Optical gain in dye-doped polymer waveguides using oxidized porous silicon cladding

Daniel Navarro-Urrios[a*], Mher Ghulinyan[a], Paolo Bettotti[a], Néstor Capuj[b], Claudio J. Oton[c], Fernando Lahoz[d], Inocencio R. Martin[d] and Lorenzo Pavesi[a]

[a] Nanoscience Laboratory, Dept. of Physics, University of Trento, Via Sommarive 14, I-38050 Povo (Trento);

[b] Dpto. de Física Básica, University of La Laguna, Tenerife, Spain;

[c] Optoelectronics Research Centre, University of Southampton, SO17 1 BJ, Southampton, UK;

[d] Dpto. de Física Fund. y Exp. Electr. y Sist., University of La Laguna, Tenerife, Spain;

ABSTRACT

We report on a novel organic/inorganic hybrid waveguide approach, which is composed of a cladding of extremely low refractive index oxidized porous silicon formed on a bulk silicon substrate and of it, a polymeric (polymethylmethacrylate) core doped with a visible laser dye (Nile-Blue) was deposited by spin coating.

The waveguiding properties of the structures have been characterised by means of the m-line technique, demonstrating that the use of oxidized porous silicon as a cladding can considerably improve the mode confinement factor of single-mode waveguides. The low refractive index achievable in the cladding (n=1.16) allows forming waveguides with a low index polymer cores.

Variable stripe length (VSL) measurements have been also performed in order to characterise the amplification properties of the waveguides. We demonstrate a clear transition from losses to gain at 694nm with a pump threshold of 28mJ/cm^2. Values of net optical gain up to 104dB/cm have been measured at this wavelength.

Keywords: optical amplifier, gain, polymer, porous silicon, laser dye

1. INTRODUCTION

Photonic polymer devices show interesting properties for optical information technologies.[1-3]. Due to their structural flexibility and their unique processing and fabrication capabilities, polymers are being increasingly used for a variety of optical applications including telecommunications, optical interconnects, high density data storage, optical processing, electro-optic modulation and switching, and displays[4]. Many encouraging results on optical amplification and even laser action have been reported in dye doped polymer based devices (see for example refs.5,6).

One drawbacks of most of the polymers used in optical applications is their low refractive indices which are not easily compatible with the request of silicon photonics. The large silicon (n=3.5) and silicon oxide (n=1.45) refractive indices do not match the low refractive index of polymers (n=1.3-1.5). Thus optical components fabricated on silicon have to be formed by a multilayer polymeric structure with a few percent index contrast between the cladding and the core layer, which in turn means large footprint for the optical components. In this work we demonstrate a system which is suitable to allow tight integration of polymer based active optical components on silicon.

We propose to use a porous silica glass cladding material obtained by strong oxidation of porous silicon (PS). It is possible to control the material refractive index of PS during the electrochemical etching of the silicon substrate [7] by modifying the etching current density. This allows the realization different vertical 1D photonic structures [8] and optical waveguides [9,10]. By oxidation, PS can be converted into a porous SiO_2 glass, which a very low material refractive indices and negligible absorption of visible light [11].

* navarro@science.unitn.it ; phone 39 0461882030; fax 39 0461882967

Photonic Materials, Devices, and Applications II, edited by Ali Serpengüzel,Gonçal Badenes, Giancarlo Righini
Proc. of SPIE Vol. 6593, 659321, (2007) · 0277-786X/07/$18 · doi: 10.1117/12.721400

Due to its common use as positive photoresist in electron beam lithography and nanoimprint technology and its transparency in the visible range, polymethylmethacrylate (PMMA) is a promising material in hybrid organic-inorganic optical component. In this work, we combine the properties of PMMA with that of porous silica to form organic/inorganic hybrid waveguides. The latter have a porous silica cladding with refractive indices of 1.16-1.45 and a polymer core obtained by spin-coating PMMA onto the cladding. In the precursor PMMA solution a laser dye was dissolved in order to form an active core. Optical gain in small modal size slab waveguides is demonstrated. Additionally, with this work we validate the use of porous silica as an extremely low refractive index material with good optical qualities to be used in compact optical devices for silicon photonics. In the present work we report on the results for the lowest refractive index which was possible to achieve in the cladding, i.e. the most compact monomodal slab waveguide with the highest confinement factors.

2. EXPERIMENTAL

Porous silicon samples were fabricated by electrochemical etching of heavily doped (0.01 Ωcm resistivity) p-type silicon. Single monolayer samples were grown by applying a current density of 80 mA/cm^2. The samples were then annealed at 900ºC for 3 hours in order to completely oxidize the structures, turning them into porous silica with a refractive index of 1.16. The polymer core of the waveguides was obtained by spin-coating PMMA onto the cladding. The core thickness was controlled by the spin-coater revolution velocity. The precursor PMMA solution was dissolved with a visible laser dye, Nile Blue (LC 6900)[12] at various molar concentrations 10^{-6}-10^{-3} M. The absorption and photoluminescence spectra of this dye are shown in Figure 1. The spin-coated films were baked for 10 min at 160°C over a hot plate. The samples were cleaved on one border to collect the guided light.

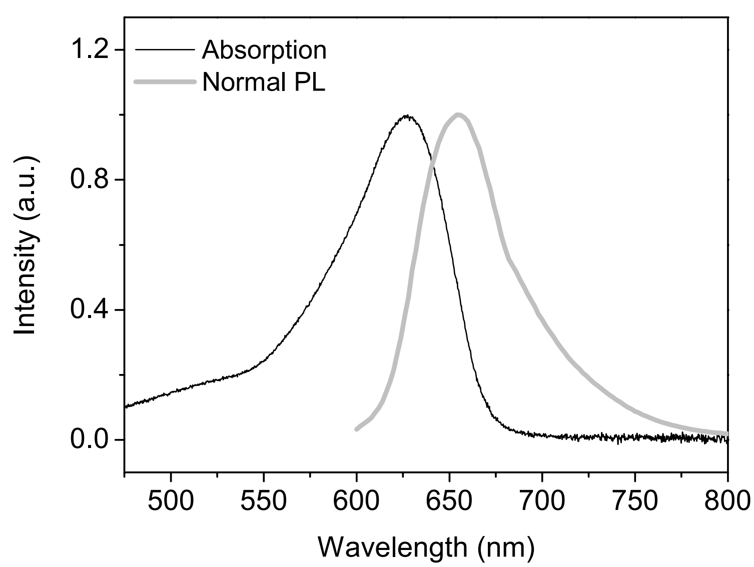

Figure 1. Absorption (black) and emission (grey) spectra of Nile Blue.

In order to characterize the optical properties of the waveguides, we have performed standard m-line prism coupling[13] measurements using the 633nm line of a He-Ne laser. With this technique, the angular dependence of the reflectance of the beam is measured, and each guided mode is observed as a sharp dark peak in the reflectance spectra. The effective index of each guided mode is directly related with the incident angle and the refractive index of the prism.

Spectroscopic measurements were performed by pumping with a frequency doubled Nd:YAG laser (532 nm) in Q-switching mode, which gave 5ns pulses with vertical polarization. A lens system focused the pump beam on the waveguide to a horizontal line which was 3 cm long and 300 micron wide. The maximum pump energy employed was 3mJ (30 mJ/cm^2). The end face guided light was collected with a lens (10cm focal length) to avoid confocal effects. The lens was partially covered with an opaque surface to avoid collecting non guided signal. The guided luminescence was imaged into a monochromator interfaced with a photomultiplier tube (PMT), which allowed single shot luminescence measurements. All the spectra were collected with 1 nm spectral resolution, and all the measurements at fixed wavelength, with 5nm resolution.

To characterize the waveguide losses of the samples We performed "shifting of excitation spot" measurements (SES)[14]. This technique consists in scanning a pump spot through the waveguide surface, and measuring the guided luminescence versus the distance *d* to the edge. The obtained signal as a function of *d* should follow the Beer law:

$$I_{SES}(d) \propto e^{-\alpha d} \tag{1}$$

where α is the loss coefficient Gain measurements were performed with the variable stripe length method (VSL) [15]. In Figure 2 it is shown a scheme of the VSL setup. The sample (either transparent or absorbing) is optically excited by a narrow stripe. A metal blade was placed in front of the waveguide in order to vary the illumination length, L. The distance between the waveguide and the blade was only 5 mm in order to minimize diffraction effects. An amplified spontaneous emission (ASE) signal I_{ASE} is therefore collected from the edge of the sample as a function of L. As a result of population inversion achieved at high pumping rates, spontaneously emitted light is amplified and an intense, and partially coherent ASE signal grows up exponentially with the excitation length. Distinctive characteristics of the ASE intensity suddenly appear, since the ASE bandwidth is appreciably narrower than that of spontaneous emission and exhibits a threshold behavior. Although ASE signal is accompanied by a shortening of the measured lifetime, within our experimental conditions it was impossible to measure this since the dye radiative lifetime was already of the order of nanoseconds, i.e. similar to the pumping pulse length. In a first approximation it is customary to interpret the VSL data within a simple model where the effect of spontaneous emission is considered within a one dimensional amplifier with a net modal gain coefficient g:

$$I_{ASE}(L) \propto \frac{1}{g}\left(e^{gL} - 1\right) \tag{2}$$

where g has to increase from negative to positive values when the pump energy (EP) is increased. It is worth noting that the coefficient α measured with the SES technique corresponds to –g measured with VSL at very low pump power. This agreement confirms the reliability of the VSL measurement.

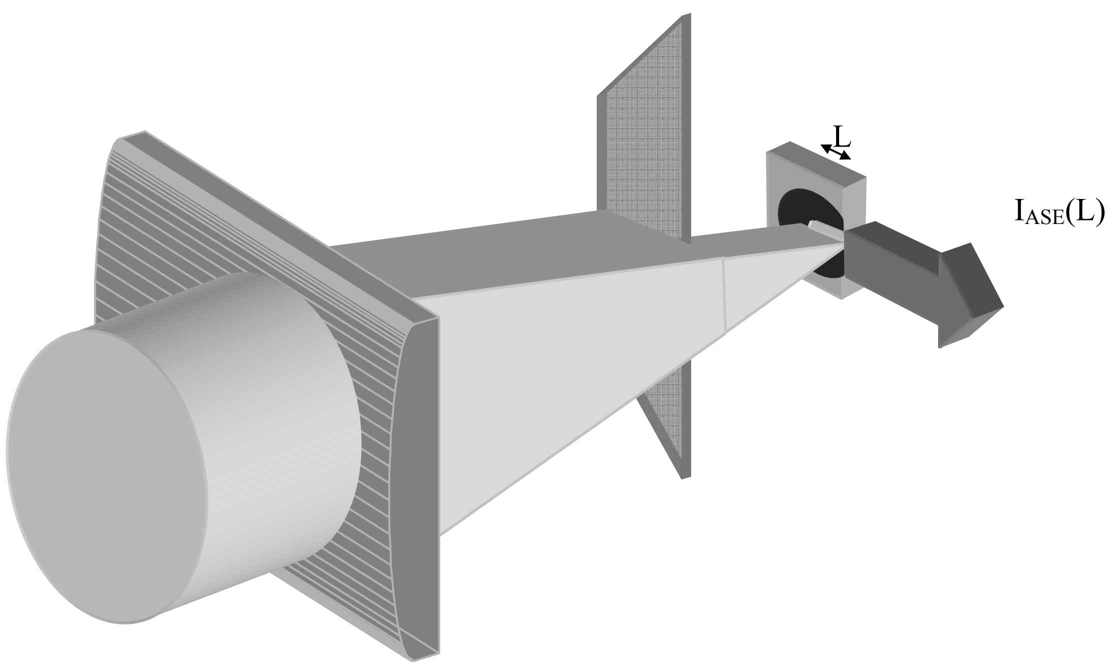

Figure 2. Scheme of the VSL setup used for the amplification measurements.

The confinement factor Γ is defined as the percentage of power within the core of the waveguide. Assuming that the energy outside the core region is totally wasted, Γ is therefore the factor by which the gain coefficient is reduced, or equivalently, the factor by which the loss coefficient is increased. The net material gain g_{net} is therefore defined as g/Γ.

It is also worth noting that in our experiment one single laser shot was used per each L value.

3. EXPERIMENTAL RESULTS

3.1 Waveguide characterization

In Figure 3 we show the results of the m-line measurements and simulations for both TE and TM polarizations together with the extracted refractive index profiles. The presence of only one dark line (arrow) in the m-line measurements indicates that the waveguide is monomodal. From the simulation, we extracted a refractive index of the core n_{core}~1.49, a core thickness d_{core}~395nm, a refractive index and thickness of the cladding n_{clad}~1.16 and d_{clad}~5μm, respectively, a material birefringence (B=3%) of the cladding layer due to the vertical structure of the pores[16,17], and an optical confinement factor for the TE mode Γ_{TE}=69% and for the TM mode Γ_{TM}=56%. The refracted index profile extracted from the simulations is represented in the bottom panel of Figure 3. It is worth noting that if instead of an oxidised porous silicon cladding we would have used bulk silica, none guided modes would have been found because of the low index contrast. A core thickness of up to 1μm should have been used in order to have a waveguide with similar confinement factors Γ.

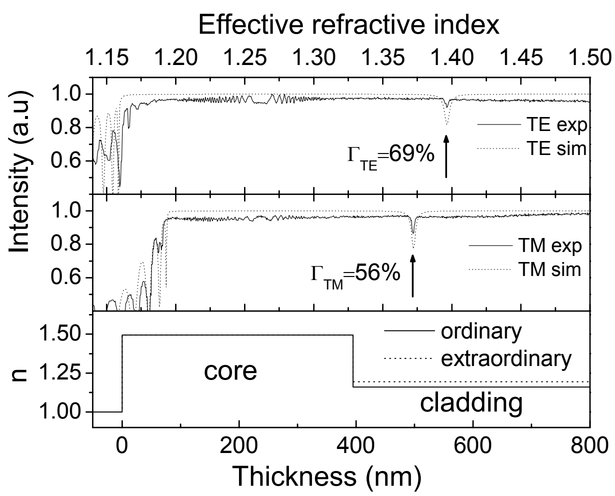

Figure 3. m-line measurements (line), simulations (dots) for TE (top panel) and TM (middle panel) polarization. The arrows show the position of the dark line due to the propagating mode. The bottom panel refers to the refractive index profile for the ordinary (line) and extraordinary (dots) rays extracted from the simulations.

3.2 Guided Photoluminescence measurements

We show in Figure 4 the guided light spectra for two different concentrations of the solution, pumping with high and low pump powers. It is worth noting that the measurements are taken in a VSL configuration pumping with the whole stripe length. For concentrations lower than the ones showed in the following figures, a broad emission spectrum and a linear intensity dependence on the pump power were observed

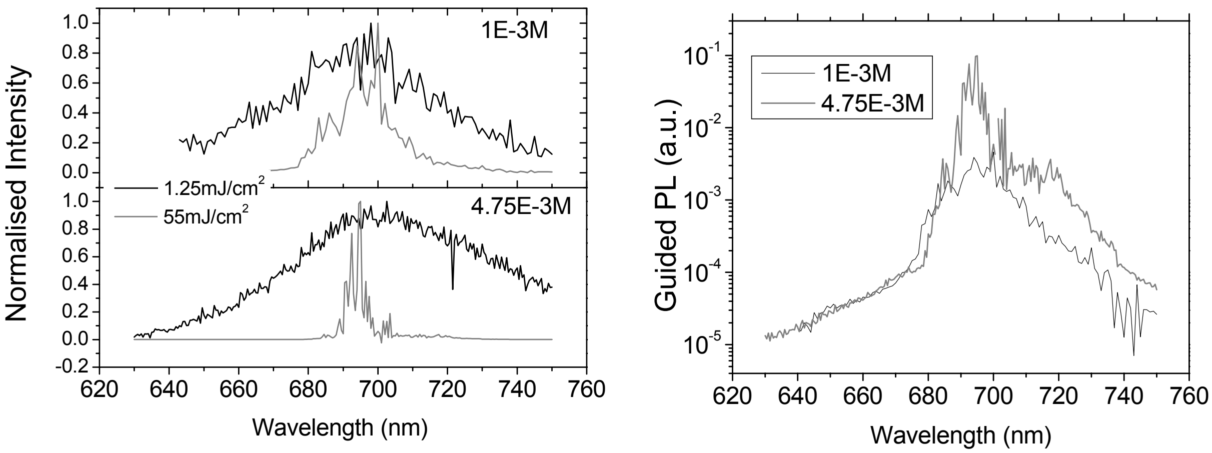

Figure 4: a) Normalised guided PL pumping with the whole line, for different molar concentrations at low and high pump density powers. b) High power PL for different solution concentrations. Intensities are comparable. Note the logarithmic scale

On the contrary, for the high concentration samples, a dramatic change in lineshape and a strong nonlinear intensity increase as a function of the pump power were observed (Fig. 4b). Let us focus our discussion on the 4.75 $x10^{-3}$ M sample. In Figure 5a) we show in log scale the guided PL spectra for high and low pump power. Therefore, by increasing the pump power 40 times, at 694nm we have increased the signal 25000 times, thus demonstrating the strong non-linear behaviour.

In order to see the behaviour of the intensity of each band versus power, we have chosen three significant 5nm wide spectral bands, centred at 676, 694and 780 nm. The PL was integrated along the bands, and the result is shown in Figure 5 b).

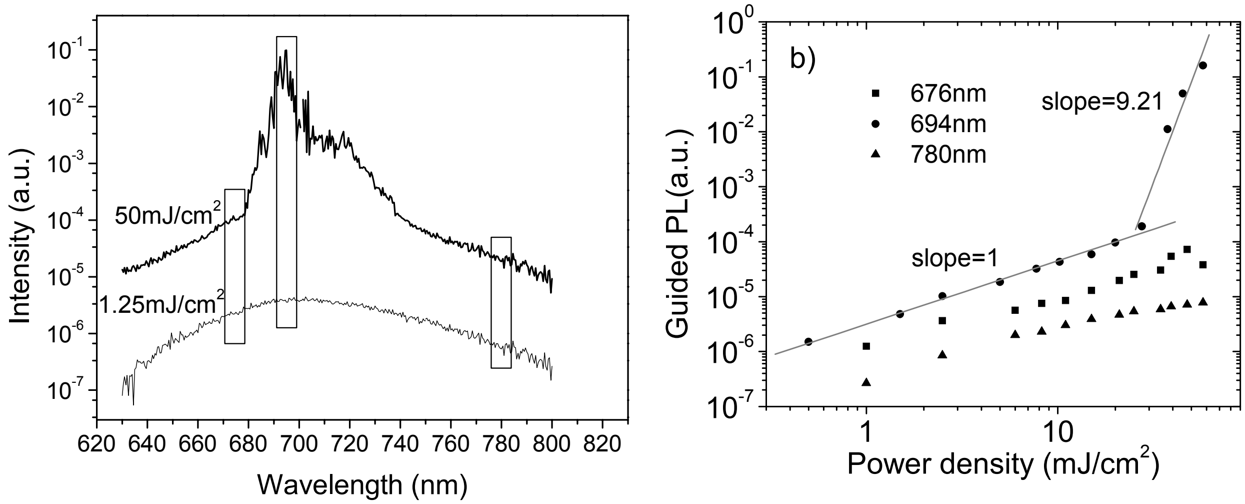

Figure 5: a) Guided PL spectra for high and low power illuminating the whole sample (~8mm). b) Integrated PL for three different 5nm wide bands around the indicated wavelengths versus pump power.

We can see that for the maximum of the amplified band the signal grows strongly superlinearly after a certain power threshold. At 676nm we are out of the band and the behaviour is almost linear. Instead, the signal at 780nm tends to saturate.

In Figures 6 we show, for high and low pump power, the spectrum illuminating with different stripe lengths. At low pump power the intensity is not much more intense when pumping with the whole stripe and even the redshift due to self-absorption is not appreciable because of the high losses, i.e. the main contribution in the edge emission is due to the region closer to the edge and this emission spectrum is indeed similar to the normal PL. At high pump power, the spectral shape of the spectrum changes and the intensity grows considerably when increasing the pumping length.

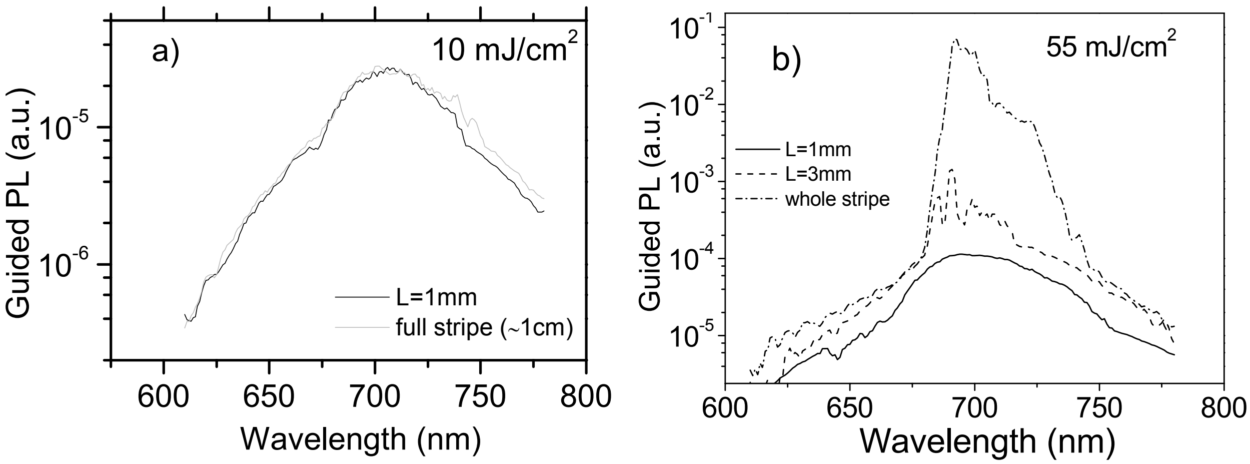

Figure 6: a) Guided PL versus different excitation lengths at low pump power and b) at high pump power.

It is well known that the use of the SES technique to measure the losses of the waveguide is quite limited because some internal source of light is needed; therefore, the spectral range available to make the measurement is thus restricted to the PL spectrum of the active material. Losses measurements using the SES technique were thus performed at the same selected wavelengths bands of Figure 5, i.e. 676, 694 and 780 nm. From an exponential decay fit, and using eq. 1 it is possible to extract the propagation losses of guided light in the slab waveguides (number on the curves in Figure 7. At 676nm the measurement allows extracting only a lower limit to the losses coefficient, which is around 65cm^{-1}. We have checked the influence of the modal confinement to the losses by changing the refractive index of the cladding and we found that the higher refractive index of the cladding, i.e. lower confinement, the lower the losses coefficient. This behaviour is opposite to what expected if the source of the losses would be in the interfaces core-cladding, because less confinement significantly increases the interaction of the optical mode with the sidewall surface roughness[18]. Thus, we argue that the main source of the propagating losses is found in the direct absorption by the dye. Quite high losses values (15cm^{-1}) are found also at 780nm, where the dye in solution does not present appreciable absorption (see Figure 1). This effect could be associated to absorption of dye agglomerates within the core or perhaps to scattering due to inclusions in the core, such as voids, cracks or bubbles, present when doping the polymer with the laser dye molecules. In any case the strong relation between the losses and the presence of the dye is demonstrated by the low losses (4 cm^{-1} at 730 nm) measured in a slab waveguide with an order of magnitude lower dye concentration.

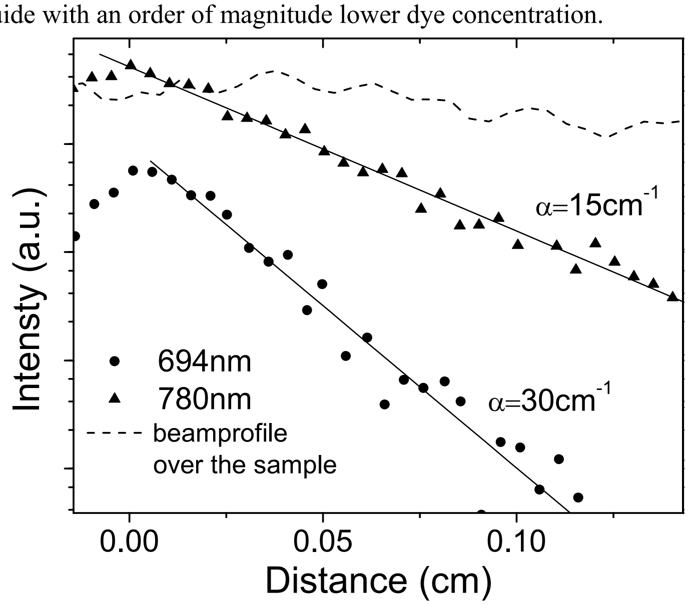

Figure 7: SES curves for different wavelengths, 694 (circles) and 780 nm (triangles) with exponential decay fits (lines). The label on the curves refers to the losses coefficients. It is also shown the beam profile over the sample (dashed line).

Figure 8 shows the VSL measurements done for different pump powers at 694nm, the maximum of the ASE signal. For low EP a sublinear behaviour of $I_{ASE}(L)$ is observed while, when EP increases, $I_{ASE}(L)$ becomes a positive exponential. This demonstrates unambiguously positive optical gain. A fit with Eq. 2 yields the gain coefficient values, that are also shown in the graph. A maximum value of $24cm^{-1}$ (104dB/cm) is measured. Note that, at maximum power, the gain is constant along the first 3 mm, reaching a total signal amplification of almost 31dB, and beyond that length the gain starts to saturate.

The losses measured with VSL at very low pump powers correspond well with the losses found by SES measurements, which confirms the reliability of the measurement.

Regarding the stability of the signal at high power, it was degrading with the number of shots. Laser dyes usually undergo bleaching after a certain number of pulses, especially when they are not circulated, as in our case. Indeed we observed that after 1000 pump pulses the amplified signal starts to decrease appreciably. After 10^4 pump pulses, the pumped region of the waveguide still showed amplification, but with ~4 times higher pump threshold.

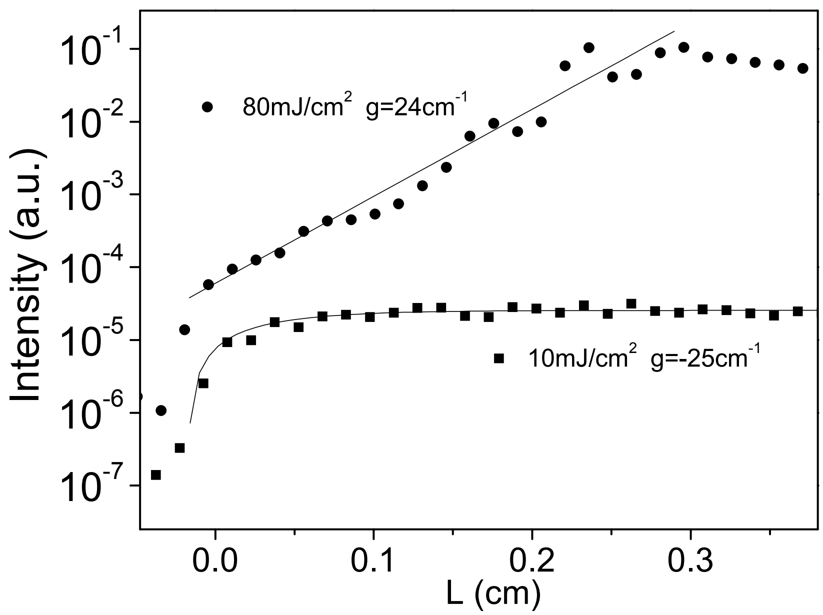

Figure 8: VSL measurements at 694nm for high (circles) and low (squares) pump powers. The fits using eq. 2 are also shown (solid lines).The results of the fits are shown on the legend. Intensities are comparable.

4. CONCLUSIONS

In conclusion, we have demonstrated slab optical waveguides using a new approach, in which the cladding layer is made of very low refractive index porous silica and the core layer is made of PMMA doped with a laser dye. Several different evidences of high values of net optical gain at 694nm have been reported, arriving to values as high as 104dB/cm, constant over 3mm. These results could open new routes towards realising low cost – high performing optical amplifiers or lasers. In addition, we emphasize that the use of oxidized porous silicon as cladding layer would allow the deposition of low refractive index polymers on silicon to form compact optical devices.

Acknowledgements

We acknowledge ITC-irst Trento for the spin-coating procedure. This work has been supported by an Italy-Spain cooperative grant, Ministerio de Educación y Ciencia (MAT2004-6868) and Gobierno Autónomo de Canarias (PI042004/018).

REFERENCES

1. N. Tessler, G. J. Denton, and R. H. Friend, Nature London, 382, 695 (1996)
2. F. Hide, M. A. Diaz-Garcia, B. Schwartz, M. Andersson, G. Pei, and A. J. Heeger, Science, 273, 1833, (1996).
3. S. Frovlov, W. Gellerman, M. Ozaki, K. Yoshino, and Z. V. Vardeny, Phys.Rev. Lett. **78**, 729 (1996).
4. Hong Ma, Alex K.-Y. Jen, and Larry R. Dalton, Adv. Mater., **14**, 1339, (2002)
5. Grace Jordan,_ Michael Flämmich_ Manuel Rüther_ Takeyuki Kobayashi, Werner J. Blau, Yasuhiro Suzuki and Toshikuni Kainom, Appl. Phys. Lett. 88, 161114 (2006).
6. K. Yamashita, T. Kuro, K. Oe, H. Yanagi Appl. Phys. Lett. 88, 241110 (2006).
7. O. Bisi, S. Ossicini, L. Pavesi , Surf. Sci. Rep. 264, 1-126 (2000)
8. M. Ghulinyan, C. J. Oton, G. Bonetti, Z. Gaburro, and L. Pavesi, J. Appl. Phys. 93, 9724, (2003)
9. C. J. Oton, D. Navarro-Urrios, N. E. Capuj, M. Ghulinyan, L. Pavesi, S. González-Pérez, F. Lahoz, and I. R. Martín, Appl. Phys. Lett., 89, 011107, (2006).
10. P. Pirasteh, J. Charrier, Y. Dumeige, P. Joubert, S. Haesaert, A. Chaillou,L. Haji, P. Le Rendu, and T. P. Nguyen, Phys. Status Solidi A 202, 1712 (2005).
11. D. Navarro-Urrios, Ph. D. Thesis, University of La Laguna, 2006.
12. U. Brackmann, Lambdachrome Laser Dyes, 3rd ed. Lambda Physik®, Germany, (2000).
13. P. K. Tien, R. Ulrich, and R. J. Martin, Appl. Phys. Lett. 14, 291 (1969).
14. J. Valenta, I. Pelant, and J. Linnros, Appl. Phys. Lett. 81, 1396 (2002).
15. K. L. Shaklee, R. E. Nahaori, and L. F. Leheny, J. Lumin. 7, 284 (1973).
16. D. Kovalev, G. Polisski, J. Diener, H. Heckler, N. Künzner, V. Yu. Timoshenko, and F. Koch, Appl. Phys. Lett. 78, 916 (2001).
17. C. J. Oton, Z. Gaburro, M. Ghulinyan, L. Pancheri, P. Bettotti, L. Dal Negro, and L. Pavesi, Appl. Phys. Lett. 81, 4919 (2002).
18. K. K. Lee, D. R. Lim, H.-C. Luan, A. Agarwal, J. Foresi, and L. C. Kimerling, Appl. Phys. Lett., 77, 1617 (2000).