Closely packed luminescent silicon nanocrystals in a distributed-feedback laser cavity

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Abstract. Silicon nanocrystals (Si-ncs) of sufficiently small size, emitting luminescence at short wavelengths (which implies the occurrence of quasi-direct radiative recombination) and being densely packed in a planar thin film (which ensures short stimulated emission (StE) lifetime) can become a suitable active material for the observation of StE in the visible region. In this paper, we describe a fabrication method of nanostructures of this type, based on enhanced electrochemical etching of silicon wafers followed by embedding porous silicon grains into an SiO2 matrix. Further, we report on time-resolved photoluminescence spectra and optical gain measurements performed via the variable-stripe-length and the shifting-excitation-spot methods. Finally, we realize a transient wavelength-tunable distributed-feedback-laser (DFL) cavity with inserted densely packed Si-ncs as an active medium. We demonstrate an increase in emission intensity on the blue emission wing (below 600 nm), which is spectrally shifting in accordance with the cavity tuning. We also present a mathematical model of the DFL cavity enabling us to simulate the experimental

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observations and analyze a realistic prospect for achieving laser action in an ensemble of Si-ncs.

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1. Introduction

The use of silicon materials as an active medium for light-emitting devices (light-emitting diodes or injection lasers) remains an open issue for basic research, even if for commercial applications in future silicon photonics, the possibility of using various hybrid solutions has appeared (AlGaNAs or GaN lasers on a silicon chip [1, 2]). The basic problem of light amplification and lasing in silicon, based on carrier recombination across the band gap, has not been fully understood and solved yet. A variety of possible approaches toward this goal have been suggested [3], silicon nanocrystals (Si-ncs) embedded in SiO$_2$ matrices (Si-ncs/SiO$_2$) being one of them [4]. Several different groups have independently reported single-passage light amplification in Si-ncs-/SiO$_2$-based samples [5]–[9], however, no laser-like emission has been reported yet. Another way being followed intensively at present is represented by Er-ions coupled to Si-ncs in a transparent matrix, for a recent review see, e.g. [10].

The key properties of a laser oscillator are (i) the presence of light amplification by stimulated emission (StE), i.e. positive net optical gain, (ii) high optical quality of the active material and (iii) sufficiently high quality of the optical cavity. For the StE onset in silicon materials, we need its build-up time to be shorter than the gain relaxation, driven by the dynamic competition between the radiative and nonradiative recombination processes. Fast nonradiative Auger recombination (Auger recombination time $\tau_A \sim 2$–$10$ ns) and efficient free carrier absorption (FCA) deplete the population inversion at high carrier densities and avoid StE, i.e. the relation $\tau_A < \tau_{\text{StE}}$, where $\tau_{\text{StE}}$ denotes the StE build-up time, usually holds. Moreover, the FCA further increases the total optical losses and lowers the net gain coefficient. However, in an ensemble of densely packed Si-ncs, $\tau_{\text{StE}}$ has been shown to be proportional to $\sim D^3(\sigma \xi)^{-1}$, where $D$ is the Si-nc diameter, $\xi$ denotes the Si-ncs volume fraction and $\sigma$ stands for the emission cross section per nanocrystal [11]. Thus, systems with a high volume fraction $\xi$ of small Si-ncs ($D$ small), in which, moreover, $\sigma$ gets considerably larger due to partial breakdown of the $\vec{k}$-selection rule (quasi-direct radiative transitions [12, 13]) seem to be favorable candidates for lasing ($\tau_{\text{StE}}$ drops rapidly and the inequality $\tau_A < \tau_{\text{StE}}$ can be achieved).
The FCA cross section decreases with decreasing Si-ncs emission wavelength as \( \sigma_{\text{FCA}} \sim \lambda^2 \), further supporting the necessity for the diameter of Si-ncs to decrease.

Preparation of such an ideal active medium composed of Si-ncs remains a technological problem. Our method described hereafter, which is based on an enhanced electrochemical etching of Si wafers combined with the sol-gel technology, yields relatively small oxidated Si-ncs \((D = 2.7 \text{ nm in average})\) embedded and passivated with a high-volume fraction \((\xi \text{ of the order of } 10^{19} \text{ Si-ncs cm}^{-3})\) in an SiO\(_2\)-based matrix. We report in this paper the detailed investigation of such thin films made of Si-ncs, aimed at attempting laser action in a resonator under optical pumping.

The paper is organized as follows. Section 2 describes techniques used for the sample preparation and characterization; section 3 is devoted to the steady-state and time-resolved photoluminescence (PL) study; section 4 shows optical gain measurements; section 5 presents the experimental realization of a distributed-feedback-laser (DFL) cavity, analyzes the DFL experimental results and compares them with a model, shown in the appendix. This comparison makes it possible to reveal the inhomogeneity of the Si-ncs distribution as one of the principal obstacles preventing the achievement of a net laser action in such samples. Finally, section 6 summarizes the results.

2. Fabrication and characterization of samples

Our preparation method is based on the electrochemical etching of crystalline Si substrate modified with respect to the original Canham work [14] and combined with sol-gel technology [15]. The first step consists of the electrochemical preparation of porous silicon (por-Si) via etching p-type silicon wafers \((\langle 100 \rangle, \sim 0.1 \Omega \text{cm})\) in a solution composed of hydrofluoric acid (HF) (50%), pure ethanol (EtOH) and hydrogen peroxide (H\(_2\)O\(_2\)). Platinum is used as the second electrode and the bath is continuously stirred. We refer to three different types of por-Si in this work: ‘standard’, ‘yellow’ and ‘white’, labeled according to the etching and post-etching bath content (differing in additional H\(_2\)O\(_2\) treatment and their color appearance under daylight).

The ‘standard’ por-Si is etched in a mixture of HF : EtOH\(= 1 : 2.5\) without the addition of H\(_2\)O\(_2\) at a constant density of 1.6 mA cm\(^{-2}\) for 2 h. The resulting porous layer is then mechanically scraped off the substrate into pure EtOH and left in an ultrasonic bath for 30–60 min in order to split larger agglomerates of nanocrystals. Finally, this colloidal solution is allowed to sediment in multiple steps to obtain small Si-ncs with narrower size distribution. The ‘1st sediment’ powder is the sediment found after \(\sim 5\) min sedimentation in pure EtOH at the bottom of the cuvette (largest agglomerates). The remaining supernatant can undergo a similar procedure several times and in this way, we obtain the 2nd, 3rd and 4th sediments. The ‘yellow’ and ‘white’ types of por-Si make use of another way to reach a similar goal, i.e. to yield small Si-ncs with a relatively narrow size distribution by modification of the etching bath by adding H\(_2\)O\(_2\) [16] and increasing the etching current density. The ‘yellow’ type of por-Si is electrochemically prepared by etching in a solution of HF : EtOH : H\(_2\)O\(_2\) (3%) \(= 1 : 2.46 : 0.54\) at 2.3 mA cm\(^{-2}\) for 2 h. Moreover, the freshly prepared por-Si layer on Si wafer is immediately dipped into pure H\(_2\)O\(_2\) (3%) for 5 min in order to carry out a post-etching procedure. The ‘white’ type of por-Si is etched even more strongly using more concentrated (30%) peroxide H\(_2\)O\(_2\): etching is performed in HF : EtOH : H\(_2\)O\(_2\) (30%) \(= 1 : 2.85 : 0.15\) solution at 2.3 mA cm\(^{-2}\) for 2 h and then post-etching follows using H\(_2\)O\(_2\) (30%) for 15 min. The
effect of $\text{H}_2\text{O}_2$ consists of additional oxidizing activity on the por-Si surface, described by the equation

$$2\text{Si} + 4\text{H}_2\text{O}_2 \rightarrow 2\text{Si(OH)}_4 \leftrightarrow \text{Si(OH)}_3 - \text{O} - \text{Si(OH)}_3 + \text{H}_2\text{O}. \quad (1)$$

During this process, the crystallite core size shrinks and its surface gets passivated by the $-\text{OH}$ groups $[17]–[19]$. No sedimentation in the liquid is obtained in the case of the ‘yellow’ and the ‘white’ por-Si, after rinsing in pure EtOH. However, the post-etched wafers are left to age for several days in stable ambient conditions to develop bright PL. The resulting porous layers are then mechanically scraped off the substrate into the pure EtOH and given an ultrasonic treatment to separate larger Si-ncs agglomerates. The fine Si-ncs powder is dried. A PL photo of the por-Si and the resulting Si-ncs powder under UV lamp irradiation is shown in figure 1(A)(a, b).

Weighted amounts (around 1–3 mg) of the Si-ncs powders are then mixed with a liquid SiO$_2$-based commercially available spin-on-diffusant P509 (Filmtronics Inc.) with phosphorus ($\sim 5 \times 10^{21}$ at. cm$^{-3}$). The aim of using a phosphorus-doped SiO$_2$ matrix is twofold—to further decrease the Si-ncs emission wavelength $[20]$ and increase the emission efficiency $[21]$. The liquid mixture is poured into spectroscopic quartz cuvettes and undergoes an additional ultrasonic treatment for homogenization. The Si-ncs powders sediment at the cuvette bottom, forming a dense Si-ncs-rich layer. To further increase the Si-ncs volume fraction, we apply an additional centrifuge treatment. Later, the samples solidify under ambient conditions within a few days. We would like to stress here that the samples were not heated above room temperature.

The resulting samples are transparent, brightly luminescent, well-passivated Si-ncs-rich SiO$_2$ planar layers of thickness around $\sim 50 \mu\text{m}$ of relatively good optical quality, close to the cuvette bottom (figure 1(A)(c)). The obtained volume fraction of Si-ncs/SiO$_2$ samples is usually between $\sim 10–20$ vol.% (corresponding to the order of $10^{19}$ ncs cm$^{-3}$), giving the highest PL intensity (figure 1(A)). The relatively large error bars in volume concentration are caused by Si-ncs-rich layer thickness fluctuations. Due to this, active Si-ncs-rich layers do not exhibit any waveguiding effects.
Figure 2. (A) Estimated size distribution (left) from an HRTEM scan (right) of a ‘yellow’ Si-nc powder. (B) Normalized Raman spectra (full lines) of (a) a ‘standard’, (b) a ‘yellow’ Si-nc/SiO$_2$ sample and (c) reference crystalline silicon wafer, fitted with two Gaussian curves (dashed lines). The sharp peak position is 517.5 cm$^{-1}$ (FWHM of 13.7 cm$^{-1}$) in (a), 515.1 cm$^{-1}$ (FWHM of 12.3 cm$^{-1}$) in (b) and $\sim$521 cm$^{-1}$ (FWHM of 5.3 cm$^{-1}$) in (c).

This method is relatively simple and easily reproducible. The size of crystallites can be tuned to some extent by changing the etching parameters and the Si-ncs volume fraction in the sol-gel-based SiO$_2$ matrix is defined by the amount of Si-ncs powder, mixed with the sol-gel before solidification. This volume fraction is virtually arbitrary, limited by the material cohesion only.

Using Fourier transform infrared spectroscopy (FTIR), we characterized the differences between the surface passivation of the H$_2$O$_2$ treated ‘yellow’ Si-nc powder and the non-treated ‘standard’ one (figure 1(B)). In the ‘yellow’ type, we observed higher signals of Si–OH-related bands at 870 and 945 cm$^{-1}$, and a strong signal at 3310 cm$^{-1}$, related possibly to remanent water, stemming from the H$_2$O$_2$ reaction with Si (equation (1)) [22]. Other peaks are related to the Si–O–Si vibration modes (792, 1050 and 1160 cm$^{-1}$). They are stronger in the ‘yellow’ powder which indicates better surface passivation when using H$_2$O$_2$.

The size distribution of the Si-nc powder was characterized using high-resolution-transmission-electron-microscopy (HRTEM). In figure 2(A), we show the HRTEM scan and an estimated size distribution of a ‘yellow’ Si-ncs powder. Taking into consideration the obtained Gaussian profile of the size distribution, we find the average size of Si-ncs to be $\sim$2.69 nm with full width at half maximum (FWHM) of $\sim$0.86 nm.

The mean size of the Si-ncs can also be measured ‘indirectly’ from Raman spectra. The mean diameter of the crystalline Si-nc core is directly connected with the shift of the sharp crystalline Raman peak at $\sim$520 cm$^{-1}$ (figure 2(B)(c)) [23]. The Raman spectra of ‘standard’ and ‘yellow’ Si-nc/SiO$_2$ samples containing dense layers of Si-ncs are plotted in figure 2(B) (a, b), as measured using an InVia Renishaw Raman micro-spectrometer with 325 nm excitation. Measured peak shifts in the ‘standard’ and ‘yellow’ samples correspond to the mean Si-ncs diameters of $\sim$3.4 and 2.5 nm, respectively, in good correspondence with the HRTEM results. This result also confirms that the presence of H$_2$O$_2$ in the etching procedure leads to reduction of the size of the Si-ncs.
3. Steady-state and time-resolved PL study

Detailed knowledge of spontaneous PL properties is a key prerequisite for optical gain studies. All experimental data shown hereafter were taken at room temperature and spectra were corrected for the spectral response of the detection system. The steady-state PL (SSPL) emission spectra were excited with a cw HeCd laser (325 nm) under \( \sim 0.5 \) W cm\(^{-2}\) pump power density.

Figure 3(A) shows SSPL spectra taken from different types of Si-ncs/SiO\(_2\) samples (i.e. after embedding the por-Si powders into the SiO\(_2\) matrix), original por-Si (inset), reference Si-ncs/SiO\(_2\) samples and the pure SiO\(_2\) matrix. The reference ‘IT’ Si-ncs/SiO\(_2\) sample is prepared using larger Si-ncs of diameter \( \sim 5 \) nm, fabricated by laser pyrolysis by Trave \textit{et al} \cite{24} and embedded in the SiO\(_2\) matrix using our sol-gel technique. Its emission spectrum is peaked at \( \sim 877 \) nm. Another reference sample was prepared by Si\(^+\)-ion implantation into an Infrasil plate. The effect of the decreasing size due to the additional H\(_2\)O\(_2\) treatment of the por-Si can be seen from the blue shift of the SSPL spectra of the native por-Si (inset in figure 3(A)) and the resulting Si-ncs/SiO\(_2\) samples (figure 3(A)): the spectrum of our ‘standard’ Si-ncs/SiO\(_2\) sample (no H\(_2\)O\(_2\) etching treatment) is peaked around 670 nm and is quite broad (FWHM of 190 nm), while spectra of the H\(_2\)O\(_2\)-treated ‘yellow’ and ‘white’ Si-ncs/SiO\(_2\) samples are noticeably blue shifted to \( \sim 630 \) and \( \sim 595 \) nm, respectively. Moreover, our numerical estimation of an inhomogeneous broadening shows that the considerable narrowing of the spectra (FWHM of 140 and 116 nm, respectively) indicates that, owing to H\(_2\)O\(_2\) treatment, also the Si-ncs size distribution gets narrower which can be considered—apart from the blue-shift—as an additional favorable factor for a gain medium with Si-ncs.
The overall blue-shifted position of the main PL band in our samples as compared with commercially red-emitting Si-ncs is accompanied, as expected, by a sharp decrease of the PL decay time from a few microseconds down to $\sim 25$ ns. An example of time-resolved spectra in a ‘standard’ second sediment sample is shown in figure 3(B). Signal was excited using a pulsed UV laser Nd:YAG (355 nm, pulse width 7 ns, repetition rate 10 Hz) and detected by a pre-triggered intensified ICCD camera with a gate width of 10 ns. The PL spectra exhibit three main spectral bands—a fast ‘blue’ one around 450 nm, a ‘green’ one at $\sim 550$ nm and a slow ‘orange’ one at 620 nm. Since the ‘blue’ and ‘orange’ bands are present in the SSPL emission spectra of the native por-Si (inset in figure 3(A)), their origin can be associated with Si-ncs (at least the Si-ncs surface states) [26]–[28]. The ‘green’ band, on the other hand, is much weaker and hardly separable from the strong ‘blue’ emission band, making its investigation difficult. We tentatively assign this band to Si-nc core exciton radiative recombination, since the non-oxidized freshly prepared H-terminated por-Si emission under UV-lamp excitation appears ‘greenish’ (to the naked eye) for a few seconds, before complete oxidation by air.

This is also supported by the PL decay and coefficient $\beta$ investigations in figure 3(C). The continuous shortening of the PL decay time with emission wavelength strongly indicates gradual switching from indirect to quasi-direct exciton radiative recombination in sufficiently small Si-ncs. Therefore, in the measurement of optical gain and realization of the DFL resonator we fix our attention mainly on the high energy wing of the emission band between $\sim 550$ and $\sim 620$ nm. The coefficient $\beta$, describing the stretched exponential behavior of the decay $I_{PL}(t) = I_0 \exp(- (t/\tau)^\beta)$ ($0 \leq \beta \leq 1$), lies for 550–650 nm between 0.4 and 0.5, which is characteristic for disordered systems like a Si-ncs ensemble [29]–[31].

The difference between the emission dynamics of ‘standard’ and $H_2O_2$-etched samples appears to be significant under femtosecond time resolution only [25], interpreted in terms of different rate of confined free carriers trapping on the surface states (figure 3(D)). This observation and interpretation have been recently confirmed by Sykora et al [13].

### 4. Optical gain measurements

Optical gain coefficient $G$ and StE spectra were measured using the standard variable-stripe-length (VSL) method [32], combined with the shifting-exciton-spot (SES) method proposed in [33] and [34]. The reason for combining the VSL and SES methods is to distinguish undoubtedly a small optical gain from experimental gain-like artifacts, caused by discrepancy between the one-dimensional (1D) VSL model and a real experimental setup [33]–[35]. Both the VSL and SES methods contain the same total optical losses $\alpha_{tot}$ and the same artifacts. The total optical losses are given by $\alpha_{tot}(\lambda) = \alpha(\lambda) + K$, where $\alpha(\lambda)$ stands for the absorption coefficient and $K$ for scattering losses. Because the SES method uses an excitation spot of a sufficiently small size ($\sim 60 \times 100 \mu m^2$) to prevent StE onset, the optical gain can occur only within the VSL signal. By comparison with the SES signal, one can decide whether the observed effects are due to real optical gain or artifacts. Moreover, because the possible gain-like artifacts are not dependent on the excitation intensity (contrary to the real optical gain coefficient), one should measure the gain coefficient dependence on the excitation intensity and find the difference between the VSL and the SES signals, the latter being integrated over the stripe length $l$:

$$I_{VSL}(l, \lambda) - \int_0^l I_{SES}(x, \lambda)dx = I_{StE}(\lambda) \left( e^{G(\lambda)l} - 1 - \frac{1 - e^{-\alpha_{tot}(\lambda)l}}{\alpha_{tot}(\lambda)} \right),$$

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Figure 4. VSL and SES results measured in a ‘standard’ second sediment Si-ncs/SiO$_2$ sample. (a) VSL (symbols) and integrated SES (lines) comparison for different excitation intensities $I_1 < I_2 < I_3 < I_4$, detected at 610 nm. (b) VSL for the lowest (black symbols; 10 times magnified) and the highest (red symbols) excitation intensity (detail from (a)), fitted with a net gain coefficient of $-73$ and $41$ cm$^{-1}$, respectively. (c) Difference between VSL and integrated SES curves (symbols) from (a), fitted by equation (2) (lines). (d) Output intensity as a function of the excitation intensity for two stripe lengths of 200 µm (red symbols) and 400 µm (black symbols). (e) Gain spectra at the lowest (black symbols) and the highest (red symbols) excitation intensity, measured with a stripe length of 300 µm.

where $I_{SpE}(\lambda)$ indicates the spontaneous emission intensity per unit length, $G(\lambda) = g(\lambda) - K$ denotes the net optical gain coefficient and $g(\lambda)$ stands for the material optical gain coefficient (negative absorption coefficient). Light amplification is observed if for some length $l$ the difference given by equation (2) becomes positive.

In figures 4(a–e), we illustrate the abovementioned procedures by presenting results obtained in a ‘standard’ second sediment Si-ncs/SiO$_2$ sample. An XeCl excimer laser (308 nm, pulse width 15 ns and 10 Hz repetition rate) was used as an excitation source. Signal was detected at the peak maxima of $\sim$610 nm. Comparison of the VSL and integrated SES results in figure 4(a) shows increasing difference between the two curves (separately shown in figure 4(c)) with increasing excitation intensity—clear evidence for StE observation. Notice that for $l$ shorter than a critical length ($\sim$200 µm) the VSL and SES curves are identical and cannot serve for gain evaluation. The total optical losses at 610 nm, evaluated from the SES measurement, were found to be $\alpha_{tot} = 73$ cm$^{-1}$.

The VSL curves for the highest and the lowest excitation intensities are plotted in figure 4(b): clear switch in the VSL curve shape with the excitation intensity was observed, caused by the change of the net gain coefficient from a negative to a positive value very much like in [36]. The difference between the VSL and integrated SES curves, plotted in figure 4(c), is always zero below the critical length and increases with the excitation intensity above this length (StE onset). Optical gain values can then be safely evaluated from the difference formula equation (2) as $g = 0$ cm$^{-1}$ (for 0.38 MW cm$^{-2}$); 1 cm$^{-1}$ (0.57 MW cm$^{-2}$); 28 cm$^{-1}$
Figure 5. (A) VSL spectra measured in a ‘standard’ second sediment Si-ncs/SiO$_2$ sample as a function of the excitation intensity with a fixed excitation stripe length of 1.3 mm. The spectral width (FWHM) decreases with increasing excitation intensity as shown in the inset. (B) Intensity dependence of time-integrated VSL signal (at 630 nm) measured in another ‘standard’ second sediment Si-ncs/SiO$_2$ sample using stripes of four fixed lengths: (a) 380 $\mu$m, (b) 950 $\mu$m, (c) 3660 $\mu$m and (d) 1 cm. Moreover, in (d) the VSL signal has been enhanced by an external cavity.

(1.18 MW cm$^{-2}$) and 114 cm$^{-1}$ (1.46 MW cm$^{-2}$), i.e. the net gain $G$ is positive only for the highest excitation intensity, where $G = g - K = 41$ cm$^{-1}$. The output intensity as a function of the excitation intensity is plotted on a log–log scale in figure 4(d). The switch from a sub-linear slope for the shorter excitation stripe length to a super-linear one for the longer stripe is further evidence for the StE onset, as well as the behavior of the gain spectrum $G(\lambda)$ in figure 4(e) (which is negative for the lowest excitation intensity and positive for the highest excitation intensity).

Spectral narrowing with increasing excitation intensity is supposed to be one of the important features of amplified spontaneous emission (ASE) spectra. This effect is, however, expected to be very low in such a wide size distribution ensemble of Si-ncs, where the inhomogeneous broadening dominates the spectral shape. In figure 5(A), we have plotted VSL spectra measured at a different place of the same sample as in figure 4, with a fixed stripe length of 1.3 mm and under variable excitation intensity (0.6–0.5 MW cm$^{-2}$). The spectral FWHM as a function of the excitation intensity is shown in the inset of figure 5(A). FWHM decreases from $\sim$180 nm for the lowest excitation intensity down to $\sim$100 nm for the highest one. The spectral narrowing of the inhomogeneously broadened spectrum can be calculated according to [37] from the Lindford formula $\Delta\nu_{ASE}^2/\Delta\nu_0^2 = (e^{G(\lambda)L} - 1)/(e^{G(\lambda)L}G(\lambda)))$, where $\Delta\nu_{ASE}$ is the Gaussian profile FWHM of the ASE spectrum and $\Delta\nu_0$ that of the spontaneous emission profile. A simple calculation yields an approximate value of $G \sim 20$ cm$^{-1}$, close to the measured gain magnitude.

Finally, time-integrated VSL intensity as a function of the pump intensity is plotted in figure 5(B), using a stripe of four different lengths (a) 380 $\mu$m, (b) 950 $\mu$m, (c) 3660 $\mu$m and (d) 1 cm. These data have been obtained with another sample containing half the amount of Si-ncs (lower Si-ncs content entails a better optical quality, but longer stripe lengths have to be used to get significant amplification effect). For the longest stripe of 3.66 mm, a clear switch from a sub-linear slope ($\sim$0.76) to a super-linear one ($\sim$1.33) at the threshold pump intensity of
∼2 MW cm\(^{-2}\) can be seen. This can be considered as another sign of the StE onset. The panel (d) in figure 5(B) deserves special attention. Here, the StE has been enhanced using an external mirror of high reflectivity (>90%) at the opposite side of the cuvette containing the sample. The mirror has been adjusted precisely perpendicular to the stripe axis, i.e. we built a simple laser cavity. In this case, we found an enhanced threshold behavior of the output–input intensity dependence with a sub-linear slope in the log–log scale of ∼0.60 together with a super-linear slope of ∼1.90 above the threshold intensity of ∼3.0 MW cm\(^{-2}\).

This result represents a continuous extension of the observations discussed above and deserves to be stressed because it demonstrates beyond doubt—and independently of the VSL fitting and without applying the SES method—that positive net gain occurs in our (best) samples. It is also important to note that the VSL/SES results differ from sample to sample and from place to place which means that the occurrence of positive optical gain cannot be guaranteed even with ideal Si-ncs size, high volume fraction and good surface passivation; sufficient optical quality of the sample is also required. Optical quality also represents the main limitation for the optical feedback application, as we will explain hereafter.

5. Distributed-feedback laser (DFL)

5.1. Basic principles

High long-distance losses estimated from the SES measurements (of the order of tens of cm\(^{-1}\)) are one of the crucial problems for external cavity applications because the emitted light loses its coherence and directionality over distances shorter than the external cavity length. Lasing and mode selection is then attainable only by using the cavity applied on shorter distances. To this end, we have applied an optically induced DFL cavity, built in Bor’s configuration [38] (figure 6). Some of our preliminary results can be found in [39, 40]. An intense pulsed excitation beam (XeCl excimer laser, \(\lambda_{\text{exc}} = 308\) nm, 10 Hz repetition rate, ∼15 ns pulse duration, ∼0.3 MW cm\(^{-2}\)) with a homogeneous rectangular beam cross section is divided by a holographic grating (grating constant \(d = 333\) nm) into the two first diffraction orders of almost equal intensities \(I_{\text{exc}}\) under an angle \(\alpha = \arcsin(\lambda_{\text{exc}}/d) = 67.5^\circ\). Both beams, focused with the aid of a cylindrical lens, are then reflected by two mirrors, revolving over the angle \(\delta\), and focused under the incident angle \(2\theta = 2(\alpha - 2\delta)\) onto the active
medium where they interfere. The stripe-like spatial profile of the excitation beams leads to a grating-like interference pattern (of maximal length \(\sim 1 \text{ cm}\) and width \(\sim 100 \mu\text{m}\)) with the grating period \(\Lambda = \frac{\lambda_{\text{exc}}}{2 \sin \theta}\), tunable by the angle \(\theta\). The intense periodic illumination in the active nonlinear medium modulates both the net gain/absorption coefficient and refractive index \(n\). The intensity-dependent refractive index is defined as 
\[ n = n_0 + \Delta n(I_{\text{exc}}) \]
where \(n_0\) is the linear refractive index and a value of \(\Delta n \approx 0.006\) was estimated from [41] for our excitation conditions.

The absorption/gain coefficient modulation contribution \(\Delta g\) to the reflectance \(R\) can be neglected, because 
\[ |\Delta g\lambda/4\pi| \ll |\Delta n| \]
The value of reflectance is low, nevertheless, for a total cavity length of 1 mm (limited by the short coherence length of the XeCl excimer laser, see the appendix) we obtain approximately \(10^4\) elemental mirrors and the total reflectance becomes \(\sim 10^{-2}\) and \(10^{-1}\), since we consider a rapid loss of the spatial coherence of waves during their propagation through the excited sample. Such a distributed feedback selects a cavity mode at a wavelength of
\[
\lambda_{\text{cavity}} \approx \Lambda 2n_0 = \frac{n_0\lambda_{\text{exc}}}{\sin \theta}, \quad \theta = \alpha - 2\delta.
\]

Tunability of the mode wavelength is realized by a synchronous change of the angle \(\delta\) on both the revolving mirrors. The selected mode is collected in the direction of the cavity axis from the edge of the sample by a plastic optical fiber, connected to a spectrometer and a cooled CCD camera, while the DFL cavity is kept in air at room temperature.

The lasing cavity mode selection is well defined and easily observed in high homogeneity samples with high-optical gain, such as e.g. in solutions of organic dyes in methanol. Our samples, however, exhibit a lower optical quality and a lower net optical gain. In this case, the DFL influence is less known and has to be carefully analyzed. Therefore we propose a simple model, presented in the appendix, to understand in detail the manifestation of the DFL cavity in our samples.

5.2. DFL experimental results and comparison with the simulations

To evaluate how the DFL cavity affects the emission mode shape and spectral position, we carefully examine the difference emission spectra. At first, both the excitation beams are incident on the sample and we record an emission spectrum \(I_{\text{on}}(\lambda)\), called ‘DFL on’ (see figure A.1(b) in the appendix). Then, only a single excitation beam (with doubled intensity) is left, i.e. no interference pattern occurs and no feedback is thus applied. We obtain a ‘DFL off’ spectrum, \(I_{\text{off}}(\lambda)\) (see figure A.1(b) in the appendix). The difference emission spectrum is then acquired as 
\[ I_{\text{diff}}(\lambda) = I_{\text{on}}(\lambda) - I_{\text{off}}(\lambda). \]

The cavity mode shows up, blue-shifts (figure 7(b)) and gets narrower (figure 7(c)) with increasing intensity from 605 nm (FWHM of 210 nm) down to 572 nm (FWHM of 76 nm). Therefore, when comparing DFL spectra under variable angle \(\delta\) (hereafter), the applied excitation intensity has to be fixed. Each time the functionality of the DFL setup was, before measuring our samples, tested using a high-optical quality reference solutions of organic dyes in methanol in which the lasing modes were successfully observed [39]. In figure 7(d), we present the DFL difference spectra measured with a fixed excitation intensity and a variable angle \(\delta\). The main cavity mode has been observed around 570 nm with FWHM of 100–145 nm, in good
Figure 7. (a) Normalized DFL difference spectra measured in a ‘yellow’ Si-ncs/SiO\textsubscript{2} sample with a fixed angle $\delta$ and a variable excitation intensity. (b) Peak position shift and (c) FWHM narrowing with increasing excitation intensity. (d) DFL difference spectra of a different ‘yellow’ Si-ncs/SiO\textsubscript{2} sample. Spectra were measured at a fixed excitation intensity and a variable angle $\delta$. The spectra were decomposed and fitted using Lorentz shape curves (dashed lines).

correspondence with the expected theoretical values (arrows in figure 7(d)) calculated according to equation (3) for refractive index $n_{0} \approx 1.731$. Side modes appear around 490–500 nm with FWHM of $\sim$40 nm, supplying experimental evidence (within our model, see the appendix) of the occurrence of ‘sharply’ defined grains of Si-ncs, distributed within the SiO\textsubscript{2} matrix. This yields very important quantitative information about the inhomogeneous distribution of Si-ncs in our active medium: the nanocrystals are crowded into larger clumps with a diameter comparable with the emission wavelength ($\sim$600 nm). Light scattering due to these inhomogeneities then represents the main contribution to the overall optical losses.

As a consequence of the ‘homogeneity’-related origin of the side modes, their occurrence is strongly sample and place dependent. Compare the spectra of three different samples as displayed in figures 7(d), 8(a) and 8(b): the side modes occur only in two of them, figures 7(d) and 8(a). On the other hand, another very important result mentioned above, namely, the observed shift of the main mode at 570–600 nm as a function of $\delta$ [39] occurs in all the samples. The shift is consistent with theoretically calculated spectral positions and this observation clearly demonstrates that the Si-ncs in the cavity ‘feel’ the positive-feedback effect.

It is also worth noting that all the presented difference spectra (figures 7 and 8) have positive values. This observation confirms again the occurrence of positive optical gain in the active medium (otherwise the difference spectrum is expected to be negative, as follows from simulations in figure A.1(c), where $I_{\text{tot}} < 1$ for $G = 0$).

6. Summary and conclusions

We have developed an original method for preparation of samples with virtually arbitrary content of small Si-ncs ($\sim$2–3 nm) exhibiting blue-shifted emission spectra. In measuring the
optical gain, we have applied an approach combining the SES and VSL techniques which enable us to extend reliably the VSL applicability to relatively low net gain values. We have revealed a positive net optical gain of $\sim 40 \text{ cm}^{-1}$ in the ‘slow’ emission component at 600 nm, originating from the ensemble of Si-ncs. This appears to be a very interesting observation, since fast Auger recombination caused by confined exciton interaction should avoid population inversion at such a long timescale. In our samples, however, ultrafast self-trapped exciton formation probably leads to Auger suppression even for longer times due to the separation of the spatial carriers.

We have made the first attempt to realize an optically pumped laser containing Si-ncs as an active medium, making use of an optically induced DFL cavity. A simple model allows us to describe the effect of such a distributed feedback on the emission of low homogeneity samples. Our central finding is twofold: (i) comparison of this DFL theoretical model with the observed difference spectra enables us to quantify the inhomogeneity of the distribution of the active centers (Si-ncs) in the SiO$_2$ matrix (clusters with typical dimensions of 600–800 nm). This causes high optical losses due to Mie scattering. (ii) The spectral tunability of the output modes in the DFL difference spectra proves that the Si-ncs feel the resonator feedback and respond to it, although output emission modes are considerably broad, mainly due to the low quality and insufficient length of the DFL resonator and to a mediocre homogeneity of the samples. Moreover, the inhomogeneous broadening of the absorption/gain spectra, related to the broad size distribution of the Si-ncs, also contributes to the mode broadening. True lasing has not been achieved, even though the positive sign of the DFL difference spectra has turned out to be additional evidence for the presence of positive optical gain in our samples, independent of the VSL and SES measurements.

Further steps toward lasing in such a system should include sample homogenization, narrowing of the Si-ncs size distribution and using an excitation laser with longer coherence length to improve the quality of the distributed cavity.

**Figure 8.** (a, b) Normalized DFL difference spectra measured in two different places of ‘standard’ third sediment Si-ncs/SiO$_2$ samples for various angles $\delta$ and a fixed excitation intensity. The theoretical spectral positions in (b) are indicated by arrows.
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Appendix. DFL theoretical description

The interference pattern quality, which defines the quality of the feedback, is given by its length and modulation depth, both driven by the temporal and spatial coherences of the two incident excitation beams interfering on the surface of the sample. The temporal coherence describes the ability of the radiation to interfere in dependence upon the difference in path lengths between the two beams. The maximum allowed path difference \(|L - L'|\) (sketch in figure A.1(a)) is equal to the coherence length \(l_{\text{coherence}}\), which in our case (XeCl excimer laser) is relatively short (~200–300 \(\mu\)m) [42, 43]. The necessary condition for the interference observation therefore reads \(|L - L'| \leq l_{\text{coherence}}\) and is always fulfilled for \(\delta = 0\) and for all the points on the axis of symmetry, where \(|L - L'| = 0\). On the other hand, for \(\delta \neq 0\), the optical path difference depends on the distance \(\Delta x\) from the axis of symmetry (sketch in figure A.1(a)), i.e. even for a small \(\delta\) only a short part of the total excited stripe will be modulated by the interference. The length of the interference pattern (DFL cavity) \(L_{\text{cavity}} = 2\Delta x\) can be calculated using a simple geometrical analysis to be

\[
L_{\text{cavity}} = \frac{l_{\text{coherence}} (\tan \alpha - \tan \delta) \cos \alpha \cos \theta}{\cos \theta - \cos \alpha}
\]

and is plotted in figure A.1(a) as a function of the angle \(\delta\). Our low value of coherence limits the cavity tuneability to e.g. \(\delta \in (-3^\circ, 3^\circ)\), when \(L_{\text{cavity}} \geq 1\) mm (full line in figure A.1(a)). Therefore, the cavity mode in our Si-ncs/SiO₂ samples with \(n_0 \approx 1.7\) can be tuned only in a narrow range \(\lambda_{\text{cavity}} \in \approx 546–595\) nm.

To describe our experimental results, we invoke the model sketched in figure A.1(b). The upper panel ‘DFL off’ describes the situation when only a single excitation beam (of intensity \(2I_{\text{exc}}\)) is incident on the sample surface, i.e. no interference pattern can be observed, i.e. no feedback is applied. The lower panel ‘DFL on’ describes the situation when two excitation beams (each of intensity \(I_{\text{exc}}\)) interfere on the sample surface and form a DFL cavity of maximal length \(L_{\text{cavity}} = N\Lambda\), where \(N\) is a natural number and denotes the total number of the interference maxima within the interference pattern with a grating period \(\Lambda\). The rest of the excitation stripe is not modulated and contributes, like in the VSL experiments, by a phase-insensitive light amplification only.

The DFL cavity influence on the emission spectra is studied via comparing the ‘DFL on’ and ‘DFL off’ spectra. Such a type of ‘comparison’ measurements represent a very powerful method in analysis and separation of weaker effects, which are present in the ‘On’ case only and are completely absent in the ‘Off’ regime (very much like in the VSL and SES methods).

Among the most important parameters of the model is the sample homogeneity. This can be described in terms of an average distance \(l_{\text{hom}}\) on which the light emitted by each single Si-nc preserves its directionality and coherence before it is scattered on inhomogeneities.
Figure A.1. (a) $L_{\text{cavity}}$ calculated using equation (A.1) as a function of the angle $\delta$ and laser coherence length $l_{\text{coherence}}$. Inset: scheme of the DFL setup situation for $\delta \neq 0$ and for a point at a distance $\Delta x$ from the axis of symmetry, where $|L - L'| \neq 0$. (b) A schematic model of the DFL mode selection calculation. (c) The DFL mode selection simulation according to equation (A.3) for different homogeneities $l_{\text{hom}}$, a fixed cavity length $L_{\text{cavity}} = 1$ cm ($N \approx 60\,000$) and $\lambda_{\text{cavity}} = 590$ nm. The average net gain coefficient $G$ was considered here to be equal to zero, the refractive index change under excitation $2I_{\text{exc}} = 288$ kW cm$^{-2}$ was taken as $\Delta n = 0.006$. The mode spectral width broadened and mode contrast lowered with decreasing sample homogeneity length $l_{\text{hom}}$. The mode position for lower $l_{\text{hom}}$ is red shifted with respect to $\lambda_{\text{cavity}}$. (d) Simulation of the cavity mode tunability for different $\delta$ using equation (A.3) with a fixed homogeneity $l_{\text{hom}} \approx 660$ nm ($K_{\text{hom}} = 4$).

The mode selection is then provided only by a part of the distributed cavity (‘cavity segment’) of length $l_{\text{hom}} = K_{\text{hom}} \Lambda$ around the single Si-nc emitter ($K_{\text{hom}}$ is a natural number and denotes the number of the unit mirrors in the cavity segment; see figure A.1(b)). The electric field of the light emitted by a single emitter, reflected backwards from such a ‘cavity segment’ of a total amplitude reflection coefficient $r_{\text{total}}$, can be expressed as $E_r = E_0 r_{\text{total}}$. The magnitude of $r_{\text{total}}$ can be estimated by many approximative methods. In our model, we have chosen the relatively simple intuitive method as known from the standard Fabry–Perot resonator, where the total amplitude reflection coefficient can be expressed by a sum of the partially reflected beams from distributed mirror units (see figure A.1(b)), each of reflectance $R$, i.e. transmittance $T = 1 - R$, decaying $\propto e^{-x/l_{\text{hom}}}$ (exponentially decaying dashed-line in figure A.1(b)). For the sake of simplicity, we calculate only with the first-order reflections, since the reflectance coefficient $R$ is very small.
The total amplitude reflection coefficient of the ‘cavity segment’ can then be written as

\[ r_{\text{total}}(\lambda) = \frac{1 - e^{-\frac{1}{K_{\text{hom}}} T} e^{2i K_{\text{hom}} \Phi(\lambda)}}{1 - e^{-(1/K_{\text{hom}}) T} e^{2i \Phi(\lambda)}} \int_{x=0}^{\Lambda} \exp({i(A(x) + B)} \Phi(\lambda)) F(x) e^{2i \Phi(\lambda)} (A + B) \, dx, \] (A.2)

where \( \Phi(\lambda) = 2\pi \Lambda n_0 / \lambda - i \tilde{G} \Lambda \) is the phase shift between the two mirrors and \( \Phi_{\text{deph}}(\lambda, x) \approx \Phi(\lambda) / 2 + 2\pi x n_0 / \lambda \) is the phase shift between the first mirror and the emitter position; \( F(x) = (B + A \cos(2\pi(x - \Lambda/2)/\Lambda))/(A + B) \) is a cosine-profile weight function to express the fact that the Si-ncs emission is excited by a cosine-like interference pattern profile (\( x \) is the emitter position, peaked at each interference maximum and also copying the interference minima; coefficients \( A \) and \( B \) define the interference pattern quality).

\( K \) is an eligible integer, defining the segment character (boundary conditions of the cavity segment). For \( K \gg K_{\text{hom}} \), we get the ‘blurred’ type of segment (material with ‘blurred’ type of inhomogeneities such as e.g. an inhomogeneous colloidal suspension), for \( K \approx K_{\text{hom}} \) we get the ‘sharp’ type (‘sharp’ type of inhomogeneities, as represented by grains of Si-ncs with contrasting refractive index in an SiO\(_2\) matrix). Hereafter, we apply the ‘sharp’ type condition. \( \tilde{G} \) stands for an average optical gain/absorption coefficient.

The total normalized emitted intensity from one emitter fixed in a position \( x_m \) is given by the sum of light emitted forward and light reflected backward in the direction of the detection system, which can be for \( l_{\text{hom}} \ll L_{\text{cavity}} \) written as \( I_{\text{m}}(\lambda) = I_0(\lambda)(1 + r_{\text{total}})^2 \), where \( I_0(\lambda) \) is the normalized spectrum of the PL emission of the ensemble of Si-ncs excited within the modulated part of length \( L_{\text{cavity}} = N \Lambda \) is given by the expression (being a sum of a geometric progression)

\[ I_{\text{tot}}(\lambda) = \frac{I_m(\lambda)}{N} T e^{\Lambda \tilde{G}} \left( \frac{1 - T e^{e^{N \Lambda \tilde{G}}}}{1 - T e^{e^{\Lambda \tilde{G}}}} \right). \] (A.3)

The numerical simulations of the cavity mode shape \( I_{\text{tot}}(\lambda) \) for a fixed mode position \( \lambda_{\text{cavity}} \) and a variable homogeneity length \( l_{\text{hom}} \) of 0.17 \( \mu \text{m} \) (\( K_{\text{hom}} = 1 \)), 0.33 \( \mu \text{m} \) (\( K_{\text{hom}} = 2 \)), 0.50 \( \mu \text{m} \) (\( K_{\text{hom}} = 3 \)), 0.66 \( \mu \text{m} \) (\( K_{\text{hom}} = 4 \)) and 0.83 \( \mu \text{m} \) (\( K_{\text{hom}} = 5 \)) are shown in figure A.1(c). The FWHM width of the selected mode obviously gets narrower with increasing homogeneity \( l_{\text{hom}} \) and also the mode contrast (i.e. the ratio of the mode minima to the maxima) increases. Due to the ‘sharp’ breakdown of a uniform grating (by e.g. the presence of a surface of large grains of Si-ncs), we can also observe additional side modes appearing around 430–490 nm, which do not show up in the spectra of samples with ‘blurred’ inhomogeneity character and high optical quality (not shown). An interesting fact is that the overall mode intensity is lower than one (note the scale on the ordinate axis) for the zero net gain coefficient \( \tilde{G} \) and zero total optical losses, i.e. the intensity is decreased by the cavity effect itself because of the losses caused by the induced grating reflectivity. The mode contrast is low (note the scale on the ordinate axis), mainly due to the low homogeneity and zero net optical gain. Due to this, it is difficult to ‘see’ the mode structure directly and we study the difference between the ‘DFL on’ and ‘DFL off’ emission spectra, i.e. DFL difference emission spectra.

The tunability of the cavity mode, simulated using equation (A.3), with a fixed sample homogeneity is shown in figure A.1(d). The mode position clearly shifts with varying angle \( \delta \), which is in the analysis of the experimental results, the most important evidence of the DFL
cavity functionality. When analyzing the difference emission spectrum of ‘DFL on’ and ‘DFL off’, equally important is that the positive sign of this difference spectrum is related to a positive net optical gain, since the difference in the case of the passive cavity will be negative, because of an increased light path in a lossy material. Comparison with experimental results is presented in section 5.

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