Waveguiding effects in the measurement of optical gain in a layer of Si nanocrystals

J. Valenta, I. Pelant, and J. Linnros

Citation: Appl. Phys. Lett. **81**, 1396 (2002); doi: 10.1063/1.1502195
View online: http://dx.doi.org/10.1063/1.1502195
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v81/i8
Published by the American Institute of Physics.

Waveguiding effects in the measurement of optical gain in a layer of Si nanocrystals

J. Valenta[a)]
Department of Chemical Physics & Optics, Faculty of Mathematics & Physics, Charles University, Ke Karlovu 3, CZ-121 16 Prague 2, Czech Republic

I. Pelant
Institute of Physics, Academy of Sciences of the Czech Republic, Cukrovarnická 10, CZ-162 53 Prague 6, Czech Republic

J. Linnros
Department of Microelectronics & Information Technology, Royal Institute of Technology, Electrum 229, S-164 40 Stockholm-Kista, Sweden

(Received 7 June 2002; accepted for publication 28 June 2002)

We discuss applicability of the variable stripe length method to experimental investigation of optical gain in a luminescent layer that behaves like a planar waveguide. We show that an interplay between the output direction of guided light modes and the numerical aperture of the collection optics may lead to an artifact manifesting itself as an apparent but false gain. We propose a way to circumvent this inconvenience by using a ''shifting excitation spot'' complementary measurement. The method is demonstrated on a layer of Si nanocrystals embedded into a synthetic silica plate. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1502195]

The recent observation of optical gain in Si nanostructures by Pavesi *et al.*[1] attracted wide attention from the scientific community. The reported modal optical gain of the order of 100 cm^{-1} should be enough to construct the first silicon laser, which could cause a revolution in the field of optoelectronics. Later, another two reports on gain in Si nanocrystals (NCs) followed. While experiments of Nayfeh and co-workers[2] were performed on specific small Si NCs, the observation by Khriachtchev *et al.*[3] was done in samples similar to those used by Pavesi's group. The main experimental method used by both Pavesi and Khriachtchev was the so called variable stripe length (VSL) method introduced by Shaklee *et al.* in 1971,[4] which has been subsequently widely applied to measure optical gain in semiconductors without being thoroughly tested.

The aim of this work is to check the applicability of the VSL method for measurement of gain in NCs formed in Si ion implanted SiO_2. We show that waveguiding that occurs in the active medium—a layer composed of Si NCs—can significantly modify not only the spontaneous emission spectral shape but also the observation of gain. It is even possible to find out, by using the VSL method, an artifact: significant gain values without genuine amplification taking place in the sample. We propose a simple modification of the VSL method that enables to correct a possible inaccuracy in determining the value of gain.

The layers of Si NCs were prepared by Si ion implantation into synthetic silica substrates using an implantation energy of 400 keV and total doses $1-6\times10^{17}$ cm^{-2}. Subsequent annealing at 1100 °C in N_2 atmosphere for 1 h lead to formation of Si NCs in a thin layer parallel to the silica surface.[5] Photoluminescence (PL) excited by the 325 nm line of a He-Cd laser shows a typical wide band in the red spectral region 650–900 nm[6,7] [see Fig. 1(a), curve A]. This ordinary PL measurement is performed by exciting and observing the front face of the implanted plane. However, when detecting PL from the facet of the implanted layer we observe huge changes of spectral shape, in particular for samples implanted with high doses 4 and 6×10^{17} cm^{-2} [Fig. 1(a), curve B]. The main features are the TE and TM modes guided by the nanocrystalline layer forming a planar waveguide. The modes can be distinguished using a polarizer [see Fig. 1(a), curves C, D]. The facets of samples were polished to a good optical quality, but the microscope image [Fig. 1(b)] shows that there are still many imperfections (due to fragility of the SiO_2 facet).

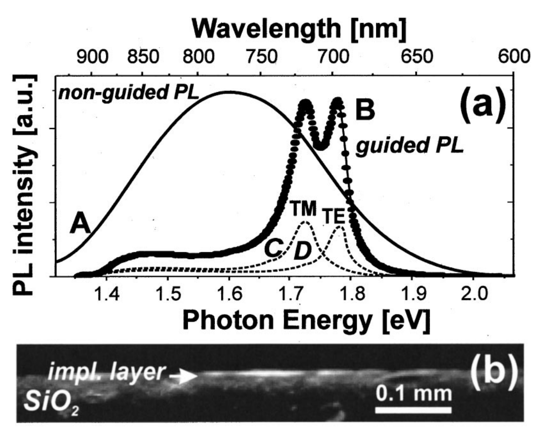

FIG. 1. (a) PL spectra of a sample implanted with dose of 4×10^{17} cm^{-2} ($\lambda_{exc}=325$ nm and $I_{exc}=0.26$ W/cm^2). Curves A and B were detected in directions perpendicular (nonguided) and parallel (guided) to the implanted plane, respectively. Dashed lines C and D are the polarization resolved TM and TE modes, respectively. (b) Microscope image (0.65×0.1 mm) of the PL emerging (in a direction perpendicular to the plane of the figure) from the implanted layer. The facet of sample is slightly illuminated by a halogen lamp.

[a)]Electronic mail: jan.valenta@mff.cuni.cz

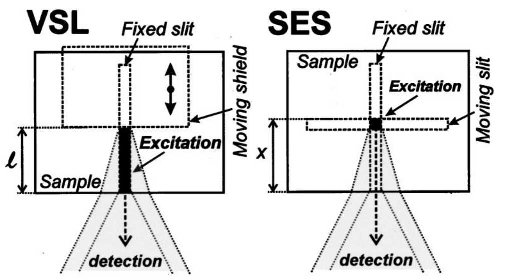

FIG. 2. The experimental setup for the VSL and SES measurements.

Let us briefly summarize principles of the VSL method.[4] The excitation light beam is focused (usually by a cylindrical lens) into a rectangular stripe, which starts at the facet of a sample and whose length can be varied by shading a part of the rectangle with a moving shield (a blade). The detection optics collects light from the facet of a sample (perpendicular to the incoming beam). If a population inversion is reached the excited stripe of sample acts as a single-pass amplifier for PL photons traveling inside the stripe. The net gain is defined as a relative change of the light intensity passing an infinitesimal distance dx along the stripe. The increment of total change of detected intensity dI_{tot} when increasing the stripe length x by dx is a sum of a gain amplification of the incoming light and of the spontaneous emission $I_{\mathrm{sp}}(\lambda)$ from stripe of length dx. Solving an ordinary differential equation for the total stripe length ℓ we retrieve the classical VSL equation

$$I_{\mathrm{tot}}(\ell,\lambda)=\frac{I_{\mathrm{sp}}(\lambda)}{G(\lambda)}\{\exp[G(\lambda)\cdot\ell]-1\}, \qquad (1)$$

where the net optical gain $G(\lambda)=[g(\lambda)\text{-}K]$. K represents losses and $g(\lambda)$ is a gain coefficient (negative absorption coefficient). Here, it is commonly accepted that $I_{\mathrm{sp}}(\lambda)$ and $g(\lambda)$ are independent of x and, more importantly, that the coupling of the output emission to a detector is constant, independent of x.

In order to verify this assumption, we performed the VSL measurement using excitation by the 325 nm line of a He-Cd laser (cw, max. power 10 mW). The sample was excited through a fixed metal slit (width of 25 or 100 μm) placed close to the sample surface (20–50 μm). The resulting stripe length was varied by moving a metal foil over the slit with a microstepping motor (see Fig. 2). The signal was collected by microscope objectives, focused on the input slit of an imaging spectrograph, and detected by a CCD camera (for setup details see Ref. 8). This arrangement ensured the constant position of the excitation spot on the sample surface, negligible influence of diffraction, and perfect control over the experiment geometry.

Figure 3(a) shows the stripe-length dependence of PL detected at wavelengths of the maxima of TE and TM modes and at 825 nm (nonguided PL). The stripe width was 100 μm, excitation intensity 0.26 W/cm^2 and detection numerical aperture (NA)$=$0.13. For guided modes we see an exponential increase of signal for stripe length $<$0.45 mm. Fitting by Eq. (1) (solid lines) yields G about 30 cm^{-1}. Nonguided PL around 825 nm gives just losses of 11 cm^{-1}. Apparent conclusion thus could be the presence of optical gain in guided modes.

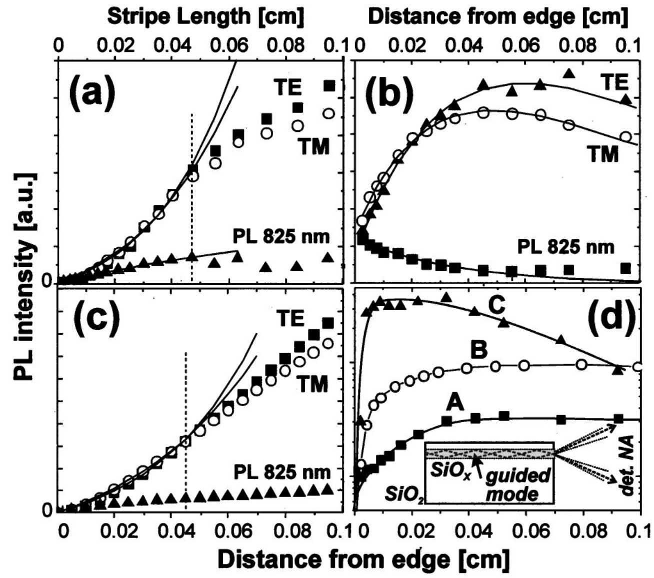

FIG. 3. (a) Results of the VSL measurement at the peak of TE and TM modes and for nonguided PL around 825 nm. The fits (lines) give values of $G=35$ and 28 cm^{-1} for TE and TM modes, respectively, and losses of 11 cm^{-1} for nonguided PL. (b) Results of the SES measurement performed under identical conditions as the VSL. The lines are fits using Eqs. (3) and (2) for TE (TM) and PL, respectively. (c) Integration of data from panel (b). The gain fits (lines) give values of $G=29$ and 22 cm^{-1} for TE and TM modes, respectively. (d) The SES measurements using detection NA of 0.075 (A-black squares), 0.13 (B-white dots), and 0.3 (C-black triangles). Lines are guides to the eye. The inset shows interplay between the exit angle of the guided mode and the detection NA.

Now, we wish to check the validity of the assumption that the signal from any infinitesimal part dx of the excited stripe is equally coupled to the detection system. For this we propose a simple modification of the VSL arrangement which we call "shifting excitation spot" (SES) measurement. The moving shield is replaced by a moving slit perpendicular to the fixed slit (see Fig. 2). Consequently, only a tiny rectangular area of the sample is excited. This excited segment is moved by changing the distance x from the edge of the sample and relevant changes of the detected signal are observed. If the abovementioned assumption is correct, we should observe only a slow exponential decrease of the signal (due to losses, because light amplification is precluded owing to dividing the excited stripe into small segments). With increasing distance x of the segment from the sample edge, we therefore expect

$$dI_{\mathrm{SES}}(x,\lambda)=I_{\mathrm{sp}}(\lambda)\exp\{-[\alpha(\lambda)+K]x\}dx, \qquad (2)$$

where $\alpha(\lambda)$ is a residual attenuation.

Results of the SES measurement for TE and TM modes, along with PL at 825 nm, are plotted in Fig. 3(b). The experiment was performed at the same part of the sample and under identical conditions as for the above described VSL measurement (excited segment 25×100 μm). It turns out that the Eq. (2) holds for the nonguided PL signal only where a loss of 29 cm^{-1} was found [lower curve in Fig. 3(b)]. The signal for the TE and TM modes shows a surprising *increase* for x up to about 0.5 mm. The observed dependence can be fitted, instead of Eq. (2), by a relation

$$I_{\mathrm{SES}}(x,\lambda)=a+bx\cdot\exp[-cx]. \qquad (3)$$

There is a constant term and a term combining a linear increase with an exponential decay. The linear and exponential terms dominate for short and large x, respectively.

If there is no amplification of PL by stimulated emission, the result of the VSL measurement must be equal to the integral of the SES dependence from $x=0$ to ℓ. The integral of the SES measurement presented in Fig. 3(b) is plotted in Fig. 3(c). One can see that the initial parts of curves for TE and TM modes have a superlinear shape, which can be fitted well with the VSL Eq. (1). Gain values of 29 and 22 cm^{-1} were found for TE and TM modes, respectively, almost identical with results shown in Fig. 3(a). This finding demonstrates that the gain-like behavior in Fig. 3(a) is a pure consequence of a nonconstant coupling of guided PL to the detection system, not a true gain.

The main reason for the observed peculiar SES dependence is the interplay between the output direction of the guided modes and the NA of the detection optics [see inset in Fig. 3(d)]. To illustrate this fact we performed the SES measurement using objectives with different numerical apertures of 0.075, 0.13, and 0.3. Results are plotted in Fig. 3(d). A gradual increase in the SES signal for x up to 0.5 mm is observed with NA of 0.075 and 0.13, but NA=0.3 gives a constant signal already at $x \sim 0.01$ mm.

In conclusion, our experiments demonstrate that the VSL method cannot be used in its classical form when a waveguiding effect takes place in the studied sample. In such a case, we have to introduce a function $\beta(\mathrm{x})$ describing the coupling of emission from an excited segment to the detection. Thus, Eq. (1) should be replaced by a general integral equation:

$$I_{\mathrm{tot}}(x)=\exp\left[G(\lambda)\int \beta(x)\,dx\right]I_{\mathrm{sp}} \times \int \beta(x)\exp\left[-G(\lambda)\int \beta(x)\,dx\right]dx. \tag{4}$$

When $\beta(x)$ is constant we retrieve the VSL Eq. (1). The coupling function $\beta(x)$ can be determined from the SES measurement that obviously yields

$$I_{\mathrm{SES}}(x,\lambda)=I_{\mathrm{sp}}(\lambda)\exp\{-[\alpha(\lambda)+K]x\}\beta(x)\,dx. \tag{5}$$

Therefore, the correct determination of G should be done as follows. Perform the VSL and the SES measurement under identical conditions at the same place of a sample. If the SES signal $I_{\mathrm{SES}}(x,\lambda)$ increases with x, a complicated coupling of the PL signal to the detection takes place (due to, e.g., waveguiding). Then either the experimental condition should be modified [with aim to have $\beta(x)$ constant] or the function $\beta(x)$ has to be determined from the SES measurement [by comparing Eqs. (3) and (5)] and the Eq. (4) solved (numerically) to find $G(\lambda)$. For our samples the real net gain $G(\lambda)$ turns out to be close to zero under excitation by a cw laser, $\lambda_{\mathrm{exc}}=325$ nm, and pump intensity up to 1 $\mathrm{W/cm^2}$.

Our observations, however, do not deny a potential presence of optical gain in a layer of Si NCs made by Si ion implantation (the presence of gain is strongly dependent on the mode of applied pumping as well as on the sample properties). But the reported values of optical gain (or its presence itself) should be confirmed by the combination of the VSL and SES measurement in order to exclude the influence of waveguiding phenomena. Similar effects cannot be *a priori* excluded in any material containing layers of different index of refraction (e.g., layers of self-organized III–V nanocrystals[9]).

Finally, we have to note that other phenomena like the total internal reflection (when using a wide stripe in the VSL measurement) or a confocal effect (when using high NA detection optics) can also affect the gain measurement. Here we can state that these effects should be at least partially corrected using our SES measurement. What is, however, not possible to correct is the suppression of gain by photons going transversally to the stripe direction (in case of wide stripe).

We are grateful to Dr. Cheylan (Australian National University, Canberra) for fabrication of high quality samples. This work was supported by GAAV CR (Grant Nos. A1010809 and AVOZ 1010914) and NATO (Grant No. PST.CLG.978100).

[1] L. Pavesi, L. Dal Negro, C. Mazzoleni, G. Franzo, and F. Priolo, Nature (London) **408**, 440 (2000).
[2] M. H. Nayfeh, N. Barry, J. Therrien, O. Akcakir, E. Gratton, and G. Belomoin, Appl. Phys. Lett. **78**, 1131 (2001).
[3] L. Khriachtchev, M. Rio, S. Novikov, and J. Sinkkonen, Appl. Phys. Lett. **79**, 1249 (2001).
[4] K. L. Shaklee, R. F. Leheny, and R. E. Nahory, Phys. Rev. Lett. **26**, 888 (1971).
[5] R. G. Elliman, M. J. Lederer, and B. Luther-Davis, Appl. Phys. Lett. **80**, 1325 (2002).
[6] S. Guha, J. Appl. Phys. **84**, 5210 (1998).
[7] J. Linnros, N. Lalic, A. Galeckas, and V. Grivickas, J. Appl. Phys. **86**, 6128 (1999).
[8] J. Valenta, R. Juhasz, and J. Linnros, Appl. Phys. Lett. **80**, 1070 (2002).
[9] J. D. Thomson, H. D. Summers, P. J. Hulyer, P. M. Smowton, and P. Blood, Appl. Phys. Lett. **75**, 2527 (1999).