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C. J. Oton[1], **D. Navarro-Urrios**[1, 2], **M. Ghulinyan**[2], **N. E. Capuj**[1], **S. González-Pérez**[3], **F. Lahoz**[3], **I. R. Martín**[3], and **L. Pavesi**[2]

[1] Dpto. de Física Básica, University of La Laguna, Tenerife, Spain
[2] Dipartimento di Fisica, University of Trento, via Sommarive 14, 38050 Povo, Italy
[3] Dpto. de Física Fund. y Exp. Electr. y Sist., University of La Laguna, Tenerife, Spain

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C. J. Oton[*, 1], **D. Navarro-Urrios**[1, 2], **M. Ghulinyan**[2], **N. E. Capuj**[1], **S. González-Pérez**[3], **F. Lahoz**[3], **I. R. Martín**[3], and **L. Pavesi**[2]

[1] Dpto. de Física Básica, University of La Laguna, Tenerife, Spain
[2] Dipartimento di Fisica, University of Trento, via Sommarive 14, 38050 Povo, Italy
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1 Introduction

Porous silicon (PS) shows very interesting optical properties due to the nanometric size of its pores. Particularly, since the report in 1990 of efficient luminescence at room temperature [1], it has been very actively investigated, because an efficient light source from a silicon-compatible material would have a considerable impact in photonics technology. There are some recent works that report optical gain in PS that have increased its interest even more [2, 3]. Another advantage of this material is the possibility of varying the porosity in depth with an arbitrary profile, allowing the fabrication of 1D photonic structures [4].

Porous silicon can be oxidized if annealed at high temperatures. If the temperature is higher than 800 ºC and the duration of the annealing is long enough, all the silicon can be converted to silica, completely losing its light emission properties, but maintaining its porosity variations. In addition, the quality of the SiO_2 that is formed after a thermal oxidation makes the material a very appealing porous host for the impregnation of different substances. Previous works report dye impregnation of this material [5–9]. However, laser action has not still been demonstrated. In this work, we report optical gain in dye-impregnated porous silicon-based planar waveguides. This paper includes a study of the power-dependent guided photoluminescence, together with variable stripe length (VSL) gain measurements at different powers and wavelengths, and a discussion of the results.

2 Sample preparation

Porous silicon samples were fabricated by electrochemical etching of (100)-oriented heavily doped (0.01 Ωcm resistivity) p-type silicon. The electrolyte was made mixing 31% of aqueous HF (48 wt.%)

[*] Corresponding author: e-mail: cxn@orc.soton.ac.uk; present address: Optoelectronics Research Centre, University of Southampton, UK.

with ethanol. Two-layer planar waveguides were grown (a core and a cladding) by applying current densities of 40 and 80 mA/cm^2 respectively to a 1cm^2 circular area. The waveguides were designed to be single-mode, one transverse electric (TE) and one transverse magnetic (TM), so the core nominal thickness was 500 nm, and the cladding was 10 µm. The samples were annealed at 900 ºC in air for 3 hours in order to completely oxidize the silicon, and convert the material into porous silica. The refractive indices of the core and cladding decreased from 1.75 and 1.36 down to 1.25 and 1.15 respectively (we report a detailed characterization of these waveguides in Ref. [10]). The samples were subsequently impregnated with Nile Blue (LC 6900) [11] by immersing them in an ethanoic dissolution. The dye concentration was varied between 10^{-6} and 10^{-4} M, and in all cases the samples were immersed and after one minute, taken out and let dry. Finally the samples were cleaved along one side to allow measuring the guided light.
The waveguide modes were characterized on a prism-coupling setup at 633 nm. Only one dark *m*-line peak was observed for each polarization, confirming that the samples were single-mode. Before and after the impregnation the position of the peaks did not change, but they became wider due to an absorption increase, confirming that the core contained an appreciable concentration of Nile Blue.

3 Experiments

A frequency-doubled Nd:YAG laser in Q-switching regime was used as the pump source. 5ns-long pulses at a wavelength of 532 nm were focused on the sample through a cylindrical lens. The spot size was 3 cm long and 300 micron wide, which gave a total area of ~10 mm^2. The maximum pump energy employed was 3 mJ (30 mJ/cm^2). A metal blade was placed in front of the sample in order to vary the illumination length. The guided light was collected with a lens of 10 cm focal length. Half of the lens was blocked to avoid collecting non-guided light. The light was introduced in a 25 cm focal monochromator, and detected with a photomultiplier tube (PMT). An oscilloscope collected each single pulse from the PMT, and transmitted its profile to the computer, where the pulse area was calculated. The guided photoluminescence spectra were collected by shooting one pulse per wavelength, with a spectral resolution of 1 nm.
Gain measurements were performed with the variable stripe length method (VSL) [12], which consists in collecting the guided amplified spontaneous emission (ASE) while varying the illumination length. Assuming a one-dimensional amplifier with a net modal gain *g*, the dependence of the signal versus illumination length *L* is given by the following equation:

$$I_{ASE}(L) \propto \frac{1}{g}\left(e^{gL} - 1\right) \tag{1}$$

where the value of *g* has to increase from negative to positive values when the pump power is increased.
The distance between the sample and the blade was set to 5 mm in order to minimize diffraction effects, and the collection was realized with a lens instead of a microscope objective to avoid confocal effects [13]. We also performed "shifting of excitation spot" measurements (SES) to characterize the losses and check the reliability of the VSL results [14]. This technique consists in scanning a pump spot through the sample, and the guided luminescence should show an exponential decay which gives the propagation losses. This value should correspond to the negative gain measured with the VSL technique at very low powers. For this experiment, the blade was substituted with a 50 µm slit and the measurement was performed in the same way. All the measurements at fixed wavelength were performed with 5 nm spectral resolution.

4 Results

We first characterized the guided luminescence of samples impregnated with different concentrations of Nile Blue. All the sample length (~8mm) was illuminated for various pump powers. The criterion employed to recognise pump-induced effects was the observation of a lineshape modification at high pump power. No change was observed for the samples impregnated with a dye concentration of 10^{-6} M and

10^{-5} M. However, the sample impregnated in a 10^{-4} M solution showed a strong line narrowing in the emission at high pump power. Figure 1 shows the guided PL spectrum of this sample for three different powers. For 1.5, 2.8 and 17 mJ/cm^2, the full width at half maximum of the PL is 72, 16 and 9.5 nm respectively. This dramatic line narrowing is a consequence of optical gain at 700 nm, as will be confirmed with the VSL measurements.

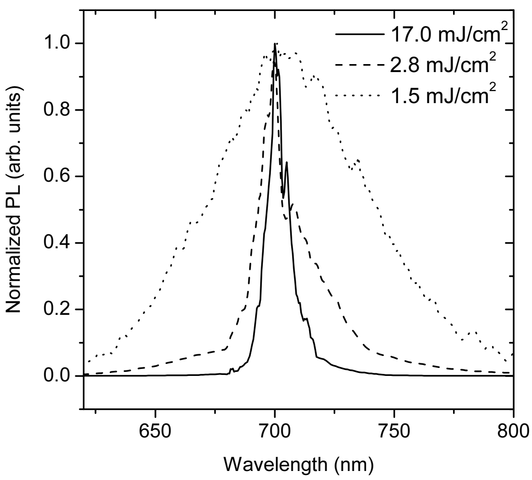

Fig. 1 Normalized guided PL spectra for three different pulse powers illuminating the whole sample with the excitation line.

Figure 2 shows the VSL curves for different pump powers. Three significant wavelengths were chosen for the gain measurements: 700, 680 and 720 nm (Figs. 2a, b and c respectively). In Fig. 2a, for low powers a sublinear behaviour is observed, and when the pump power increases, the dependence turns into a positive exponential, demonstrating unambiguously positive optical gain. The fit with Eq. (1) is shown for each case, and the obtained gain value is shown in the legend. Figure 2d shows the gain coefficient versus power density for the three analyzed wavelengths. The gain threshold at 700 nm is ~3 mJ/cm^2, at 680 nm is ~10 mJ/cm^2, and at 720 nm no gain was observed, although almost transparency was achieved at the highest power. Table 1 summarizes the obtained results for the three wavelengths. To ensure the reliability of the VSL technique, we performed SES losses measurements at the same wavelengths in order to compare the results with the low-power VSL gain coefficient [15]. Table 1 shows these results too, and one can see that there is good agreement between both techniques.

It is worth noting the fact that the gain coefficient at 700nm varies from -10 cm^{-1} up to 8.7 cm^{-1} when the power is increased. The gain seems to saturate at this value (see Fig. 2d), which could imply that total population inversion has occurred. In order to model this behaviour, let us assume a simple two-level system, where N is the total number of active dye molecules per cm^3 and N^* the number of excited dye molecules. In addition, we assume that the emission and absorption cross sections are the same, and equal to σ. If these dye molecules are in the core of a waveguide with intrinsic losses α_{wg} and optical mode confinement Γ, the gain coefficient would be given by:

$$g = -\alpha_{wg} + \left(2N^* - N\right)\Gamma\sigma \tag{3}$$

Therefore the minimum and maximum gain (g_{min} and g_{max}) would be obtained substuting $N^* = 0$ and $N^* = N$ respectively. If the intrinsic losses of the waveguide are low, $g_{\max}$ should get to values near $-g_{min}$. In our case at 700 nm, $g_{min} = -10$ cm^{-1} which under high pumping rates becomes $g_{max} = 8.7$ cm^{-1} gain, which means that almost total population inversion has been achieved and that the intrinsic losses are of the order of 1 cm^{-1} or less.

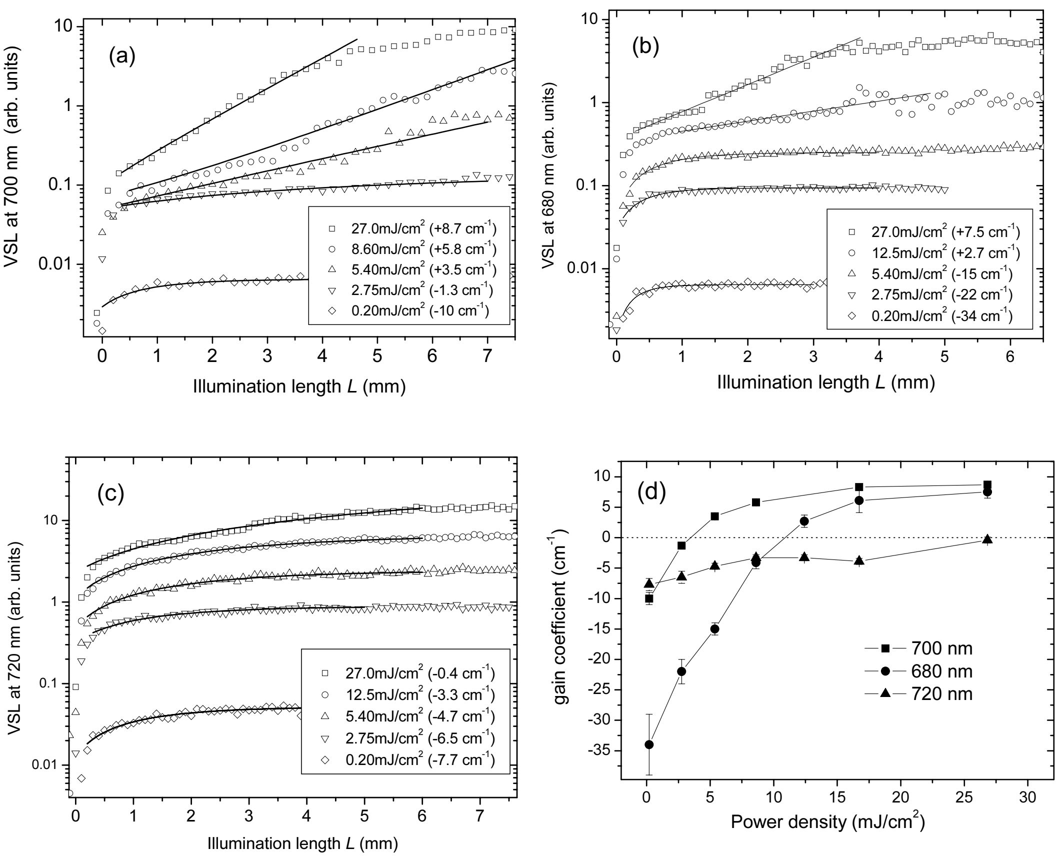

Fig. 2 VSL measurements at 700nm (a), 680nm (b) and 720nm (c) for different pump densities, with their fits to Eq. (1). The legends indicate the power density of the pulses and the gain coefficient obtained from the fit. (d) Gain coefficient versus pump energy density at different wavelengths.

Table 1 Gain and losses results at three different wavelengths.

Wavelength (nm)	SES losses (cm^{-1})	VSL gain coefficient at minimum power (cm^{-1})	VSL gain coefficient at maximum power (cm^{-1})	Threshold (mJ/cm^2)
700	10.2	-10	8.7	3.0
680	30	-34	7.5	10
720	7.7	-7.7	-0.4	-

Laser dyes usually undergo bleaching after a certain number of pulses, especially when they are not circulated, as in our case. We observed that after 1000 pump pulses the amplified signal starts to decrease appreciably. After 10^4 pump pulses, the waveguide still showed amplification, but with ~4 times higher pump threshold. However, the original behaviour is recovered when the sample is rinsed and impregnated again.

5 Conclusions

Optical gain has been observed in oxidized porous silicon planar waveguides impregnated with Nile Blue molecules. We performed guided PL measurements, together with VSL and SES techniques, and highest gain values of 8.7 cm^{-1} have been measured at 700 nm. This work can open the way to a dye laser in a silicon compatible material.

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