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Nonlinear Optical Properties of Si Nanocrystals

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ABSTRACT

A systematic study of nonlinear optical properties of silicon nanocrystals (Si-nc) grown by plasma enhanced chemical vapor deposition (PECVD) is reported. Nonlinear optical refraction and absorption have been measured by z-scan technique at three different time regimes and at different wavelengths to investigate both the thermal and electronic responses. For this purpose three different laser sources have been used. Different behaviors, as expected from the theory, for different pump pulse durations are observed.

INTRODUCTION

Nonlinear photonic materials are widely used in many key-devices for the telecom industry such as switches, routers, wavelength converters. As an example, optical logic gates realized with nonlinear Mach-Zehnder interferometer (MZI) offer a very attractive feature for mass-manufacturing such as scalability and flexibility. Silicon nanocrystals (Si-nc) are widely studied because their efficient visible emission [1] and they have been already demonstrated as a very promising material for nonlinear applications both in the form of porous Silicon and Si-nc embedded in SiO_2 [2, 3] offering the opportunity to be exploited in Si photonics [4]. One of the main advantages of using Si-nc in SiO_2 is their full process-compatibility with mainstream CMOS technology.

Previous studies have demonstrated a large nonlinear optical response of Si-nc in the range of visible wavelength down to 800 nm, but a systematic study of non linear properties of Si-nc at 1500 nm is not yet known in literature [2, 3].

Different physical mechanisms can contribute to Si-nc optical nonlinearities, which are differentiated by their response time. In order to characterize the material for the application in all-optical gates, it is of fundamental importance to distinguish among the different contributions. Here we report a comprehensive study on the different physical mechanisms contributing to Si-nc optical nonlinearities. It is found that the fast nonlinear response is increased by one order of magnitude with respect to that of bulk silicon.

THEORY

The optical nonlinearities in semiconductors are a combination of bound electronic, free carrier and thermal effects. Bound electronic response involves a distortion of the electronic cloud about an atom by the optical field. If the atom is highly polarizable, it can exhibit a significant electronic nonlinearity (n_{2be}) [5]. This kind of response is very fast; in fact it follows instantaneously the field.

Interband transitions are possible when the frequency of the incident radiation is near a resonant energy transition. Single or two photon absorption processes can excite free carriers in a semiconductor. In turn these free carriers absorb the incident radiation: an effect which is related by Kromers-Kronig relation to a change of the refractive index. Thus excitation via one or two photon absorption of a significant population gives an additional change in the refractive index (n_{2fr}).The induced free-carrier refraction depends on the density of photogenerated carriers (ΔN) produced by the photon absorption and occurs on a time scale typical of free carrier recombination in semiconductors [6].

The increase of the sample temperature leads to a thermal nonlinear refraction (n_{2th}). There are two situations for which these thermal effects can be of importance. The first is when long pulses are used (~ ns), the second when a high repetition rate (> 10 kHz) can produce cumulative effects [7].

Finally, the nonlinear response n_2 of a semiconductor is the result of these terms: $n_2 = n_{2be} + n_{2fr} + n_{2th}$. To separate the various contributions the main method is to perform experiments at different excitation temporal regimes: with low repetition rate and fast pulses one emphasizes the bound electronic contribution, with high repetition rate or long pulses one emphasizes the others.

Table I: Structural parameters of 1250°C annealed samples

Sample	Si(%)	O (%)	N (%)	*n*, *k* @ 830 nm	*n*, *k* @ 1550 nm	Si-nc mean size (nm)
G01	34	66	-	1.47, $< 10^{-4}$	1.46, $\sim 1.6\text{x}10^{-5}$	-
G02	40	54	6	1.50, $< 10^{-4}$	1.48, $\sim 1.6\text{x}10^{-5}$	< 1
G03	48	44	8	1.93, 0.008	1.91, $\sim 1.6\text{x}10^{-5}$	3.9 ± 0.8
G04	51	43	6	1.99, 0.020	1.95, $\sim 1.6\text{x}10^{-5}$	4.8 ± 1.5

EXPERIMENTAL

A set of SiO_xN_y layers (600 nm thick) was deposited by plasma enhanced chemical vapor deposition (PECVD) onto transparent silica substrates with different values of Nitrogen and Silicon (Table I). The presence of Nitrogen is a consequence of the use of N_2O as one of the gaseous precursor in the deposition process. Silicon nanocrystals were formed by a high temperature annealing process in N_2 atmosphere. Nonlinear transmission z-scan experiments were performed as function of the silicon content and the annealing temperature. Three kinds of pulsed sources were used in the measurements: i) a Ti:Sapphire laser pumped optical parametric generator, $\lambda = 1550$ nm, 1 kHz repetition rate with 100 fs pulse duration, peak intensity $I_0=6\text{x}10^{11}\text{W/cm}^2$ ii) a Ti:Sapphire laser, $\lambda = 830$ nm, 82 MHz repetition rate with 70 fs pulse duration, peak intensity $I_0=1.15\text{x}10^{10}$ W/cm^2 and iii) a Nd:YAG laser pumped optical parametric oscillator, $\lambda = 1550$ nm, 10 Hz repetition rate with 4 ns pulse duration, $I_0=10^8\text{-}10^9\text{W/cm}^2$.

Whereas in the cases of 82 MHz repetition rate and of nanosecond pulse duration we have nonlinear contribution from all the three different effects, in the case of fs pulses with 1 kHz repetition rate we have only bound electronic nonlinearities. The transmission of the laser beam through the sample is recorded while a computer driven continuous motor moves the sample along the optical path (*z* axis). The transmission through the sample is monitored by a photodiode (D_1). An aperture is placed in front of D_1 for closed aperture measurements. A small portion of the input beam was monitored by another photodiode (D_2) and the ratio (D_1/ D_2) is recorded as a function of the sample position, *z* (Figure 1). The transmission with and without the aperture was measured in the far field as the sample moved through the focal point, enabling the separation of the nonlinear refractive index from the nonlinear absorption [2, 3].

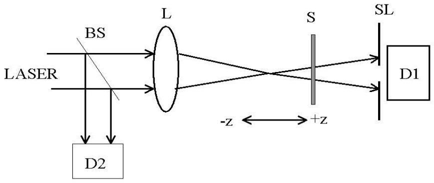

Figure 1: Schematics of the experimental set-up used for the z-scan measurements [2].

DISCUSSION

Measurements at 1.55 µm with fs pulses

Representative experimental data are shown in Figure 2 for samples G02 and G03 annealed at 1100°C. It was found a positive z-scan trace. Repeated measurements on all the samples show similar results. Thus the nonlinear refraction is positive for all the Si-nc samples. This kind of nonlinearity is due to the bound electronic response. The nonlinear refraction values are estimated of the order of 10^{-13} cm^2/W. In addition a nonlinear absorption is also observed, whose nonlinear absorption (β) values are in the range of - ($10^{-9} \div 10^{-8}$) cm/W. The presence of a strong nonlinear absorption, for the samples with higher Silicon content (G03, G04), distorts the measured closed aperture z-scan trace; in fact the nonlinear absorption enhances the valley and reduces the peak height [8], as shown in Fig. 2 (c ,d).

Our measurements show that the nonlinear refractive index is higher for higher linear refractive index, as shown in Figure 3. The linear refractive index is related to the Si-nc sizes and density, thus the nonlinearities increase as the Si-nc size and density increase. This effect is in contrast with expectations dictated by quantum confinement effects, where it is predicted that the nonlinearities increase as the nanocrystal size decreases. A probable explanation is that at 1500 nm the nonlinearities are also affected by the dielectric mismatch between the Si-nc and the oxide. The local electric field experienced by the nanocrystals is enhanced compared to the incident field because of the dielectric mismatch. Local field enhancement factor Q_{NF} can be calculated with the Lorenz-Mie scattering theory [9]. The effective third order susceptibility $\chi^{(3)}$ is enhanced by a factor $(Q_{NF})^{3/2}$ due to the local field effect. Since Q_{NF} increase with the increase of the nanocrystals size, the effective $\chi^{(3)}$ increase too. The relative importance of the two effects (dielectric mismatch and quantum confinement) is weighted by the energy at which the nonlinearities are measured. For infrared light, the nonlinearities seem more influenced by dielectric mismatch.

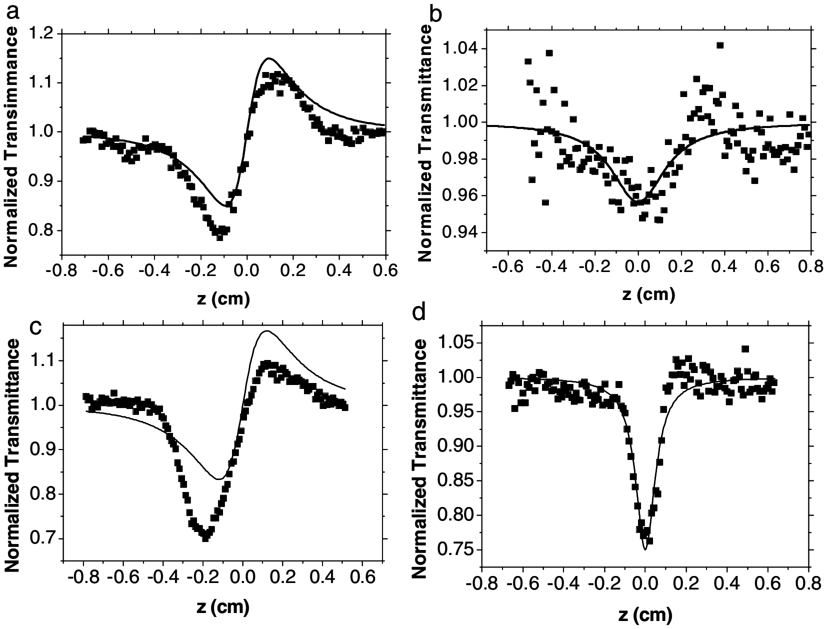

Figure 2: Representative z-scan traces for G02 closed (a) and open (b) aperture and for G03 closed (c) and open (d) aperture. Both the samples are annealed at 1100 °C. The theoretical fitting is also included as a solid curve.

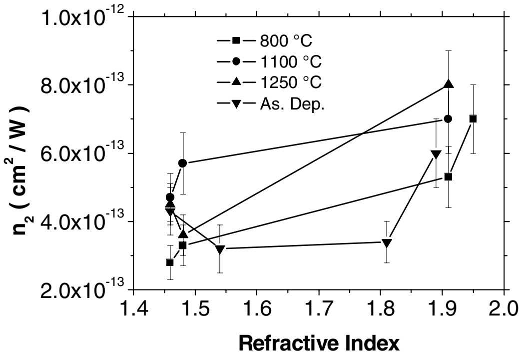

Figure 3: Nonlinear refractive index as a function of the linear refractive index for different annealing temperature (100fs, 1kHz, 1550 nm).

Measurements at 0.83 μm with fs pulses at 82 MHz

The nonlinear refraction values are in the range of ($10^{-13} \div 10^{-12}$) cm^2/W for all the samples, and the nonlinear absorption values are of the order of -10^{-8} cm/W. Some samples show a negative nonlinear refractive index, probably due to the superposition of thermal cumulative (high repetition rate) and free carrier effects (830 nm is near the absorption edge). Figure 4 reports the dependence of n_2 as a function of the linear refractive index.

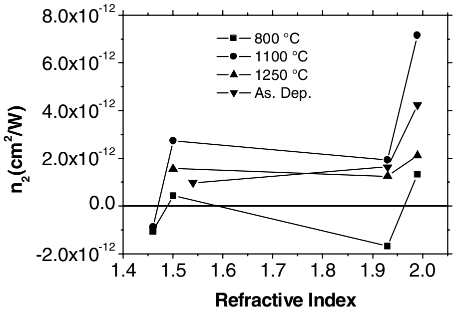

Figure 4: Nonlinear refractive index as a function of the linear refractive index for different annealing temperature (70fs, 82MHz, 830 nm).

Measurements at 1.55 μm with ns pulses at 10 Hz

Z-scan measurements for all the samples have shown negative nonlinearities associated to thermal effects, measured values are in the range of - ($10^{-9} \div 10^{-8}$) cm^2/W.

Figure of merit

The material properties with respect to device performances can be assessed by the introduction of the nonlinear figure of merit $F = n_2 / \beta\lambda$ [10]. The estimation of F for our samples have evidenced that the Si-nc demonstrate always higher values respect to that reported for bulk Si and bulk GaAs [10] as showed in Figure 5. The highest F value is achieved with low size silicon nanocrystals because of their lower nonlinear absorption coefficient.

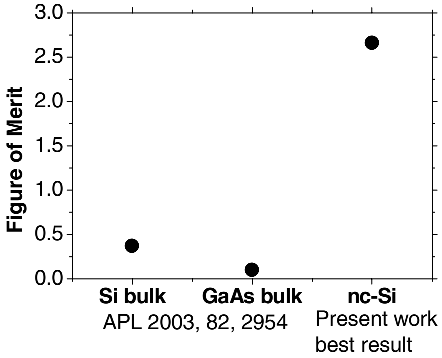

Figure 5: Comparison between F from Si-nc, bulk Si and bulk GaAs at 1550 nm [10].

CONCLUSIONS

A systematic study of the optical nonlinearities of Si-nc was performed in three different excitation regimes to separate and evaluate the different contributions to the nonlinear refraction. Positive nonlinearities were found for all the samples in fs and low repetition rate regime, due to pure bound electronic response. Negative nonlinearities were found for all the samples in the ns regime, ascribed to thermal effects. The study of bound electronic nonlinearities revealed the increase of nonlinear response with respect to the increase of nanocrystals size, probably due to the dielectric mismatch between the Si-nc and the oxide embedding medium. The analysis of the nonlinear figure of merit has demonstrated that the Si-nc is a promising material for application in optical logic gates. In particular small size silicon nanocrystals have demonstrated the best features, in terms of figure of merit, due to their low nonlinear absorption properties.

ACKNOWLEDGMENTS

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