Available online at www.sciencedirect.com
SCIENCE DIRECT®
ELSEVIER
Optical Materials 27 (2005) 745–749
OPTICAL Materials
www.elsevier.com/locate/optmat

Optical gain in different silicon nanocrystal systems

P.M. Fauchet [a,*], J. Ruan [a], H. Chen [a], L. Pavesi [b], L. Dal Negro [b,1], M. Cazzaneli [b], R.G. Elliman [c], N. Smith [c], M. Samoc [c], B. Luther-Davies [c]

[a] *Department of Electrical and Computer Engineering, University of Rochester, Rochester, NY 14627, USA*
[b] *University of Trento, Trento, Italy*
[c] *Australian National University, Canberra, Australia*

Available online 13 October 2004

Abstract

An extensive experimental study of optical gain in silicon nanocrystals is under way. Different types of samples have been tested (e.g., nanocrystalline silicon superlattices prepared in Rochester, ion implanted silicon dioxide prepared in Canberra) using different measurement techniques (e.g., variable stripe length method in Trento and in Rochester, prism coupling in Canberra) and different pump laser sources (from femtosecond to cw). All the results presented here have been reproduced in at least two different laboratories, making it unlikely that experimental artifacts play a role. So far, we have observed nsec-duration gain in the visible/near infrared range in nanocrystalline silicon superlattices under high photoinjection conditions with short laser pulses. For other conditions (e.g., lower photoinjection, ion implanted samples with a lower concentration of quantum dots), we have not observed gain.

1. Introduction

The desire for faster computers is pushing the microelectronics industry to increase the integration density and operating frequency of electronic chips. However, this trend is not without limits. The electrical interconnect is one of the major problems we will face within 10 years, not only between chips but also at the single chip level [1]. Certainly finding a solution to this problem would have a huge impact for the semiconductor industry. Replacing electrical interconnects with optical interconnects is one of the promising ways to solve this problem. In optical interconnects there is no distance related loss or distortion of the signal, no deleterious fringing effect and no heat dissipation in the interconnect itself. The possibility of using wavelength division multiplexed networks makes optical interconnects even more attractive [2]. A basic optical interconnect system includes a light source, a detector and the optical path between them. Silicon can already be used for most of the elements of a complete interconnect system. One-dimensional photonic bandgap structures made of a porous silicon microcavity impregnated with liquid crystal molecules form an electrically-addressable mirror [3] and can be useful as an optical switch. An all-silicon modulator capable of GHz speed has also been demonstrated [4]. Detectors made from silicon are also available, including some capable of a very high response speed [5]. However, due to the poor efficiency of silicon based light emitters there are no appropriate silicon light sources.

In 2000 evidence for optical gain was presented in silicon nanocrystals that were produced by implanting silicon in silicon dioxide layers grown on silicon wafers [6]. In that work, ~3 nm silicon nanocrystals luminescent in the near infrared around 1.5 eV were inserted into a crude waveguide structure. The light emitted by the

* Corresponding author. Tel.: +1 585 275 1487; fax: +1 585 275 2073.
E-mail address: fauchet@ece.rochester.edu (P.M. Fauchet).
[1] Present address: Department of Materials Science and Engineering, Materials Processing Center, Massachusetts Institute of Technology.

0925-3467/$ - see front matter
doi:10.1016/j.optmat.2004.08.008

material was amplified as it traveled along the waveguide. Net modal gains between 10 and 100 cm^{-1} were measured using the variable stripe length (VSL) method. Although gain has been independently reported by other groups [7–12], several important questions remain unresolved. It is not clear why no gain is observed in some studies [13–15] or why groups that do observe gain report that only a fraction of the samples yield gain. The differences may be due to the use of different samples or experimental conditions such as measurement setup or pump fluence.

In this paper we present a comparative study done on different types of samples prepared in different groups (e.g. nanocrystalline silicon superlattices prepared in Rochester, ion implanted silicon dioxide prepared in Canberra). Those samples were tested using different measurement techniques and different pumping strategies at different laboratory independently.

2. Sample preparation and experimental setup

Silicon nanocrystalline superlattices were prepared by magnetron sputtering in Rochester. A thick (1 μm) SiO_2 layer was grown on silicon wafers to form the cladding layer for the waveguide structure. Alternating layers of amorphous silicon (a-Si) and SiO_2 were then deposited in the same chamber to form a superlattice (SL) structure. Samples with different layer thicknesses have been grown and tested. However, in all cases, the thickness of both types of layer was kept between 2.5 and 4 nm. The a-Si layers were then transformed into dense arrays of nanocrystals by a procedure that involves high temperature furnace annealing, possibly preceded by a quick rapid thermal annealing step. We have shown that under these conditions, the entire a-Si layer is transformed into densely-packed nanocrystals that form a monodispersed size distribution [16,17]. Indeed, the diameter of the nanocrystals is given by the thickness of the initial a-Si layer. (Thereafter, we will call these the Rochester samples.) Ion implanted samples were prepared in Canberra. Waveguide structures were formed by implanting Si at an accelerating voltage of several hundred eVs into 5 μm thick SiO_2 layers grown on (1 0 0) Si wafers by high temperature, high pressure oxidation. After annealing at 1100 °C for one hour in a flowing N_2 ambient nanocrystals were formed with the size distribution between 3 and 5 nm with a peak nanocrystal density of around 5×10^{18} cm^{-3} [13]. (Thereafter, we will call these the Canberra samples.)

Studies of optical gain were conducted on all samples using the variable stripe length (VSL) method [18]. Normal PL and VSL measurements on the Rochester samples were performed using two different setups in two different laboratories (Trento [7] and Rochester [19]). In Trento, continuous wave measurements were performed using a UV enhanced argon ion laser (365 nm) and a double monochromator with a photomultiplier tube (PMT) for photon detection. The time resolved measurements were performed using high fluence short optical pulses (6 ns, 10 Hz, 355 nm) produced by the third harmonic of a Nd:YAG laser and a single grating spectrometer with a picosecond visible streak camera for photon detection. In Rochester, an amplified frequency doubled Ti-sapphire system with ultrashort pulses (~200 fs, 1 kHz, 405 nm) was used and the signal was analyzed by a scanning monochromator equipped with a PMT and a gated photon counter. Measurements on the Canberra samples also employed a pump–probe technique in which the probe beam propagates in a waveguide structure containing the silicon nanocrystals. For these pump–probe measurements, a probe beam (~150 fs, 250 Hz, 755 nm) was coupled into the nanocrystal waveguide using a prism coupler while the pump, produced using the frequency doubled light from the same laser beam, illuminated the top of the waveguide. Detection was performed by a Si photodiode.

3. Experimental

3.1. VSL results on the Rochester samples

The normal incidence PL from the samples tested for gain increases linearly with pump power in the range of interest and its spectrum remains unchanged. These results indicate that Auger recombination does not play an important role and confirm that our size distribution is very narrow [20].

Fig. 1 shows results obtained on a sample that did exhibit gain under cw mode-locked pumping. When the pump intensity is raised from 10 W/cm^2 to 1 kW/cm^2

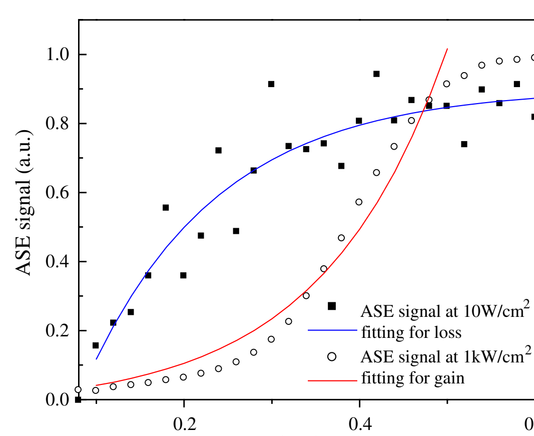

Fig. 1. Amplified spontaneous emission (ASE) measured under CW mode locked pumping on a sample at two different pump intensities. The solid squares show loss for low pump intensity and the open circles show gain at high pump intensity. The signal was measured at 800 nm.

(corresponding to a fluence of 0.12 μJ/cm^2 to 12 μJ/cm^2 per pulse), we observe a switch from a loss of ∼20 cm^{-1} to a gain of ∼100 cm^{-1}. These results strongly suggest that under intense pumping, it is possible to produce enough gain to overcome the waveguide losses. The magnitude of the gain is greater than in most results obtained using other samples and we attribute this enhancement to the greater density of nanocrystals in the Rochester samples. When a similar sample was tested in Trento using cw pumping, no gain was observed at the maximum pumping intensity [11]. These combined results suggest that intense pulsed pumping is required to overcome the waveguide losses.

The results from time-resolved studies performed in both Trento and Rochester confirm this conclusion. In addition to the slow (μs) emission component characteristic of nc-Si systems, we measured a fast (ns) emission component when either the pump fluence was increased for a fixed pumping length or when the pumping length was increased at a fixed pump fluence. Fig. 2 shows time-resolved results obtained by varying the pumping length at a fixed pump fluence (550 mJ/cm^2). As the pumping length increases, a very clear fast emission peak with a full width half maximum (FWHM) of about 10 ns, comparable to the pump duration, appears on top of the slow emission background. We interpret this effect as due to the appearance of stimulated emission. This result, obtained in Trento under ns optical pumping, was confirmed in Rochester using fs pumping. Results of the fast and slow components measured in Rochester are shown in the inset of Fig. 2. Only loss was measured with the slow component whereas gain could be measured with the fast component. (The signals are normalized since the intensity of the fast component is much higher than that of the slow component.) The data were recorded with an observation time window of 10 ns in the case of the fast component and 2 μs in the case of the slow component. (The time resolution of the detection system is in the nsec time regime). Since nothing except the observation time window was changed, the potential artifacts of the VSL technique (e.g., finite numerical aperture of the collecting optics or total internal reflection at the output surface [15,21]) cannot explain the results under high pulsed pumping conditions. Fig. 3 summarizes typical VSL results using the setup in Trento. Fitting yields a gain value of 50 cm^{-1} at the maximum fluence. In this case, switching from loss to gain was achieved by simply increasing the pump fluence as shown in the inset of Fig. 3. This behavior was also qualitatively reproduced in Rochester. In Fig. 4 we plot the peak intensity of this fast component vs pump fluence. The sublinear behavior in the low pump fluence region changed abruptly to superlinear behavior beyond a threshold

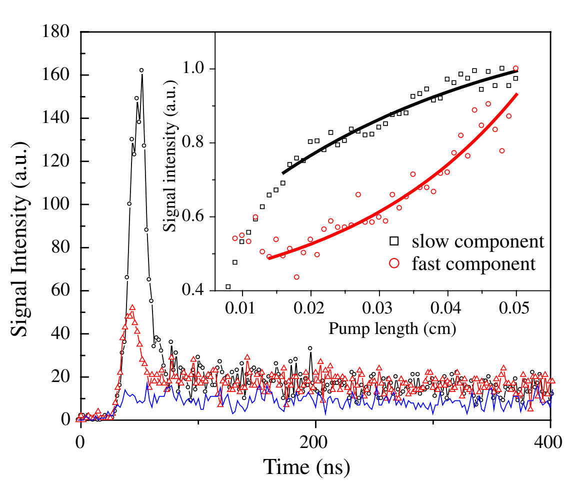

Fig. 2. Time resolved results obtained using the VSL configuration with a pump fluence of 550 mJ/cm^2 and ns optical pulses. The solid line, triangles and circles correspond to a pumping length of 0.3 mm, 0.5 mm and 1 mm respectively. Inset shows normalized VSL results on the fast component (circles) and slow component (squares) using fsec optical pulses. Fitting (line) yields a gain of 36 cm^{-1} for the fast component and a loss of 27 cm^{-1} for the slow component.

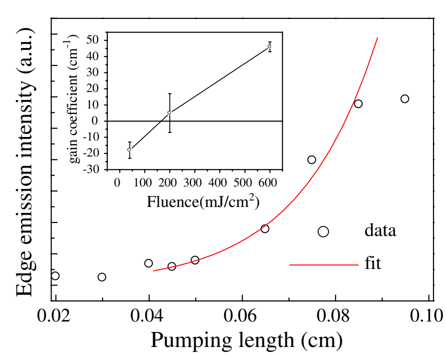

Fig. 3. Typical VSL results (circles) with a pump fluence of 550 mJ/cm^2 and ns optical pulses. Fitting (line) yields a gain of 50 cm^{-1}. The result of fitting the data at three fluences plotted in the inset shows the transition from loss to gain.

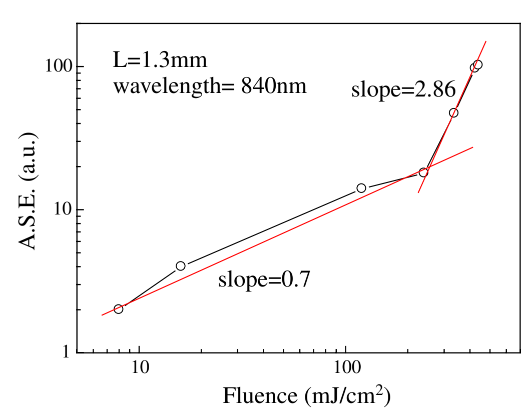

Fig. 4. The peak intensity of the fast component is plotted vs pump fluence The solid lines are power-law fitting with a slope of 0.7 and 2.86 for low fluence and high fluence part respectively. A clear threshold is observed near 250 mJ/cm^2.

value, as expected when stimulated emission dominates over Auger recombination.

The results obtained independently in the two different laboratories on the Rochester samples are consistent. They show that under intense pulsed pumping it is possible to produce enough gain to overcome the waveguide loss, especially in the visible/near infrared range. In addition, the fast (ns) optical gain can compete with the non-radiative Auger recombination that is usually invoked to rule out the possibility of population inversion (and optical gain) in nc-Si [20].

3.2. Pump probe and VSL measurement on the Canberra samples

Results from the pump–probe measurements on a Canberra sample are plotted in the inset of Fig. 5, where the time response of the probe beam is shown following excitation with pump pulses of increasing fluence. The probe beam is attenuated instead of amplified by the pump beam. The magnitude of the attenuation increases with increasing pump fluence and eventually saturates as shown in Fig. 5. Based on the excited state carrier absorption model proposed in Ref. [13], a reasonable fit to the data is achieved as shown in Fig. 5. The fitted value for the excited state absorption coefficient is about $4.6 \times 10^{18}\text{cm}^2$ in this case. This value is consistent with the free carrier absorption cross-section in bulk silicon [22].

A similar sample was tested in Rochester using the VSL technique and fsec pulses. We observed no fast component in the response, only the normal slow component. As shown in the inset of Fig. 6 the typical VSL measurement indicates loss, not gain, consistent with what we observe for the slow component in the Rochester samples. In Fig. 6 the fitted loss value from the VSL measurement is plotted vs. pump fluence. The loss increases and eventually tends to saturate with increasing pump fluence. This behavior is consistent with the results of the previous pump probe measurement, although the values of the fluence differ by one order of magnitude.

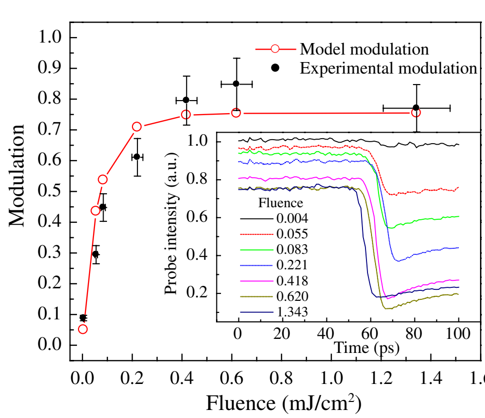

Fig. 5. The modulation of the probe beam in a pump probe experiment is plotted vs pump fluence. The solid line is a fit to the data using a simple excited state absorption model (Ref. [13]). The inset shows the probe signal measured at the exit from the waveguide as a function of time for different pump fluences.

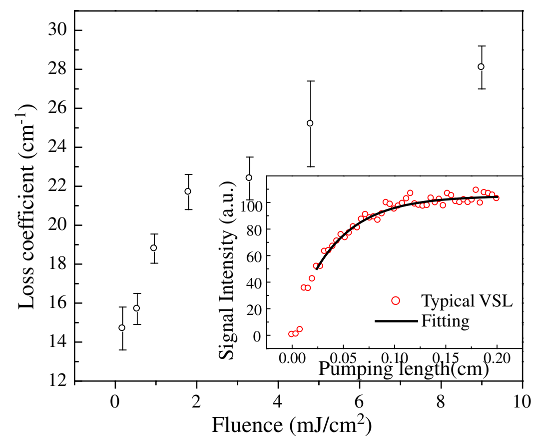

Fig. 6. Loss coefficients measured from VSL measurements plotted vs pump fluence. The inset shows a typical VSL curve and a fit with a loss of $24\,\text{cm}^{-1}$.

3.3. Discussion

In this paper, we have shown experiment data obtained using different setups in different laboratories. With the Rochester samples we showed that it is possible to produce enough gain to overcome the losses in the VSL method under intense pulsed pumping. The use of different experimental setups and the time-resolved experiments themselves ruled out any possible artifacts associated with the VSL method. The fluences used to achieve gain with the nsec pump pulses in Trento were approximately one order of magnitude greater than those used with the fsec pump pulses in Rochester. This difference may indicate that recombination channels exist with a time constant $\leqslant 1$ nsec. With the Canberra samples pump-probe experiments have been done in addition to VSL experiments. In both experiments, only loss was measured. Note that the fluence used in Rochester for the Canberra samples was comparable to the fluence used on the Rochester samples exhibiting gain. One of the possible reasons for the lack of gain in the Canberra samples is the lower concentration of quantum dots. More detailed conclusions will require more extensive comparative studies.

4. Conclusions

In conclusion, two different types of samples were measured independently in different laboratories using different techniques and different pump sources. The results are consistent for both samples. Gain with ns-duration in the visible/near infrared range was observed from the Rochester samples under high photoninjection conditions with short laser pulses. No gain was observed in the Canberra samples. The different results may be related to the different density of nanocrystals in the two types of samples.

This research was supported in part by the Semiconductor Research Corporation (Contract 2002-MC-995) and National Science Foundation (Grant ECS-0112612).

References

[1] International Technology Roadmap for Semiconductors 2001, <http://www.itrs.com>.
[2] M.J. Kobrinsky, B.A. Block, J.-F. Zheng, B.C. Barnett, E. Mohammed, M. Reshotko, F. Robertson, S. List, I. Young, K. Cadien, Intel Technol. J. 8 (2004) 129.
[3] S.M. Weiss, P.M. Fauchet, Phys. Stat. Sol. A 197 (2003) 556.
[4] A. Liu, R. Jones, L. Liao, D. Samara-Rubio, D. Rubin, O. Choen, R. Nicolaescu, M. Paniccia, Nature 427 (2004) 615.
[5] S. Alexandrou, C.-C. Wang, T.Y. Hsiang, M.Y. Liu, S.Y. Chou, Appl. Phys. Lett. 62 (1993) 2507.
[6] L. Pavesi, L. Dal Negro, C. Mazzoleni, G. Franzò, F. Priolo, Nature 408 (2000) 440.
[7] L. Dal Negro, M. Cazzanelli, N. Daldosso, Z. Gaburro, L. Pavesi, F. Priolo, D. Pacifici, G. Franzò, F. Iacona, Physica E 16 (2003) 297.
[8] L. Dal Negro, M. Cazzanelli, L. Pavesi, S. Ossicini, D. Pacifici, G. Franzò, F. Priolo, F. Iacona, Appl. Phys. Lett. 82 (2003) 4636.
[9] L. Khriachtchev, M. Rasanen, S. Novikov, J. Sinkknen, Appl. Phys. Lett. 79 (2001) 1249.
[10] M. Nayfeh, S. Rao, N. Barry, A. Smith, S. Chaieb, Appl. Phys. Lett. 80 (2002) 121.
[11] J. Ruan, P.M. Fauchet, L. Dal Negro, M. Cazzanelli, L. Pavesi, Appl. Phys. Lett. 83 (2003) 5479.
[12] K. Luterova, K. Dohnalová, V. Svcrek, I. Pelant, J.-P. Likforman, O. Crégut, P. Gilliot, B. Honerlage, Appl. Phys. Lett. 84 (2004) 3280.
[13] R.G. Elliman, M.J. Lederer, N. Smith, B. Luther-Davies, Nucl. Instr. and Meth. B 206 (2003) 427.
[14] L. Khriachtchev, M. Rasanen, S. Novikov, Appl. Phys. Lett. 83 (2003) 3018.
[15] J. Valenta, I. Pelant, J. Linnros, Appl. Phys. Lett. 81 (2002) 1396.
[16] L. Tsybeskov, K.D. Hirschman, S.P. Duttagupta, M. Zacharias, P.M. Fauchet, J.P. McCaffrey, D.J. Lockwood, Appl. Phys. Lett. 72 (1998) 43.
[17] G.F. Grom, D.J. Lockwood, J.P. McCaffrey, H.J. Labbé, P.M. Fauchet, B. White, J. Diener, D. Kovalev, F. Koch, L. Tsybeskov, Nature 407 (2000) 358.
[18] K.L. Shaklee, R.E. Nahory, R.F. Leheny, J. Lumin. 7 (1973) 284.
[19] P.M. Fauchet, J. Ruan, Towards the first silicon laser, in: L. Pavesi, S. Gaponenko, L. Dal Negro (Eds.), NATO Science Series, 93, Kluwer Academic Publishers, Dordrecht, 2003, p. 197.
[20] D. Kovalev, H. Kheckler, G. Polisski, F. Koch, Phys. Stat. Sol. B 215 (1999) 871.
[21] L. Dal Negro, P. Bettotti, M. Cazzanelli, D. Pacifici, L. Pavesi, Optics Commun. 299 (2004) 337.
[22] J.M. Moison, F. Barthe, M. Bensoussan, Phys. Rev. B 27 (1983) 3611.