Size dependence of nonlinear optical response in Si-nanocrystals

S. Hernández,[1] A. Martínez,[1] O. Jambois,[1] P. Pellegrino,[1] P. Miska,[2] M. Grün,[2] H. Rinnert,[2] M. Vergnat[2] and B. Garrido[1]

[1] MIND-IN2UB, Departament d'Electrònica, Universitat de Barcelona, Martí i Franquès 1, 08028 Barcelona, Spain.
[2] Institut Jean Lamour, UMR CNRS 7198 – Nancy Université – UPV Metz, France.

Contact e-mail: shernandez@el.ub.es

Silicon nanocrystals (Si-nc) embedded in oxide matrices have been proposed as active material for nonlinear photonic applications, as their nonlinear optical properties were found to be larger than the ones in silica or Si. A strong size dependence of their nonlinear optical properties has been observed by different authors, nevertheless they show a large scatter in their data [1, 2]. Therefore, an accurate knowledge of their nonlinear optical properties as a function of the Si-nc size is crucial for the conception and design of highly efficient new photonic structures and the control of their performance.

Here we present a *z*-scan study of SiO/SiO_2 multilayers exciting in the ns-regime with pulses of $\lambda = 1064$ nm. Films were deposited by evaporation and annealed up to 1000 °C in a conventional furnace for 10 minutes. As a consequence, Si-nc were precipitate with sizes from 2 nm up to 5 nm. In Fig. 1 we present an energy filtered TEM image of a typical sample with Si-nc of 3 nm. A high density of Si-nc can be observed homogeneously distributed along the films. Similar results were also obtained in the whole set of samples. Raman measurements have revealed a sharp feature around 518 cm^{-1} indicating that the Si-precipitates have high crystal quality. A linear optical characterization of the layers has been performed by means of photoluminescence and optical absorption, finding that the energy emission and the band increase as the Si-nc size is reduced.

The nonlinear absorption and nonlinear refractive index have been measured by z-scan experiments using a ns-pulsed Nd:AG laser working at $\lambda = 1064$ nm. The z-scan traces have shown a position dependence transmittance in both the open and close aperture configurations, indicating that both absorptive and refractive nonlinearities arise in the Si-nc/SiO_2 system under these excitation conditions. The films have been analyzed in open and close aperture configurations, in order to determine both non-linear absorption and non-linear refractive index in the ns-regime. Using expression developed by Sheik-Bahae [3], we fitted the experimental from open and close aperture configurations and we extracted the nonlinear absorption coefficient and the nonlinear refractive index of the SiO/SiO_2 multilayer system.

In Fig. 2 we present the nonlinear absorption coefficient and nonlinear refractive index for SiO/SiO_2 multilayered samples with different Si-nc size. The nonlinear absorption coefficient and the nonlinear refractive increase from 1.0×10^{-6} to 1.8×10^{-5} cm/W and from -4.2×10^{-11} to -7.6×10^{-10} cm^2/W, respectively, as the Si-nc increases from 2 to 5 nm. A similar increasing trend with the Si-nc size can be observed when the nonlinear response is normalized to the Si excess, as shown in the inset of Fig. 2, in contrast to previous studies performed in Si-nc where a reduction of the nonlinear optical response is observed for larger Si-ncs [1].

In order to determine the origin of the observed nonlinear behavior in the Si-nc/SiO_2 multilayered system, we have analyzed their nonlinear optical response using a nonlinear model which includes the changes in the free-carrier concentration and possible thermal contributions induced by the high excitation fluencies. In this time domain (ns), we found that the excitation of free carriers is the main mechanism contributing to the nonlinear optical response.

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References

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Figures

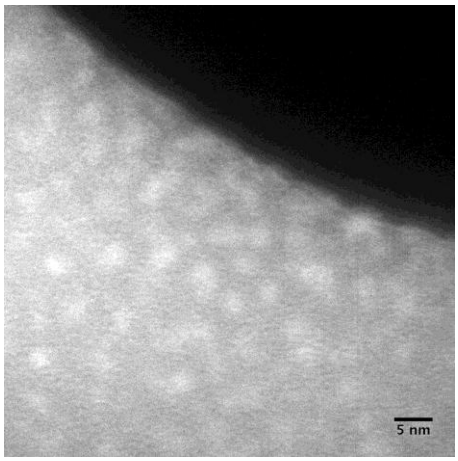

Figure1. EFTEM images of sample with Si-nc of 3 nm. The light contrast is referred to Si-nanoparticles.

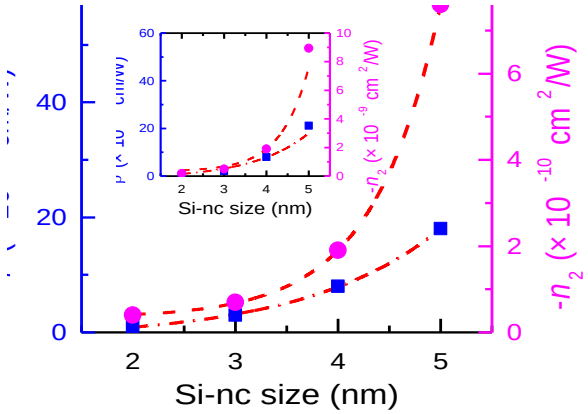

Figure 2. Nonlinear absorption coefficient (left axis, in blue) and nonlinear refractive index (right axis, in magenta) of the SiO/SiO_2 multilayered system as a function of the Si-nc size.). In the inset, β and n_2 as a function of the Si-nc size, once normalized by the Si excess in the samples.