Light emitting devices based on Si nanoclusters: the integration with a photonic crystal and electroluminescence properties*

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We present the properties and potentialities of light emitting devices based on amorphous Si nanoclusters. Amorphous nanostructures may constitute an interesting alternative to Si nanocrystals for the monolithic integration of optical and electrical functions in Si technology. In fact, they exhibit an intense room temperature electroluminescence (EL). The EL properties of these devices have been studied as a function of current and of temperature. Moreover, to improve the extraction efficiency of the light, we have integrated the emitting system with a 2D photonic crystal structure opportunely fabricated by using conventional optical lithography to reduce the total internal reflection of the emitted light. The extraction efficiency in such devices increases by a factor of 4 at a resonance wavelength.

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Si nanocrystals embedded in a SiO_2 matrix are currently attracting a great interest as a candidate system to solve the physical inability of bulk Si. The band gap of Si nanocrystals is enlarged with respect to the bulk material due to quantum confinement effects and, as a consequence, the emission is shifted in the visible region. The demonstration that Si nanocrystals can exhibit optical gain[1] and that they can be used as the active layer in efficient light emitting devices[2-12] has greatly increased. Also amorphous Si nanoclusters (nc) have received a considerable attention as light emitting materials, and some reports about their photoluminescence (PL) properties[13], as well as theoretical studies on their electronic properties[14], have been published in the last years. However, if compared with the very large amount of already available data about Si nanocrystals, amorphous clusters are still a relatively unexplored material. In particular, although electroluminescence (EL) from amorphous nanostructures has been already demonstrated[2,15,16], the few reported experiments refer to low temperature data[16]. On the other hand, amorphous Si nc may represent an interesting alternative to nanocrystals for the development of Si-based light emitting devices, mainly because their use should allow to remarkably decrease the thermal budget needed for the nanostructure formation (amorphous Si nc are formed already at 900 °C [17]), allowing an easier integration of an optical source in an electronic device.

In this paper we present our results on the application of amorphous Si nc for the fabrication of light-emitting devices. A new device structure, based on the integration of the emitting system with a photonic crystal (PhC) will be also proposed. The optical properties of the devices will be presented and discussed, with a particular attention to their potentialities for practical applications in silicon optoelectronics.

SiO_x thin films were deposited on top of Si substrates (p-type Si (111) wafers of 10-20 mΩ cm resistivity) by using a parallel plate plasma enhanced chemical vapor deposition system, consisting of an ultra high vacuum chamber and a rf generator (13.56 MHz), connected through a matching network to the top electrode of the reactor; the bottom electrode is grounded and acts also as a sample holder. All deposition processes have been performed by using 50 W of input power. The source gases used were high purity SiH_4 and N_2O; the N_2O/SiH_4 flow ratio was set to obtain SiO_x films with a total Si concentration of 46 at.%. The film thickness is about 70 nm. After deposition, the SiO_x films were annealed at 900 °C

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for 1 h in nitrogen atmosphere in order to induce the separation of the Si and SiO_2 phases with the formation of amorphous Si nc embedded in SiO_2[17]. The annealed SiO_x films have been used as the active layer for the fabrication of MOS light emitting devices. In such devices the top electrode is a 300 nm thick n-type (as doped) polysilicon layer, having a resistivity of 1 mΩ cm, deposited by low pressure chemical vapor deposition; the bottom electrode is the p-type Si substrate. The same simple structure has been previously successfully employed for the realization of light emitting devices based on Si nanocrystals[8,9]. The active area of the devices ranges from 0.9×10^{-3} to 10×10^{-3} cm^2 and is metal-free to allow the exit of the light. The devices were also integrated with a PhC structure to increase the extraction efficiency of the light. The polysilicon layer acting as the top electrode has been etched with conventional optical lithography to produce a triangular two-dimensional PhC composed of holes having a diameter of 400 nm with a lattice constant of 900 nm. The depth of the holes is 200 nm, hence leaving a continuous 100 nm thick polysilicon layer for the electrical contact.

Scanning electron microscopy (SEM) analyses were performed by a field emission microscope Zeiss Supra 25. EL measurements were taken by biasing the device with a square pulse at a frequency of 55 Hz, using a fast Agilent Pulse Generator. EL signal was analyzed by a single-grating monochromator and detected by an infrared-extended liquid nitrogen cooled tube. Time-resolved measurements were made by pumping the system at steady state, then switching off the applied voltage and detecting how the EL signal at a fixed wavelength decreased with time. Low temperature measurements were performed by using a close cycle liquid He cryostat with the samples kept in vacuum at a pressure of 10^{-3} Pa.

Fig.1 reports typical room temperature EL spectra of a device based on amorphous Si nc obtained by applying under forward bias conditions voltage values ranging from -13.5 to -19 V, corresponding to current density values ranging from 2.2 to 22 A/cm^2. On ore harel The EL spectra are characterized by a broad peak in the 700-1 200 nm range and their intensity increases by increasing the current density flowing through the device because the number of injected carriers in the matrix increases[18]. The peak shape is characterized by a main component at a wavelength of about 1000 nm and by a weaker one at about 740 nm. The presence of two well distinct maxima is not a peculiarity due to the electrical excitation. In fact, the PL spectrum of the active layer is characterized by a peak shape and is very similar to that one reported in the figure[18]. On the other hand the PL spectrum resembles a more typical shape, consisting of a single peak centered at about 800 nm, in absence of the poly-Si layer on top of the film. This demonstrates that the two-maxima shape reported in Fig.1 is not a peculiar characteristic of the active layer we have used but it depends on the optical properties of the poly-Si/SiO_x stack. Indeed, the high refractive index contrast at the poly-Si/SiO_x interface creates multiple interference phenomena in the poly-Si layer and, in turn, strong oscillations in the value of the transmission coefficient, therefore, acting as an anti-reflection coating favours the extraction of selected wavelengths.

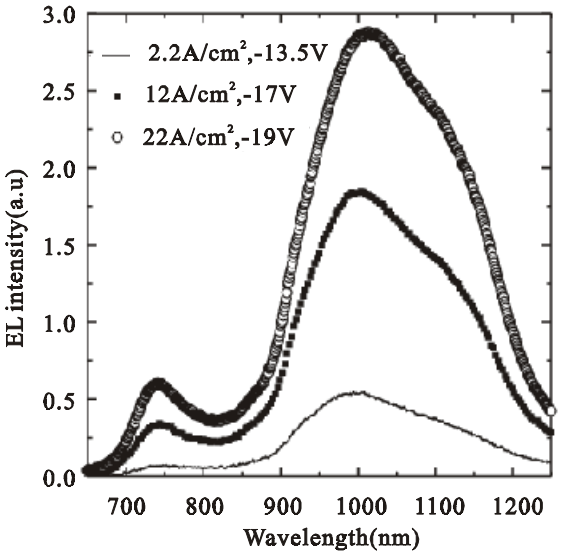

Fig.1 Room temperature EL spectra of a device based on amorphous Si nc obtained at different current densities and voltages.

Fig. 2 reports the EL intensity at 950 nm as a function of the temperature measured under forward bias with a current density passing through the device of 0.16 A/cm^2. The EL signal reproduces the typical behaviour already reported in literature[9] for devices based on Si nanocrystals, which has been attributed to the singlet-triplet energy splitting[19] of a self-trapped exciton in an interfacial level localized at the Si nanocrystal/SiO_2 interface[20,21]. The same dependence on temperature has been found also for the PL signal[18], demonstrating that photon emission from amorphous Si nc involves the same emitting centers under both the electrical and the optical excitation.

The de-excitation properties of the devices have been characterized by measuring the time-decay of the EL signal at 950 nm as a function of the current density passing through the device. The data have been collected by forward biasing the device with a square pulse at a frequency of 55 Hz; when the excitation is switched off we observe the decay of the EL signal as a function of time. The results are reported in Fig. 3.

The lifetime of the EL signals has been extracted by fitting the experimental curves with the equation:

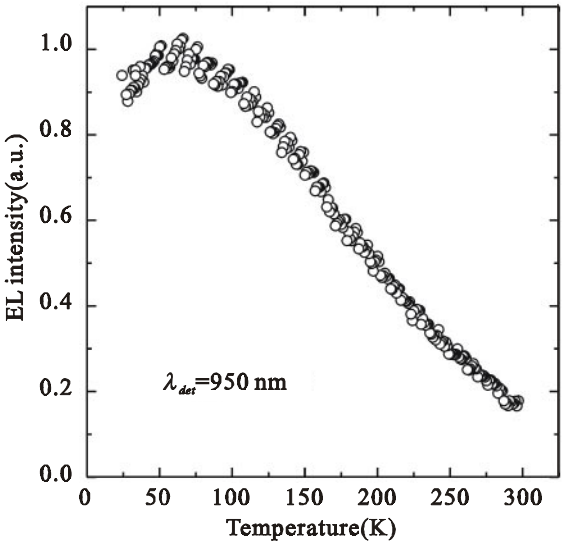

Fig.2 EL intensity measured at 950 nm as a function of temperature with a current density of 0.16 A/cm².

$$I(t)=I_0\exp[-(t/\tau)^{\beta}] \quad (1)$$

where $I(t)$ and I_0 are the EL intensity as a function of time and at $t = 0$, respectively, τ is the EL decay time, and β is a dispersion factor ≤ 1. The smaller β is, the more "stretched" the exponential is. The stretched behaviour characterizes the PL time decay of interacting Si nanocrystals[22,23]. For the curves reported in Fig.3, it is possible to observe that the EL lifetime becomes faster by increasing the current density. In particular, by increasing the current density from 0.4 to 22 A/cm², τ decreases from 1.5 μs to 300 ns, while β holds an almost constant value of about 0.90. It is important to note that the lifetime measurements are not limited by the excitation time resolution since the voltage fall-time is about 50 ns. The strong reduction of τ by increasing the current density suggests the occurrence of an Auger process between carriers and a nearby excited nc. Furthermore, we remark that the τ values reported in Fig.3 are extremely shorter than those measured for Si nanocrystals[9] that are typically of the order of tens of μs for current density values of a few A/cm², compared with the value of 600 ns we have found here for a current density value of 3 A/cm².

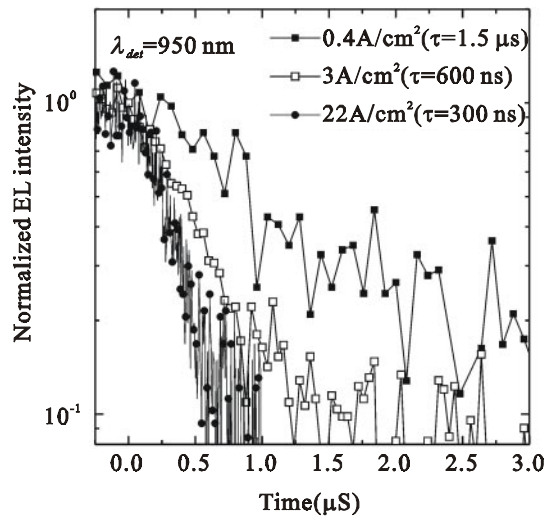

Fig.3 EL time decay curves measured at 950 nm for different current densities.

The above exposed data demonstrate that amorphous Si nc may constitute a promising active material for the fabrication of light emitting devices compatible with the Si technology, since their use should allow to remarkably decrease the thermal budget needed for the Si nanostructures formation step (900 °C) with respect to the case of Si nanocrystals, where the usual annealing temperature is 1100 °C or higher[17]. However, devices based on amorphous clusters, exhibit very different operating conditions from those based on Si nanocrystals[8,9]. In particular, for the same applied voltage, they exhibit current densities several orders of magnitude higher than those based on Si nanocrystals.

The main reason for the reduced quantum efficiency in amorphous clusters is represented by the fast lifetime values shown in Fig.3. However, this explanation is not sufficient. In fact, the lifetime of amorphous nc is about two orders of magnitude smaller than that of nanocrystals, while at a constant current density, the EL of nc is only one order of smaller magnitude . This suggests that indeed part of the decrease in lifetime for the amorphous clusters should be lifetime due to a much shorter radiative. Since amorphous nc are smaller than nanocrystals, this assumption is very likely. This observation suggests that, if non-radiative processes can be controlled, amorphous Si nc represents an extremely promising material for practical applications in optoelectronics. during high current density operation The high stability and reliability exhibited by the devices is also very encouraging. Indeed, the devices have been continuously tested for several weeks without any sign of deterioration.

In a typical silicon LED, as a result of the high silicon refractive index, most of the emitted light remains trapped inside the device by total internal reflection, thus severely limiting the overall performance. To increase the efficiency of the above described devices, we have integrated them with a properly designed PhC. Fig.4(a) reports the schematic of the device; with the aim of enhancing vertical light extraction, the polysilicon layer has been patterned by means of con-

ventional optical lithography to produce a triangular two-dimensional photonic crystal. The SEM image shown in Fig.4 (b) illustrates the top view of the structure and the good uniformity of the hole shape and spacing. The lattice constant of the photonic crystal has been tuned in order to introduce a fourth-order diffraction escape path at 860 nm (i.e., within the emission band of silicon nanostructures) for those light modes which are index guided on the top Si layer. In fact, we observed at around 860 nm, in resonance with the designed diffraction spot of the two dimensional photonic pattern, a sharp emission peak[24]. In Fig.5, we report the ratio between the EL intensities of the PhC device and of the device without PhC structure; it is possible to observe that the PhC increases the intensity of the emission by a factor of 4 at 860 nm. It is important to note that the EL signal coming from a device in which a random distribution of holes has been realized is very similar to the unpatterned one, indicating that a random hole distribution has almost no effect on the extraction of trapped light.

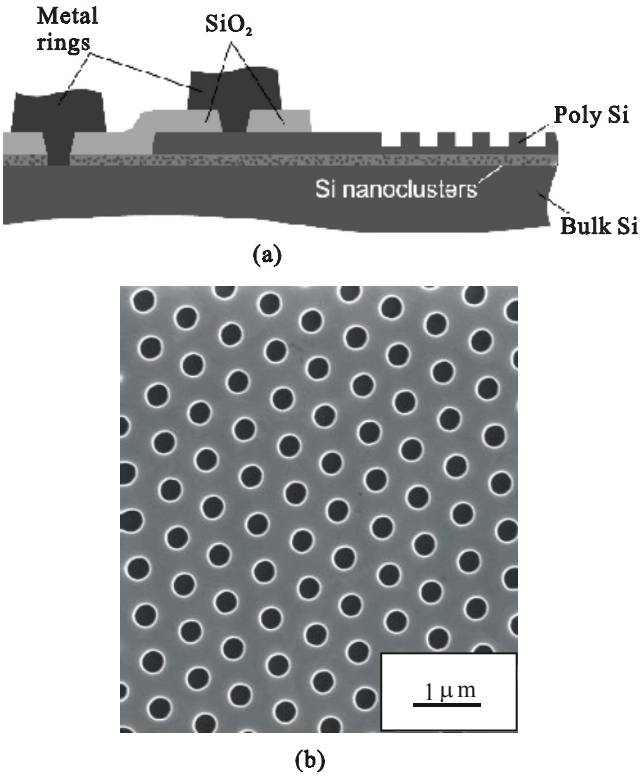

Fig.4 (a) Schematic of the device with PhC structure. The active region containing amorphous Si nc is sandwiched between two conducting layers. (b) SEM image of the PhC fabricated on top of the device.

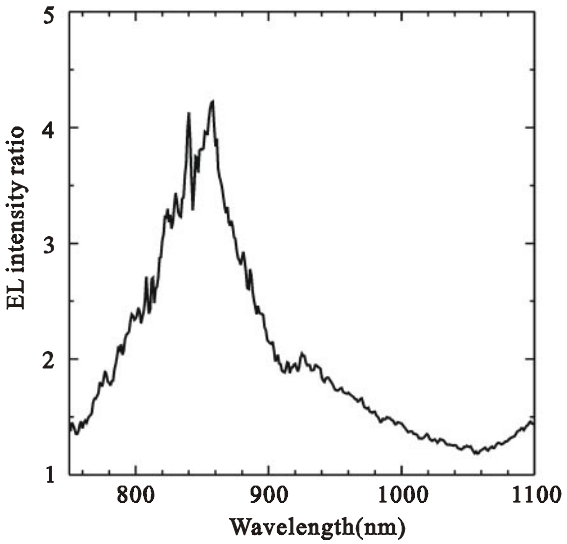

Fig.5 Ratio between the EL intensities of the device with PhC structure and of the device without PhC structure.

In this paper, we have reported the fabrication and the characterization of light emitting devices based on amorphous Si nc. These devices exhibit an intense and stable EL emission at room temperature. The results demonstrate that light emitting amorphous Si nc may constitute a very promising material for the fabrication of a Si-based light source, since they should allow us to couple interesting optical properties with the possibility to remarkably decrease the thermal budget of the preparation process with respect to the case of Si nanocrystals. The efficiency of these devices is similar to that found in devices based on Si nanocrystals; the high current density needed to observe intense EL signals is balanced by the low operating voltage and these unusual working conditions have no detrimental effect on the device reliability.

Moreover, we have obtained a four-fold enhancement of light extraction in amorphous Si nc devices by exploiting diffraction effects introduced by the coupling with a PhC structure. We have succeeded in conciliating VLSI compatibility, electrical pumping, and enhanced extraction efficiency in Si nanostructure LEDs. However, having demonstrated that even simple PhC structures are effective in improving the performance of Si nc light sources, we expect that these results can open the way to a whole new generation of silicon devices in which photonic and electronic functions are integrated together.

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