

Guided photoluminescence study of Nd-doped Silicon Rich Silicon Oxide and Silicon Rich Silicon Nitride waveguides

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Abstract: Planar waveguides made of Nd³⁺-doped silicon rich silicon oxide (SRSO) and silicon rich silicon nitride (SRSN) have been fabricated by reactive magnetron sputtering and characterized with special emphasis on the comparison of the guided photoluminescence (PL) properties of these two matrices. Guided fluorescence excited by top surface pumping at 488 nm on planar waveguides was measured as a function of the distance between the excitation area and the output of the waveguide, as well as a function of the pump power density. The PL intensity increased linearly with pump power without any saturation even at high power. The linear intensity increase of the Nd³⁺ guided PL under a non-resonant excitation (488 nm) confirms the efficient coupling between either Si-*np* and RE ions for SRSO or radiative defects and rare earth ions for SRSN. The guided fluorescences at 945 and 1100 nm were observed until 4 mm and 8 mm of the output of the waveguide for Nd³⁺ doped SRSO and SRSN waveguides, respectively. Co-propagation has been only carried out on silicon nitride planar waveguide for a sample length superior to 1.5 cm. The guided fluorescence decays of Nd³⁺-doped-SRSO and -SRSN planar waveguides have been measured and found equal to

$97\mu\text{s} \pm 7$ and $5\mu\text{s} \pm 2$, respectively. These results show notably that the Nd^{3+} -doped silicon rich silicon oxide is a very promising candidate on the way to achieve a laser cavity at $1.06\mu\text{m}$.

1. INTRODUCTION

A main objective in silicon photonics research is the development of novel Si-compatible materials for the engineering of light sources directly integrated on optical chips [1-3]. Rare-earth (RE) doped-silicon-based matrices has gathered much attention as a possible way of realizing a Si-based source. Thus, erbium-doped films of Silicon Rich Silicon Nitride (SRSN) or Silicon Rich Silicon Oxide (SRSO) have been investigated as promising host matrices because of the efficient sensitization of RE ions that favours an enhancement of their emission [1-13]. Erbium is one of the most studied ions in different matrices these last years because of its leading position in telecommunication applications. This is why numerous works have been carried out on Si-based waveguides doped with Er^{3+} ions [1, 2 and 5] but up to now, there is only few papers demonstrating the achievement of a net gain [16].

By contrast, Nd^{3+} -doped SRSO or SRSN systems have not been extensively studied. But their spectroscopic four-level scheme prevents photons emitted at $1.06 \mu\text{m}$ to be reabsorbed from the ground level. Consequently, such Nd-doped materials could be good candidates for the demonstration of the first Si-based laser emitting at $1.06 \mu\text{m}$, taking advantage from high absorption cross section of Si-np and efficient Si-np: Nd^{3+} transfer energy [17-19].

Based on these considerations, it is of interest to study the optical properties of guided luminescence of active Nd-doped Silicon Rich Silicon Oxide and Silicon Rich Silicon Nitride planar waveguides by top surface pumping.

In this paper, we describe the optical properties of Nd^{3+} -doped silicon rich silicon oxide (SRSO) and silicon rich silicon nitride (SRSN) thin layers synthesized by reactive magnetron sputtering by means of m-lines and reflectance spectra measurements. The optical properties of planar waveguides were investigated by measuring their optical losses and through guided photoluminescence (PL) by top surface pumping. The evolution of the PL was

studied as a function of the pumping power and as a function of z , the distance between the excitation area by top surface pumping and the output of the waveguide. We compare the different obtained PL spectra and discuss on the conditions to obtain efficient PL of Nd^{3+} but also its limitation according to the type of matrix and the pumping scheme.

2. EXPERIMENTAL CONDITIONS

The Nd^{3+} -doped SRSO films were synthesized by reactive magnetron co-sputtering of pure Nd_2O_3 and SiO_2 targets, followed by a thermal treatment at 900°C during one hour under a pure nitrogen flow. Using the ability of H_2 to reduce the oxide species released by the sputtering of the SiO_2 target, the incorporation of Si excess was monitored by a gas flow of H_2 that was mixed to Ar in the sputtering gas. The RF power densities on SiO_2 and Nd_2O_3 targets were 7.6 W.cm^{-2} and 0.3 W.cm^{-2} , respectively.

The SRSN layers were also obtained by reactive magnetron co-sputtering of pure Nd_2O_3 and Si targets. After standard piranha cleaning procedure, the substrate has been introduced in the vacuum chamber and heated at 200°C during 1 hour prior to deposition. The Ar/ N_2 gas flow ration was fixed at 10% and the plasma pressure was set at 2mTorr. The RF power densities on Si and Nd_2O_3 targets were 4.5 W.cm^{-2} and 0.45 W.cm^{-2} , respectively. The films were subsequently annealed at 700°C during 1 hour in a pure nitrogen flow. More details of the fabrication process can be found elsewhere [19, 21].

The planar waveguides were obtained from a guiding layer of SRSO or SRSN, deposited on an $8\mu\text{m}$ -thick thermally grown SiO_2 , used as a cladding layer, on Si wafer.

The optical properties (refractive index, reflectance) of the fabricated layers were analyzed by means of either a m-lines method at different wavelengths (633, 1310, and 1550 nm) or a LAMBDA 900 Perkin Elmer beam spectrometer equipped with a specular

reflectance module with a 6° fixed angle. These measurements enabled the optical thickness to be determined by evaluating the beat of the interference fringes on the basis of the effective medium theory (Bruggeman model) [22, 23]. Thus the refractive index as a function of the wavelength can be deduced. In addition, the interface roughness σ of both layers was measured by means of a profilometer (Dektak150).

Optical losses were measured by studying the scattered light from the surface of the planar waveguide [24-26]. Laser light was microlensed fiber-coupled into the waveguide. The intensity of the scattered light was recorded with a digital camera placed above the sample. Transverse scanning along the light propagation direction enabled the 2-D light intensity distribution of the guided modes to be obtained. Then, the intensity curve was obtained by integrating the data along each sampling transverse line. The light intensity decreased exponentially with the z-propagation distance.

In this study, the attenuation values were the average of several measurements performed on a lot of samples. The first 1-2mm from the input facet were not considered to improve the signal to noise ratio and also to avoid the multiple scattering at the injection point. The polarization of the coupled light into the waveguide was not controlled.

The guided PL measurements were performed at room temperature using an argon laser operating at 488 nm ($3.3 \cdot 10^{19}$ photons/cm²/s) in top or propagated pumping configuration. For this latter, the pump light is coupled at the input of the planar waveguide using an optical fiber. The 488 nm excitation wavelength is non-resonant line with Nd³⁺ ions so that only an indirect excitation of Nd through the Si-np can occur [19]. The PL was dispersed in a Jobin-Yvon iHR320 monochromator and detected by a cooled InGaAs detector in the 850-1650 nm wavelength range. A lock-in amplifier was used to record the near infra-red spectrum if necessary. The guided fluorescence was optimized by observing the guided mode at the output of the planar waveguide by means of the InGaAs camera. For each figure of the guided PL

spectra, the normalization factor is equivalent to the maximum intensity of the most important spectrum. Then, the other spectra are normalized according to this value for both SRSO and SRSN spectra. Since the experimental spectra were obtained in the same experimental conditions, we can suppose the maximum intensity is equal to 1 for each group of spectra.

The lifetime was obtained by excitation with a Nichia GaN laser diode emitting 1 W at 444 nm. It was modulated by a pulse generator; the sample excitation was done by 500 μ s square pulses width. The detection was carried out by a Hamamatsu InGaAs detector cooled to -20 °C. High-pass filters cutting at 1 μ m were used to select the Nd^{3+} emission. A preamplifier FemtoDHPCA-100 converts the photodiode current into voltage; the time response of the measurement chain is 2 μ s.

3. RESULTS and DISCUSSION

3.1 Physical and optical characterizations

Firstly, monolayers with different thicknesses and bilayers were fabricated to measure the refractive index and to study its variation with the layer thickness. Then, these measurements, added to the numerical simulation, allowed us to elaborate the planar waveguide structures. The refractive index was measured by using m-lines method at different wavelengths (633, 1310, or 1550 nm). These measurements have been confirmed by recording the reflectance spectra of the layers and then fitting them using transfer matrix formalism. This formalism includes the Bruggeman effective medium approximation for SRSO and for SRSN layers [22, 23]. For the former, a system composed of Si-np embedded in a silica matrix is considered. For the latter SRSN, the system is composed by a Si_3N_4 matrix containing a Si excess. This method allows us to determine the evolution of the refractive index of a SRSO or SRSN layer as a function of the wavelength.

Figures 1(a) and (b) show an example of good agreement between the experimental and theoretical reflectance spectra for a Nd³⁺-doped SRSO or SRSN deposited on an 8μm-thick thermally grown SiO₂ on Si substrate. The refractive index at 1.06 μm was found to be equal to 1.54 and 2.09 for SRSO and SRSN, respectively (Table 1). The evolution of the refractive index of a SRSO layer fits well with the values obtained from the M-lines measurement (Fig. 1(c)).

From the knowledge of the refractive index at 1.06 μm, the propagated modes were calculated using the dispersion equation for a planar waveguide as a function of the guiding layer thickness (Fig. 2 (a) and (b)) for TE and TM polarizations. The accuracy of the guiding layer thickness is not very important because the planar waveguides will be single-mode up to a certain layer thickness. On the basis of these results, 1.1 μm-thick Nd³⁺-doped SRSO and 0.38 μm-thick Nd³⁺-doped SRSN were deposited on a 8 μm-thick thermally grown SiO₂ on Si wafer to obtain the single-mode waveguides as shown in Figure 2. More details of the physical characteristics of SRSO and SRSN layers are reported in Table 2 [19, 20].

Optical losses were measured on these planar waveguides at 1.55 and 1.06 μm. They are in the same order of magnitude for both waveguides and are about of 1 dB/cm at 1.06 μm and 0.6 dB/cm at 1.55 μm (Table 2). The optical loss difference can be explained by the fact that the main contribution to such losses in the near infrared range is due to surface scattering which decreases with the wavelength[27].

3.2 Guided PL spectroscopy and discussion

The guided PL measurements on SRSO and SRSN planar waveguides were performed at room temperature using an argon laser operating at 488 nm. Firstly, the alignment is optimized between sample, objective and spectrometer by fiber-coupling laser light at 1.06 μm into the waveguide. The guided fluorescence achieved by top surface pumping is studied

as a function of the pump power and also as a function of z , the distance between the excitation area and the output of the waveguide (Fig. 3(a)).

Figure 3(b) represents the evolution of normalized guided PL spectra of SRSO and SRSN for a pump power equal to 150 mW ($3.3 \cdot 10^{19}$ photons/cm²/s) at room temperature. Spectra reveal two principal bands centered about 930 and 1100 nm corresponding to $^4F_{3/2} - ^4I_{9/2}$ and $^4F_{3/2} - ^4I_{11/2}$ radiative transitions of Nd³⁺ ions. The presence of the guided PL of Nd³⁺ excited by a non resonant excitation at 488 nm confirms the efficient sensitization of either Si-np for SRSO layers or radiative defect for SRSN one towards RE ions. The band centered at 1340 nm is present only in the case of SRSN corresponding to $^4F_{3/2} - ^4I_{13/2}$ radiative transitions of Nd³⁺ ions. But this weak band is due to the low branching ration of the transition. It's not observed in the SRSO, it can be explain by a lower branching ratio. In this wavelength range a strong background can also cover up the emission band (Fig. 3(b)).

Moreover, the two spectrum shapes are not identical. The guided PL of SRSO waveguide reveals a broad band centered at 1100 nm that contains two peaks at 1090 and 1126 nm. Taking into account the geometry of our waveguides these two peaks are only attributed to the contribution of active layer. This is confirmed by measurements performed on an undoped SRSO planar waveguides fabricated in the same conditions for which no guided PL has been evidenced. So, we can affirm that the guided PL is coming from the active guiding layer and not from the Si substrate. Furthermore, the cladding layer is an 8 μm thick layer which avoids the radiate modes of an emission from the Si substrate towards the guiding layer.

In the case of SRSN waveguides, a broad band that contains one only peak centered at 1104 nm has been observed.

Figures 4(a) and (b) show the normalized spectra of guided PL for doped planar SRSO and SRSN waveguides recorded at room temperature and for different pump power densities (150 mw corresponds to $3.3 \cdot 10^{19}$ photons/cm²/s). The spectra have the same shape whatever the applied pump power and the PL intensity increases with excitation power. The insets of Figures 4(a) and 4(b) display the guided PL integrated intensity for the two Nd³⁺ emission peaks. The PL intensity increased linearly for low pump power and no saturated intensity was observed at higher power for both waveguides. The linear increasing of the guided PL intensity of Nd³⁺ excited by a non resonant excitation at 488 nm confirms the strong coupling between the Si-np or defects and RE ions in the two different host matrices. However, in the case of SRSO waveguide, we can notice an increasing of the base line intensity with pump power. This increase is wavelength dependant. It could be attributed to some luminescent defects created during the growth, but more investigation must be done to confirm this hypothesis.

Then, the guided fluorescence by top surface pumping at 488 nm on planar waveguides is studied as a function of the distance between the excitation area and the output of the waveguide (z) as shown in Figure 5(a). Figures 5(b) and 5(c) represent the evolution of normalized PL spectra according to the distance z for both waveguides. The guided fluorescences at 945 and 1100 nm were observed until 4 mm from the output of the SRSO waveguide and until 8 mm from the output of the SRSN waveguide. As expected, the guided fluorescence at 945 and 1100 nm decreased when the excitation area moved away the output. This is attributed to the optical loss in the active layer as well as to the beam divergence in the planar waveguide. The larger propagation length observed for SRSN layer is attributed to a better light confinement in the guiding layer due to a higher index contrast (See mode field profiles in Figures 2). The positions of the peaks correspond to the typical transitions between

internal energy levels in Nd³⁺ ions as described in figure 3. The related integrated intensities in the 850-1000 nm and 1000-1200 nm wavelength ranges are reported as a function of the distance in the insets of figures 5(b) and (c). The exponential decrease with the distance has been fitted leading to the attenuation values of 10.4 and 7.3 cm⁻¹ at 945 and 1100 nm respectively in the case of the SRSO waveguide. For the nitride waveguide, the attenuation coefficients are at least two times lower with deduced values equal to 4.4 and 2.3 cm⁻¹ at 945 and 1100 nm respectively. These measurements allows us to believe that a better luminescence efficiency is obtained in the case of SRSN waveguides than in the case of SRSO ones. It is worth noticing that the attenuation value at 945 nm is higher than the one at 1100 nm, because of the reabsorption effect from the ground level.

The comparison of the two samples by measuring the guided PL is not straightforward. One of the reasons is the difference of thickness of the active layers for both matrices (1.1 μm for SRSO while it is of 380 μm for SRSN). The other concerns the difference of the excitation cross section of the SRSN and SRSO matrices. Consequently, lifetime measurements have been performed to determine their luminescence efficiency. Figures 6 present guided fluorescence decays (τ) of ⁴F_{3/2} - ⁴I_{11/2} radiative transition excited at 444 nm by top surface pumping of Nd³⁺-doped- SRSO and SRSN planar waveguides. Due to the non-exponential decay of the luminescence, the fitting by an exponential curve cannot be easily done. The lifetimes (τ) of the ⁴F_{3/2} - ⁴I_{11/2} radiative Nd transition were determined by the equation $\tau = \int I(t) dt / I(0)$, where I stands for fluorescence intensity. The lifetime corresponds to the average lifetime of the level taking into the response time of the amplifier system account. The fluorescence decays of Nd³⁺-doped- SRSO and SRSN planar waveguides were found to be 97 μs ± 2 and 5 μs ± 2, respectively. We notice that the lifetime in the nitride matrix is very low due to the presence of defects.

With these measurements, we can notice a better luminescence efficiency of SRSN waveguides than SRSO ones. Indeed, the guided fluorescence by top surface pumping was observed for some more important length from the output in the case of SRSN waveguide. These measurements confirm that the spontaneous emission is encouraged in the SRSN matrix because of better luminescence efficiency and a low lifetime, whereas the lower PL and a higher lifetime in the case of SRSO matrix should ensure to achieve a better stimulated emission. Consequently, Nd³⁺-doped SRSO waveguides are a promising matrix to achieve a laser cavity at 1.06 μm.

4. CONCLUSION

We have measured low optical losses of about 1 dB/cm at 1.06 μm for both Nd³⁺ doped- SRSO and SRSN waveguides. Guided fluorescence by top surface pumping at 488 nm on planar waveguides was measured as a function of the distance between the excitation area and the output of the waveguide, as well as a function of the pump power. The PL intensity increased linearly with pump powers with no saturation in the range of applied powers. The linear increase of the guided PL of Nd³⁺ excited by a non resonant excitation at 488 nm confirms the strong coupling between either Si-np and RE ions for SRSO or radiative defects and RE ions for nitride matrix. The guided fluorescences at 945 and 1100 nm were also observed until 4mm (SRSO) and 8 mm (SRSN) of the output of the waveguide and, of course, decreased when the excitation area moved away the output. So, waveguides based on Nd³⁺-doped SRSN, and notably SRSO because of its higher fluorescence decay, are very promising candidates on the way to achieve a laser cavity at 1.06 μm.

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Figure caption:

Figure 1: (a) Experimental and theoretical reflectance spectra of a 1100-nm-thick Nd³⁺-doped SRSO layer deposited on 8 μm-thick thermally grown SiO₂ on Si wafer and its schematic representation of the sample, (b) Experimental and theoretical reflectance spectra of a 500-nm-thick Nd³⁺-doped SRSN layer deposited on Si substrate and its schematic representation of the sample, (c) Evolution of refractive index of silica layer, Nd³⁺-doped SRSO layer and Nd³⁺-doped SRSN layer as a function of the wavelength obtained from reflectance spectrum (line) and by m-lines measurements (dots).

Figure 2: (a) Calculated number of modes as a function of the thickness of SRSO guiding layer deposited on 8 μm-thick thermally grown SiO₂ on Si wafer for TE and TM polarization at 1064 nm, and field mode profile for TE polarization and for SRSO thickness equal to 1.1 μm at 1064 nm (b) Calculated number of modes as a function of the thickness of SRSN guiding layer deposited on 8 μm-thick thermally grown SiO₂ on Si wafer for TE and TM polarization at 1064 nm, and field mode profile for TE polarization and for SRSN thickness equal to 0.4 μm at 1064 nm (The interface between air and guiding layer is localized for a thickness equal to zero meter).

Figure 3: (a) Schematic representation of the top surface pumping at 488 nm and near-field of guided PL at the output of planar optical waveguide, (b) Comparison of guided PL spectra for the three lines corresponding to ${}^4F_{3/2} - {}^4I_{9/2}$, ${}^4F_{3/2} - {}^4I_{11/2}$ and ${}^4F_{3/2} - {}^4I_{13/2}$ radiative transitions for Nd³⁺-doped- SRSO and SRSN planar waveguides obtained by top-pumping.

Figure 4: (a) Evolution of guided PL spectra by top surface pumping as a function of the pump power for Nd³⁺-doped- SRSO planar waveguides, inset: Integrated PL for the two lines

corresponding to ${}^4F_{3/2} - {}^4I_{9/2}$ (850 to 1000 nm) and ${}^4F_{3/2} - {}^4I_{11/2}$ (1000 to 1200 nm) radiative transitions as a function of the pump power for Nd³⁺-doped- SRSO planar waveguides, (b) Evolution of guided PL spectra by top surface pumping as a function of the pump power for Nd³⁺-doped- SRSN planar waveguides, inset: Integrated PL for the two lines corresponding to ${}^4F_{3/2} - {}^4I_{9/2}$ (850 to 1000 nm) and ${}^4F_{3/2} - {}^4I_{11/2}$ (1000 to 1200 nm) radiative transitions as a function of the pump power for Nd³⁺-doped- SRSN planar waveguides

Figure 5: (a) Schematic representation of the top surface pumping at 488 nm, (b) Evolution of guided PL spectra as a function of z, the distance between the excitation area and the output of the waveguide for Nd³⁺-doped- SRSO planar waveguides, inset: Integrated PL for the two lines corresponding to ${}^4F_{3/2} - {}^4I_{9/2}$ (850 to 1000 nm) and ${}^4F_{3/2} - {}^4I_{11/2}$ (1000 to 1200 nm) radiative transitions for Nd³⁺-doped- SRSO planar waveguides, (c) Evolution of guided PL spectra as a function of z, the distance between the excitation area and the output of the waveguide for Nd³⁺-doped- SRSN planar waveguides, inset: Integrated PL for the two lines corresponding to ${}^4F_{3/2} - {}^4I_{9/2}$ (850 to 1000 nm) and ${}^4F_{3/2} - {}^4I_{11/2}$ (1000 to 1200 nm) radiative transitions for Nd³⁺-doped- SRSN planar waveguides.

Figure 6: (a) Guided fluorescence decays of ${}^4F_{3/2} - {}^4I_{11/2}$ radiative transition excited at 444 nm by top surface pumping of Nd³⁺-doped- SRSO planar waveguides, (b) Guided fluorescence decays of ${}^4F_{3/2} - {}^4I_{11/2}$ radiative transition excited at 444 nm by top surface pumping of Nd³⁺-doped- SRSN planar waveguides.

Table 1: Refractive indices for different wavelengths and for the different films

λ (nm)	SiO ₂	SRSO	SRSN
1550	1.444	1.536	2.078
1340	1.447	1.536	2.080
1064	1.450	1.543	2.085
945	1.451	1.546	2.089
633	1.457	1.559	2.111
488	1.463	1.576	2.140

Table 2: Physical characteristics of SRSO and SRSN films

	SRSO	SRSN
Roughness (nm)	≈ 3	≈ 3
Optical losses at PIR (dB/cm)	≈ 1	≈ 1