Silicon nanocrystals to enable silicon photonics

Invited Paper

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Low dimensional silicon, where quantum size effects play significant roles, enables silicon with new photonic functionalities. In this short review, we discuss the way that silicon nanocrystals are produced, their optoelectronic properties and a few device applications. We demonstrate that low dimensional silicon is an optimum material for developing silicon photonics.

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1. Introduction

Silicon photonics is the technology where photonic devices are produced by standard microelectronics processes by using the same paradigm of electronics: integration of a large number of devices to yield a high circuit complexity which allows high performances and low costs[1]. Thus, the real truth is to develop photonic devices that can be easily integrated to improve the single device performance and to allow high volume production: integration and mass-manufacturing are where silicon photonic can outperform other photonic platforms, such as InP-based or glass-based ones. To this aim, one further add-on is making silicon do something where it is not able to do in its standard (bulk) form. Low dimensional silicon, where small silicon nanocrystals or nanoclusters (Si-ncs) are developed, is one way to compel silicon to act as an active optical material[2]. In this short review, we will go through a few applications where Si-nc enables silicon doing photonic functions otherwise not possible by using bulk silicon.

Figure 1 reports the number of results one gets if looks for Si-nc in Google^{TM} Scholar. It is observed a steady rise in the number of publications witnessing the rising interest in this material. The first results found in this search refer to four papers reporting about different properties of Si-nc: the first is our paper on the observation of optical gain[3], the second is the paper by Tiwari *et al*. on the use of Si-nc for memories[4], the third is the paper by Wilson *et al*. on the demonstration of quantum size effects in Si-nc[5], and the fourth is the paper by Mutti *et al*. on the observation of room temperature luminescence in Si-nc[6]. While if one makes the same search on Google^{TM}, the first result concerns the use of Si-nc for photovoltaics[7]. All the key ingredients that make Si-nc appealing for photonics are discussed in these papers: quantum size effects make new phenomena appear in silicon, such as room temperature visible photoluminescence, optical gain, Coulomb blockade, and multiexciton generation. In this short review, we will discuss these topics and see how they can be exploited in working devices. For more detailed information, we refer readers to other reviews that we wrote in the past[8−11].

2. Silicon nanocrystals: production

Silicon nanocrystals are produced by a wealth of different techniques[12]. They can be essentially distinguished into three types: direct synthesis, phase separation in a silicon-rich dielectric, and top-down production from bulk silicon. Examples of the first type are cluster deposition[13] or chemical synthesis in a solution[14]. The second class of techniques is more widely diffused and essentially is based on the production of a silicon-rich dielectric (e.g., silicon oxide or silicon nitride) and on the phase separation of the constituent phases (silicon and the dielectric) by an annealing treatment. The duration and temperature of the annealing treatment determine the size and crystallinity of the nanoparticles. Various methods can be used to produce the starting silicon-rich dielectric: ion implantation[6], sputtering[15], chemical vapor deposition (CVD)[16], sol-gel synthesis[17], etc. The third class of techniques is the one used in the production of porous silicon (electrochemical etching) where silicon is partially dissolved and the remaining skeleton is composed of interconnected Si-ncs[18]. Alternatively, lithographic technique followed by repeated oxidations on a silicon wafer can define small silicon islands or columns where Si-ncs are formed[19]. This is useful for single Si-nc production, or when the geometrical arrangement of Si-nc has to be controlled.

The preparation method strongly influences electrical

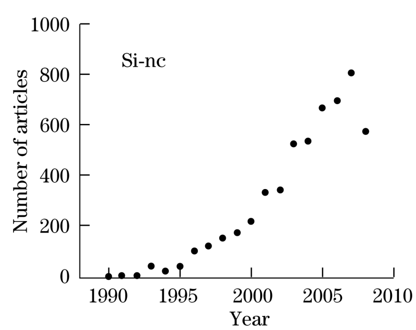

Fig. 1. Number of articles versus year as reported by Google^{TM} Scholar in a search performed on 31/12/2008. The search keys were "low dimensional silicon" OR "silicon nanocrystal/s" OR "silicon nanocluster/s". The total number of articles integrated over the years is 6220, while the results for "silicon photonics" is 1740 and for "porous silicon" is 28800. The single point referred to year 2008 is low due to the time delay between the search date and the actual database construction.

and optical properties of Si-nc. Consider the case of Si-nc formed in a dielectric by phase separation. Figure 2 gives a sketch of Si-ncs dispersed in a dielectric matrix. Figure 2(a) shows the ideal case where all Si-ncs have the same diameter and shape, orderly arranged in space. In addition, the dielectric has a homogeneous composition and the interface between the Si-nc and the dielectric is atomically sharp. Figure 2(b) shows the real case where the single Si-nc has a layered structure characterized by an inner crystalline core and a diffused interface[20]. The Si-ncs are characterized by a distribution of Si-nc sizes, shapes, and inter-dot distances[21]. Moreover, the composition of the dielectric is strongly inhomogeneous with various localized defects which can be either due to excess Si atoms still remaining in the host (about 50% of all the Si atoms in excess with respect to the stoichiometric value is still in the dielectric after annealing treatment[22]) or host based (e.g., vacancies). And the interface between the crystalline core of the Si-nc and the dielectric is diffused over a distance of roughly 0.5 nm and is composed by a strongly stressed dielectric whose composition is still under debate[20].

Most of the authors refer to Si-nc as nanocrystals, while it should be noted that in a few applications what is more relevant is not the use of nanocrystals but of nanoclusters. The term cluster is usually used to indicate the amorphous nature of the nanoparticle. The difference between nanocrystals and nanoclusters is quite subtle and sometimes semantic. One can say that nanoclusters are characterized by the lack of regular lattice planes when observed by transmission electron microscopy (TEM) or by the lack of a distinct longitudinal optical (LO) phonon peak at about 520 cm^{-1} when observed by Raman spectroscopy or are characterized by a broad (extending even below the silicon band gap) and weak emission when observed by photoluminescence.

3. Silicon nanocrystals: linear optical properties

Figure 3 shows a qualitative diagram of the dependence of the band gap, emission intensity, and refractive index on the Si-nc size. It is observed that quantum confinement effect causes an enlargement of the band gap with respect to that of bulk silicon[2]. The smaller the Si-nc is, the larger the band gap is. Consequently, it is possible by using properly sized Si-ncs to cover a wide spectral range, all across the visible. What is remarked from the start is that Si-ncs have strong emission intensity at room temperature. This is a result of three main facts: firstly, an increased electron and hole wavefunction overlap in the momentum space as a result of quantum confinement of electrons and holes in small Si-nc; secondly, the spatial localization of excited free carriers in a small region where the probability to find a non-radiative recombination center is low; thirdly, the reduced effective refractive index which allows more light to escape from the material. Quantum confinement effects also explain the reduced lifetime of excitons in Si-nc: microsecond level for Si-nc and tenths of milliseconds for Si. Figure 3 also shows that the emission intensity peaks for average size Si-nc[23]. Large Si-ncs have low emission intensity due to long lifetime; small Si-ncs have low emission intensity due to an increased escape probability of excitons which tend to spill-over into the dielectric.

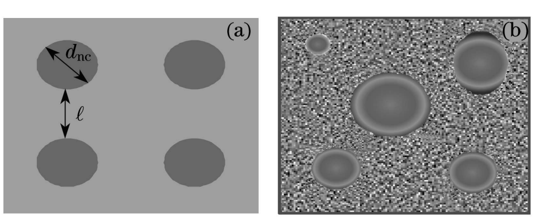

Fig. 2. Qualitative sketch of (a) the ideal case and (b) the real case. $d_{\rm nc}$ is the diameter of the Si-nc while ℓ is the inter-dot distance.

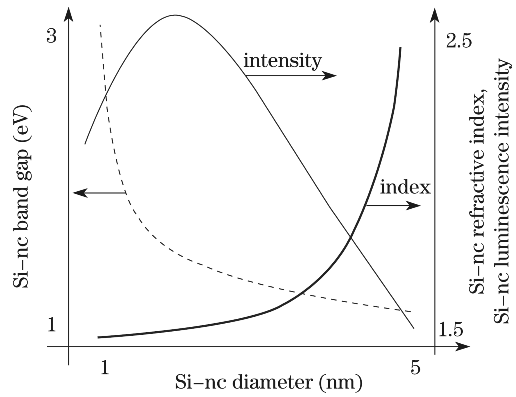

Fig. 3. Qualitative sketch of the dependence of the Si-nc band gap, the Si-nc luminescence intensity, and the refractive index versus the Si-nc diameter. The numerical values refer to Si-nc formed in SiO_2.

The overall scheme shown by Fig. 3 can be used to describe the general phenomenology for Si-nc synthesized by different methods. The details are however different since the dielectric matrix affects strongly the recombination dynamics of Si-nc.

Figure 3 also shows the trend of the refractive index. The smaller the Si-nc is, the lower the refractive index is. The actual value depends strongly on the dielectric in which Si-ncs are formed. In a simple scheme, the refractive index can be considered as an average of the one of the matrix and the one of silicon weighted by the volumetric fraction of the two components. In reality, the situation is more complex and the size of Si-nc is also relevant[24].

Figure 4 shows the phenomenology of the emission and absorption spectra of Si-nc[10]. Looking at the emission, it is observed that Si-ncs emit a wide spectrum peaked in the visible region. The large emission width is the sum of the indirect nature of the recombination[19] and of the wide size distribution. The spectrum is wider when the

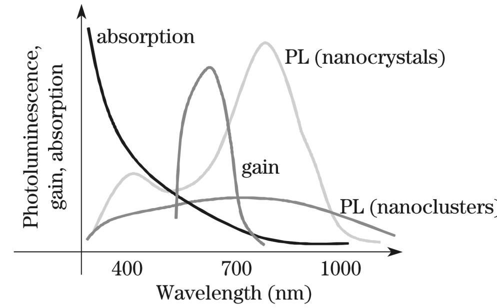

Fig. 4. Linear optical spectra of Si-nc of a typical size of 3 nm and formed in SiO_2. The two luminescence spectra refer to the emissions from nanocrystals and nanoclusters.

nanoparticles are still amorphous as in silicon nanoclusters while it is narrower when they are nanocrystals. In addition, emission can be observed at short wavelengths due to recombination in defects present in the matrix, mostly vacancies. Small Si aggregates emit at short wavelengths: Si-ncs smaller than 1 nm have blue emission which is however instable with respect to recombination in interface defects[25]. Room temperature visible emission paves the way to application of Si-nc in lightening both as phosphors and as light emitting diodes. External quantum efficiency of Si-nc as high as 60% has been reported at 790 nm[26].

The absorption is blue-shifted with respect to emission due to the indirect nature of the Si-nc band gap. Defining the absorption coefficient $\alpha = \sigma_{\mathrm{abs}} N_{\mathrm{NC}}$, where N_{NC} is the Si-nc density and σ_{abs} is the Si-nc absorption cross section, one finds that $\sigma_{\mathrm{abs}} = 3.5 \times 10^{-18}$ cm^2 at 830 nm[27].

Since the refractive index of Si-nc in a dielectric is larger than the one of SiO_2, these two materials can be used to form waveguides. Figure 5 shows various geometries used to produce waveguides. Low loss waveguides can be realized where the propagation loss can be lower than 2 dB/cm at 1.55 μm[27]. In this region, losses are due to scattering losses caused by Mie scattering and waveguide surface roughness. At shorter wavelengths, losses due to absorption start to be relevant[28].

As the density of excited carriers in Si-nc increases, another power dependent loss mechanism settles in: excited carrier absorption[29]. This depends on the density of excited carriers and can be described by $\alpha_{\mathrm{xc}} = \sigma_{\mathrm{xc}} N_{\mathrm{xc}}$, where σ_{xc} is the excited carrier absorption cross section and N_{xc} is the density of excited carriers. For Si-nc and at $\lambda = 1.54$ μm, $\sigma_{\mathrm{xc}} = 4 \times 10^{-19}$ cm^2, that is when one carrier is excited per nanocrystal, $\alpha_{\mathrm{xc}} \sim 1$ cm^{-1}.

Due to the low loss of Si-nc based waveguides, more interesting structures can be produced such as ring resonators, optical cavities or microdisks. For microdisks, quality factor as high as 3000 has been demonstrated at around 800 nm which makes this structure very interesting for lasing applications[30].

Another interesting application of Si-nc is to activate the emission of other species, such as rare-earth impurities. In fact, the coupled system of Si-nc and Er represents a very interesting alternative for fabrication of infrared emitter where the active species is Er while Si-nc plays the role of sensitizer of Er luminescence[2].

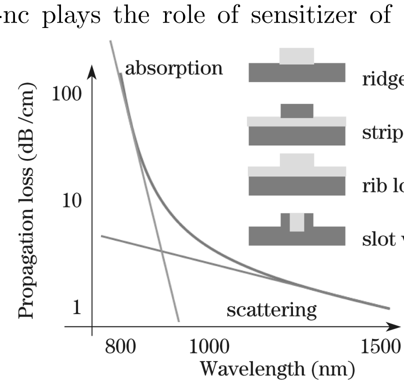

Fig. 5. Propagation losses in a Si-nc waveguide. The inset shows different geometries that can be used to form the waveguides, where the dark layer is formed by SiO_2 while the grayish layer is formed by Si-nc embedded into a dielectric matrix.

Absorption of the excitation occurs in Si-nc which in turn transfers the excitation to Er by a mechanism which is still under debate[31]. The Er/Si-nc system can be realized in the form of a waveguide for all-optical amplifier applications[32].

4. Silicon nanocrystals: nonlinear optical properties

Heavily photoexcited Si-nc shows several interesting nonlinear optical properties which widen even further the scope of its applications. Here we detail two main effects: optical gain[3] and Kerr-based nonlinearities[33].

Figure 4 shows the gain spectrum measured in Si-nc when pulsed laser excitation is used[34]. The main characteristics of light amplification in Si-nc are that it is fast (a few nanoseconds), and occurs at longer wavelengths than absorption and at shorter wavelengths than the typical emission. The first characteristic is due to the delicate balance among stimulated emission and other nonlinear effects such as excited carrier absorption and Auger recombination. The spectral position difference between absorption and gain is explained by assuming a four-level model for the gain[35]. The blue shift of the gain spectrum with respect to the spontaneous emission is still under debate. It could be due to the fact that spontaneous emission occurs in large Si-nc while stimulated emission in small Si-nc. Or alternatively, that stimulated emission is due to active centers different from the Si-ncs responsible for spontaneous emission. These active centers can be either interface radiative centers due to Si=O double bonds[35] or small Si aggregates in the matrix[36]. It should be noted that gain has been observed in Si-ncs embedded into SiO_2 but not when they are embedded into Si_3N_4[37]. The measured value for the gain coefficient is few cm^{-1} which allows to use Si-ncs as active materials into an optical cavity to achieve lasing.

High excitation effects are also observed by a nonlinear dependence of the optical constants of Si-nc. In fact, one can consider the refractive index (n) and the absorption coefficient (α) as $n = n_0 + n_2 I$ and $\alpha = \alpha_0 + \beta I$, where n_0 and α_0 are the linear terms discussed in the previous section, while n_2 and β are the nonlinear refractive index and the nonlinear absorption coefficient, respectively, and I is the intensity of the pump beam. Nonlinear refraction is of the Kerr-type when it involves a direct polarization of the electronic clouds around an atom caused by the pump beam. This mechanism is intrinsically very fast. However, in a semiconductor, other kinds of nonlinearities are present due to thermal lensing effects and free carrier effects. Measurements on Si-nc have demonstrated that all these kinds of nonlinearities are present and one can distinguish them by using different lengths of the excitation light pulses[33]. For very short (femtosecond) light pulse, Kerr nonlinearities dominate. In this case, the nonlinearities are due to the polarization of the strained bonds at the interface of the Si-nc and the dielectric matrix. Also quantum effects due to carrier confinement in the Si-nc cannot be discarded. Nonlinear absorption effects in Si-nc are mostly due to two-photon absorption. They are particularly detrimental because, when two-photon absorption is present, it generates a large amount of excited carriers which in turn absorb light by excited carrier absorption but also

Table 1. Comparison of Nonlinear Optical Properties of Several Materials at 1550 nm

	Si	Si-nc (PECVD)	Si-nc (LPCVD)	SiO_2	GaAs	$Al_{0.18}Ga_{0.82}As$	Polymer
n_2 (cm^2/W)	4×10^{-14}	8×10^{-13}	2×10^{-12}	2.5×10^{-12}	1.6×10^{-13}	1×10^{-13}	1.7×10^{-13}
β (cm/GW)	0.8	20	70	0	10	<0.3	not Available
$F = n_2/\beta\lambda$	0.35	0.25	0.19	>10	0.1	2	not Available
$\gamma = n_2/(\lambda A_{\text{eff}})$ $(\text{W}\cdot\text{cm})^{-1}$	0.26	10	26	0.003	0.34	0.21	2.2

n_2 is the nonlinear refractive index, β is the nonlinear absorption coefficient, F is the figure of merit for nonlinear application, λ is the wavelength, γ is the waveguide nonlinear parameter, and A_{eff} is the optical mode cross section in the waveguide. For Si-nc, two different deposition methods have been studied (PECVD, plasma enhanced CVD, and LPCVD, low pressure CVD).

but also cause, via a Kramers-Kronig transformation, excited carrier refraction which affects n_2.

Table 1 shows a comparison of the results of different semiconductors where it is evident that Si-nc is a very interesting system for applications in nonlinear integrated optics since the waveguide nonlinear parameters are larger than those of other semiconductors or polymers. In fact, an application of Si-nc in fast all-optical switching has been demonstrated with switching speed as fast as a few picoseconds which allows its use for >100 Gb/s all-optical networks[38].

5. Silicon nanocrystals: optoelectronic properties

Photoluminescence is quite intense in Si-nc, while electroluminescence is, on the contrary, quite weak. Best results for the power efficiency of Si-nc based light emitting devices (LEDs) are about 0.37% in oxidized porous silicon[39] and 0.15% in Si-nc at room temperature[40]. Figure 6 shows the two main tunneling mechanisms for charge injection: Fowler-Nordheim tunneling and direct tunneling. The first is characteristic of high applied bias and produces electroluminescence by impact excitation. It is characterized mostly by unipolar injection where a single kind of carrier is injected, usually electrons. The second is characteristic of low applied bias and produces electroluminescence by direct injection of both electrons and holes into the Si-nc. This is strongly affected by size dispersion and by inter-dot distance variation.

Figure 7 shows the comparison between two LEDs which have a 50-nm-thick single layer of Si-nc or a 50-nm-thick multilayer structure as active region[40], respectively. The multilayer structure is formed by a periodic sequence of 2-nm-thick SiO_2 layer, and of 3-nm-thick Si-nc rich layer. The aim of the multilayer structure is to control the size and the inter-dot distance in the vertical direction (i.e., the direction of current flow). The injected current and the electroluminescence intensity are reported versus the applied electric field. It is observed that the single layer structure has a larger injected current but a lower electroluminescence intensity than the multilayer sample. This directly shows the role of the size and inter-dot distance dispersions which strongly obstruct direct tunneling in favor of Fowler-Nordheim tunneling. Most of the current in the single thick Si-nc layer flows through parasitic shunting paths formed by defects in the dielectric. As a result, the multilayer sample, where injection is by direct tunneling, has higher electroluminescence at lower bias than the single homogenous thick layer.

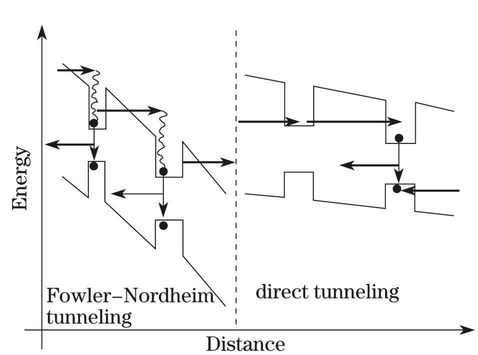

Fig. 6. Two main tunneling mechanisms for charge injection: Fowler-Nordheim tunneling and direct tunneling.

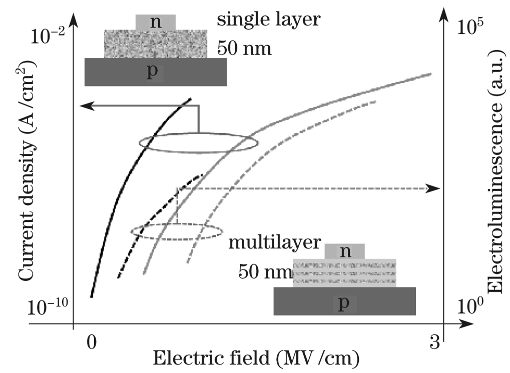

Fig. 7. Comparison between two LEDs with a 50-nm-thick single layer of Si-nc or a 50-nm-thick multilayer structure as active region, respectively.

Another interesting application of Si-nc is in photovoltaics[7]. By using a layer of Si-nc between the collecting electrodes in a solar cell, we show that a multiplication stage is realized due to secondary carrier generation in the Si-nc[41]. This is an intrinsic mechanism caused by infrared absorption and hot carrier injection. Figure 8 shows the mechanism. Visible light is absorbed by the standard silicon cell, and electrons which flow to the n-type electrode are injected through a Fowler-Nordheim tunneling into the Si-nc. Here carriers are generated in the interface states by infrared light absorption. The injected carriers loose the excess energy by exciting other carriers from the trap states into the Si-nc. These in turn participate in the current and the mechanism is reactivated. As a final result, a single photoexcited carrier yields shorter circuit current, hence a

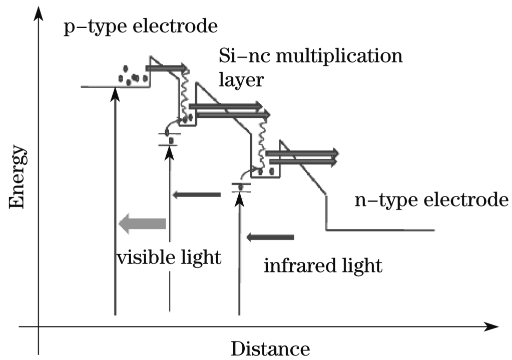

Fig. 8. Qualitative sketch of the mechanism of internal multiplication (secondary carrier generation) in our Si-nc solar cell.

larger conversion efficiency in the cell is achieved.

6. Conclusion

In this short review, we have discussed a few interesting properties of Si-nc and demonstrated a few effects which are enabled by using Si-nc as the active material. We propose Si-nc as a common material for many different device applications not only in active electro-optic devices as laser or solar cells, but also in all-optical devices or in sensing applications. The physics of Si-nc is not yet completely clear, and a lot of room is still open for interesting new developments.

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