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Emission properties of a distributed feedback laser cavity containing silicon nanocrystals

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Abstract

We report on light emission from silicon nanocrystals (Si-nc) in a laser cavity. Using modified electrochemical etching of Si wafers we prepare Si-nc with blue-shifted photoluminescence spectrum down to 580–620 nm, embedded at high-volume fractions in a SiO_2-based solid matrix. We insert this active medium into an optically pumped resonator. Since our samples are only partially homogeneous, we cannot use external mirrors in order to achieve optical feedback: we induced optically an internal distributed cavity by intense, spatially periodical excitation. Mode selection was simulated by a simplified theoretical model, based on an approach of multiple reflections. In the framework of the model we discuss the experimentally observed spectral emission changes induced by the distributed cavity.

Keywords: Silicon nanocrystals; Optical gain; Distributed feedback laser cavity

1. Introduction

The two most important steps towards the realization of efficient electrically driven silicon-based laser are light amplification in a Si-based material as well as a positive optical feedback. Both were recently obtained on different types of optically pumped crystalline Si-based materials, emitting in the infrared region [1,2]. Both devices, however, are still far from possible on-chip integration. A different approach uses silicon nanocrystals (Si-nc) in SiO_2-based matrices, where one-passage light amplification in the visible region based on amplified spontaneous emission (ASE) was reported [3–7], but no optical feedback in Si-nc system has been demonstrated up to now. Crucial items for the onset of the stimulated emission (SE) are good optical quality of the sample and the dynamical behavior of radiative and competitive fast nonradiative processes [8–11]. The SE buildup time is given by $\tau_{SE} \sim R^3(\xi)^{-1}$, where R is the size and ξ is the volume fraction of Si-nc [9]. Thus systems with high-volume fractions of Si-nc, showing a blue-shifted emission, seem to be the most favorable candidates for lasing [12].

2. Experimental

We fabricated samples of an active medium on the basis of porous silicon (por-Si) grains, embedded in a sol–gel-based SiO_2 matrix doped with phosphorus [7,13]. The process resulted in a brightly luminescent, well-passivated Si-nc rich (~10 vol%) planar layer of sufficiently good optical quality. The samples exhibit steady-state PL spectrum peaked at ~580–620 nm. Both application of H_2O_2 in different steps of por-Si etching process [7,13] and the presence of phosphorus in SiO_2 matrix contributes to this blue shift [14]. At higher-volume fractions Si-ncs tend to aggregate and to form inhomogeneities (of a size comparable with the emitted wavelength ~1 μm), inducing higher losses due to the scattering of emitted light. This leads to a decrease of the net gain coefficient, poor light directionality and loss of the emission

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coherence on ~μm scale. Use of an external mirror cavity for larger volumes of the active material is then questionable. Therefore, we used in our samples optically induced internal distributed feedback laser (DFL) cavities, built in Bor's configuration [15] (Fig. 1, bottom), where the cavity length is comparable with the emission wavelength. An intense-pulsed excitation beam with homogeneous stripe profile (XeCl excimer, $\lambda_{exc} = 308$ nm, 10 Hz repetition rate, 20 ns pulse duration, beam energy ~90 mJ/pulse), is divided by a holographic grating (3000 lines/mm) into its two first diffraction orders of almost equal intensities under angles α. Both beams are then reflected by mirrors, revolving over the angle δ, and focused under the incident angle $2\theta = 2(\alpha - 2\delta)$ onto the active layer, where they interfere. The linear stripe profile of the excitation beams leads to a grating-like interference pattern (Fig. 1, top) (length L~1 mm, width ~130 μm, with a tunable grating period of $\Lambda = \lambda_{exc}/2 \sin\theta$). The intense periodic illumination in the active nonlinear medium leads to a periodically modulated refractive index n via the third-order nonlinear optical susceptibility $\chi^{(3)}$ [16–19]. For a simple estimation of the reflectance R we assume a step-like modulation $n_2 = n_1 + \gamma I_{exc}$, where $n_1 = \sqrt{1 + \chi^{(1)}}$ is given by the linear optical susceptibility $\chi^{(1)}$, I_{exc} is excitation intensity and γ is proportional to $\chi^{(3)}$. R is then given by a simple Fresnel formula $R = (n_2 - n_1/n_2 + n_1)^2 = r^2$. Such a distributed cavity selects longitudinal modes from the broadband emission of the active medium at tunable $\lambda_{cavity} = 2n\Lambda$ [15]. The emitted signal was detected by a plastic optical fiber, connected to a spectrometer and a CCD camera, cooled with liquid nitrogen to about 100 K. Experiments were done at room temperature.

The above described DFL cavity leads to a laser emission provided that: (i) sufficient optical gain due to SE occurs, (ii) the quality of the cavity is high enough and (iii) the sample is optically sufficiently homogeneous. The crucial parameters affecting the quality of the cavity are (i) the modulation depth of n and (ii) the cavity length L. The modulation depth is driven basically by the magnitude of γ [16–19] (we estimate $\gamma = 10^{-8}\,\mathrm{cm^2/W}$ under our experimental conditions). L depends crucially on both temporal and spatial coherence of the two excitation beams, since the interference pattern can occur only if their path difference is smaller than the excimer laser coherence length l_c (which is of the order of hundreds of μm only). Tuning the

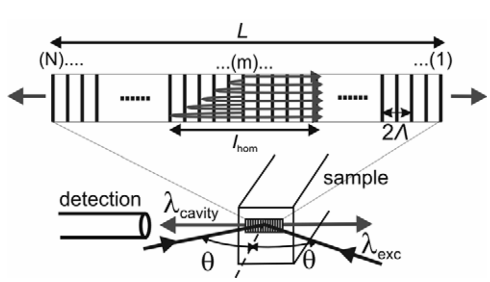

Fig. 1. Top: distributed mirror, used in the model calculation. Amplifying/absorbing material is separated from the mode-selective system. Bottom: DFL experimental setup.

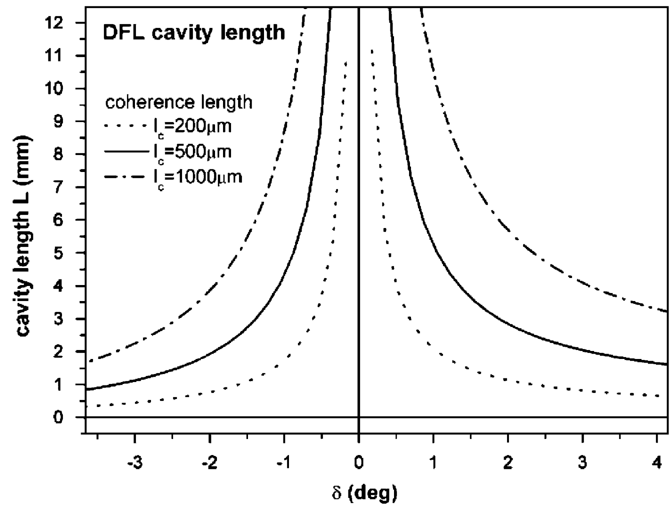

Fig. 2. Length of the interference pattern L as a function of δ for different l_c.

revolving angle δ of the mirrors therefore entails reduction of L with respect to $\delta = 0$, as shown in Fig. 2. This limits severely the accessible range of Λ in our experiments.

3. Model

The main experimental results have been published in Ref. [13]. Here we wish mainly to discuss a model that we have developed to draw conclusions from the experimental data. The model is based on a multiple reflection approach to simulate the mode selection in a partially homogeneous nonlinear active system with an optically induced distributed cavity. The constructive interference leading to the mode selection is supposed to occur only between the waves coupled on a distance equal or less to the mean homogeneity length l_{hom}, which is supposed to be of the order of few μm. Due to the relatively small change of the absorption/gain coefficients, we split our model system into two subsystems: a distributed cavity (Fig. 1, top), selecting the cavity modes, and an absorbing/amplifying material with average net absorption (α)/gain ($g = G - \alpha$) coefficient. The emitting points (Si-ncs) we assume to be distributed along the excitation maxima. The interfering beams come from the reflections on the distributed mirrors only within the short distance $l_{hom} = 2K\Lambda$ (Fig. 1, top) (K be an integer >0) where the light is still in phase, forming an isolated segment with constructive interference. The rest of the material in the cavity with total length $L = N\Lambda$ (N be an integer >0) contributes only by a phase insensitive amplification/absorption of the light. Let $S\Lambda$ (S be an integer >0) be the gain saturation length. The electric field from one segment in the position (m) is for $N \gg S,K$ given by the equation

$$E^{(m,K)} = (t^m e^{-m\Lambda\alpha})E^{(K)},$$

$$E^{(K)} = E_0 e^{K\Lambda\alpha} e^{S\Lambda G} e^{2(K+1)\Lambda g} e^{-iK\varphi} \times \left(1 + rt e^{-i2\varphi} \sum_{j=0}^{K-1} (t^2 e^{-i2\varphi})^j\right),$$

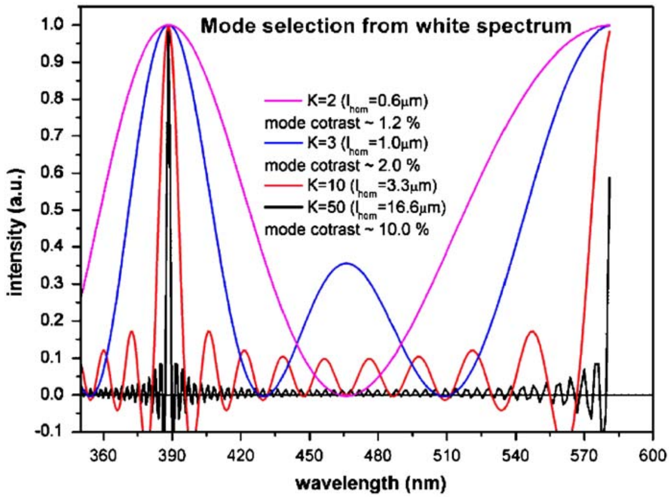

Fig. 3. Mode selection simulation for an ideally white spectrum, calculated for several values of l_{hom}. Spectra are normalized.

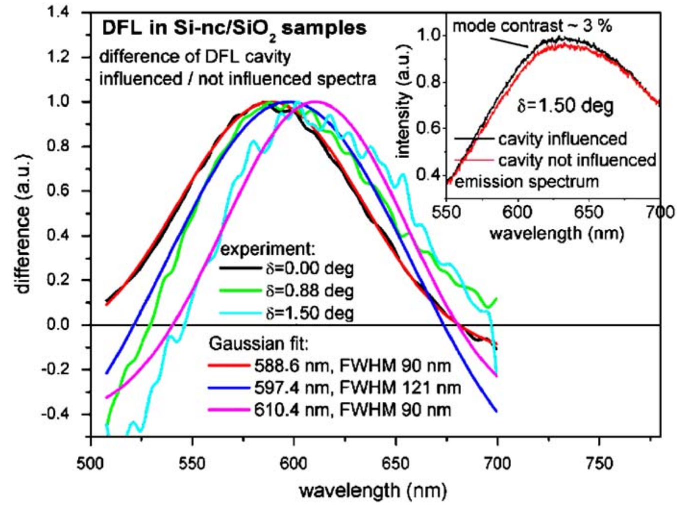

Fig. 4. Experimentally observed DFL modes excited in Si-nc/SiO_2-based material. Curves are fitted by Gaussian shapes.

where E_0 is the emission amplitude of a single segment (excited by I_{exc}), t/r are the transmitivity/reflectivity coefficients and φ is the phase shift. The overall electric field produced by all N segments can be written as $E^{(N,K)} = E^{(K)}\sum_{m=0}^{N}(\mathrm{e}^{-\alpha\Lambda}t)^m$. The intensity $I^{(N,K)}$ is simply given by $I^{(N,K)} = E^{(N,K)*}E^{(N,K)}$. The mode contrast (modulation depth) and the width of the selected modes are mainly given by the number K, i.e. the homogeneity of the material l_{hom} (Fig. 3). Such model gives mode selection from an ideal white spectrum (ideal continuum of emitting states); the real resulting modes would be given by the convolution with the emission (gain) spectra.

4. Results and discussion

The model was already used for simulation of experimental results [13], using parameters taken from measurements of VSL in our type of samples [7], $g{\sim}20\,\mathrm{cm}^{-1}$, $S\Lambda \approx 1\,\mathrm{mm}$ ($S \approx 6000$) and for the reflectance value $R{\sim}5.10^{-5}$ (calculated using $\gamma = 10^{-8}\,\mathrm{cm}^2/\mathrm{W}$ [17,18] and excitation intensity $I_{\mathrm{exc}} = 2.5\,\mathrm{MW/cm}^2$). The total number of the distributed mirrors $N \approx 30000$ was calculated from the length of the interference pattern $L = N\Lambda{\sim}0.5\,\mathrm{cm}$ (equal to the active area length). To demonstrate potential of the method, here we display in Fig. 3 calculated normalized DFL spectra for the homogeneity mean distance $l_{\mathrm{hom}}{\sim}600\,\mathrm{nm}$ ($K = 2$), $1.0\,\mu\mathrm{m}$ ($K = 3$), $1.7\,\mu\mathrm{m}$ ($K = 5$), $3.3\,\mu\mathrm{m}$ ($K = 10$) and $16.6\,\mu\mathrm{m}$ ($K = 50$). The mode contrast is given in percents of the initial intensity and depends for low net gain mainly on l_{hom}.

Optical feedback leads to the increase in the optical path length, which in passive material (no amplification) should reduce the overall intensity of observed spectra, while in the active system ($g>0$) the overall intensity increases due to the light amplification. Because of low-mode contrast in partially homogeneous system, experimentally more available are the differences between spectra influenced/not influenced by the distributed cavity (under the same excitation conditions), which are expected to be positive in active material. Such difference spectra were always positive in our case, indeed, which proves the occurrence of positive gain ($g>0$) in our samples. This is demonstrated in Fig. 4, which displays DFL spectra in a Si-nc/SiO_2 layer as measured for different angles δ, i.e. for different adjusted values of the emission wavelength. It can be seen that the peak maximum undergoes a shift as a function of δ, demonstrating that the Si-ncs "feel" the distributed feedback. However, the spectra show low-mode contrast (around 3%, Fig. 4 inset) and wide FWHM of ~100 nm. These observations are obviously due to the low homogeneity of the sample and our model enables us to estimate l_{hom} to be below $1\,\mu\mathrm{m}$ [13]. The limited space here does not allow us to discuss the comparison with experiment in more details and we leave this point to a separate publication.

5. Conclusion

We have realized an internal optically induced tunable DFL laser cavity, containing an active nonlinear close-packed film of luminescent Si-nc. We have developed a simple model, describing the behavior of partially homogeneous system in good agreement with observed results. We have verified the presence of light amplification, we have proved the influence of the cavity on Si-nc emission and we are able to quantify the homogeneity degree of the samples by comparison with the model. The optical feedback quality must be improved in the future, predominantly by using excitation laser sources with a longer coherence length and assuring stronger nonlinear changes of the index of refraction.

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