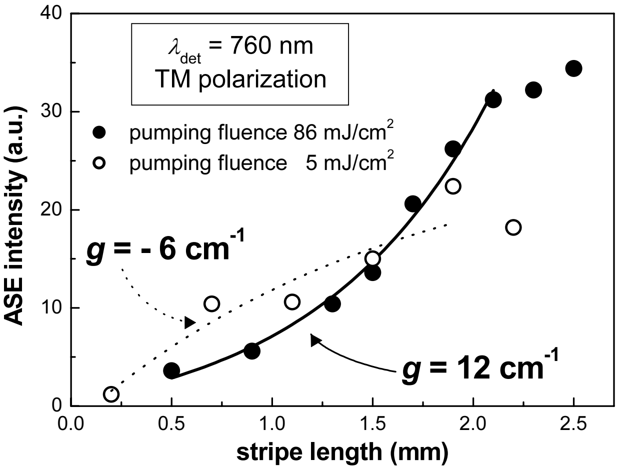

Fig. 4 Time resolved VSL curves at 760 nm for two different excitation intensities. Amplified spontaneous emission (ASE) peak intensity is plotted as a function of the stripe length. Theoretical fit of the data (full line) yields net modal gain value of $g = (12 \pm 2)$ cm^{-1} at a pumping fluence of 86 mJ/cm^2 and losses $\alpha = (6 \pm 6)$ cm^{-1} at 5 mJ/cm^2.

change of the amplified spontaneous emission (ASE) spectrum due to optical gain is frequently used for evaluation of the optical gain spectrum. Therefore, in the case of the substrate leaky-mode emission, the true optical gain spectra should be derived from the VSL curves recorded for a set of emission wavelengths.

2.2 Optical gain

Results of the CW VSL measurement are summarised in Fig. 3. Panel a) shows an example of the VSL curves measured at the wavelengths corresponding to the TE and TM modes, respectively. After initial sharp increase due to the diffraction on the moving slit, both curves show a sublinear increase indicating optical losses in both cases. However, spectra of the optical losses shown in Fig. 3 b) and c) reveal reduced optical losses in both modes. This observation is explained by the fact that the leaky modes propagate partially in the substrate (close to the core/substrate boundary) where Mie scattering and absorption losses are lower [13]. Indeed the TE and TM modes “leak out” step by step to the optically transparent SiO_2 substrate. On the other hand, the normal waveguide modes (e.g. emission at wavelengths different from the one of the TE or TM modes) show larger losses due to the presence of the Si-nc and also due to scattering by the sidewall roughness [16].

Pulsed excitation in the VSL experiment, nevertheless, can achieve higher pumping fluence while reducing thermal effects and achieving the inversion factor needed to overcome the critical competition between the stimulated emission rate and the fast nonradiative Auger quenching [3]. Under pulsed excitation conditions, a new fast component ($\tau \sim 100$ ns) appears for long stripe lengths and above certain pumping threshold, which is superimposed to the usual slow component ($\tau \sim 1$ μs) [4]. Figure 4 plots the peak emission intensity collected from the sample edge as a function of the stripe length for emission wavelength of 760 nm (TM leaky mode) and for two very different excitation fluences. A sublinear dependence showing optical losses of 6 cm^{-1} at low excitation fluence changes into an exponential increase at 86 mJ/cm^2 indicating the presence of stimulated emission. The estimated net modal gain is 12 cm^{-1}.

A supporting observation, characteristic of stimulated emission and proving the presence of optical amplification, is shown in Fig. 5. A clear threshold at ~50 mJ/cm^2 in the excitation fluence dependence separates two different recombination kinetics. Above the threshold, a superlinear emission regime is observed, accompanied by a decrease of the emission lifetime.

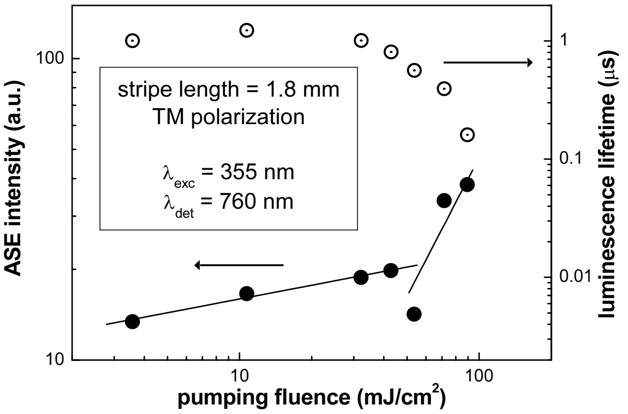

Fig. 5 Full circles: ASE peak intensity as a function of the excitation fluence for fixed excitation length 1.8 mm. Open circles: fast component lifetime. Solid lines are guides for the eyes.

The estimated magnitude of the gain, however, remains slightly lower than the values obtained in samples containing Si-nc by other authors [2–6]. Also the above-threshold slope (~1.5) of the ASE intensity dependence on the pumping power (see Fig. 5) is remarkably smaller [3, 5]. We believe that this can be mainly due to two facts. First, optical gain in the leaky modes is expected to be lower – if any – since the excitation energy leaks out step by step from the population-inverted core. Second, the damage threshold of this sample is quite low which prevented us to use as high fluences as the one used in other works.

3 Conclusions

In conclusion, we have demonstrated that an active planar optical waveguide made of silicon nanocrystals exhibits peculiar polarization-resolved substrate modes in the photoluminescence collected from the sample edge. In these "leaky" modes, time resolved VSL experiment under ns excitation revealed the presence of stimulated emission (net modal gain ~12 cm^{-1}). The presence of optical amplification was further supported by a superlinear ASE increase above a pumping threshold of ~50 mJ/cm^2 together with a fast recombination lifetime shortening.

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