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Spectral- and time-resolved electroluminescence of silicon nanocrystals based light emitting devices

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Abstract
In this work we study the electroluminescence (EL) from a high efficiency multilayered silicon nanocrystals light emitting diode. The spectral analysis of EL under dc condition shows a spectrally modulated optical emission. Through reflectivity measurements we evaluated the effects of interference on the EL lineshape due to device structure and ascribed the emission to recombination in size dispersed silicon nanocrystals (Si-NCs). By studying the time resolved current–voltage $I$–$V$ and EL–$V$, we evidenced that injected carriers are both accumulated separately and concurrently in Si-NCs. At the bias transition the accumulated carriers either are extracted from the gate oxide giving rise to a short current pulse or they diffuse to large Si-NCs giving rise to an EL overshoot which decays with $\mu$s time constant.

Keywords: silicon, nanocrystals, electroluminescence, time-resolved, overshoot, spectra, reflectivity

(Some figures may appear in colour only in the online journal)

1. Introduction

Silicon nanocrystals (Si-NCs) light emitting diodes have been developed with the aim to give an integrated light source for silicon photonics. Unfortunately, their power efficiencies are still too low to be considered for application [1]. In many reported devices, the light emission under high voltage direct-current (dc) bias is due to the inefficient impact excitation process, which follows Fowler Nordheim (FN) tunneling injection. This injection mechanism causes damages to the dielectrics and, eventually, the failure of the device. By looking at the spectral shape of the emission, either excited by injection or by optical pumping, many authors claimed a multi-centre origin of the electroluminescence (EL) where unspecified interface states are in competition with exciton recombination in Si-NCs [2]. Moreover, sequential FN injection of holes and electrons into Si-NCs by using an alternating polarity bias voltage has been invoked to obtain high EL efficiency [3]. In previous works, we demonstrated that it is possible to achieve bipolar injection at low voltages under dc conditions in multilayered devices by alternating nm-thick SiO$_2$ layers to nm-thick Si-NCs layers [4]. In this letter, we address the time and spectrally resolved EL under dc and alternate-current (ac) driving in a multilayered device to get insight on the recombination mechanisms.

2. Experimental details

The studied devices are based on a metal oxide semiconductor (MOS) structure, where alternating stoichiometric SiO$_2$ and silicon rich silicon oxide (SRSO) layers are used as gate dielectric (figure 1). Si- NCs are grown by plasma enhanced chemical vapour deposition (PECVD) and annealed at 1100 °C. The multilayer stack is formed by nominally 2 nm SiO$_2$ + (3 nm SRSO/2 nm SiO$_2$) repeated five times to yield
a total gate thickness of 27 nm. As substrate, p-type doped silicon is used while the gate is a n-type polysilicon layer with a nominal thickness of 100 nm (figure 1). Details on the fabrication and the structural characterization of the active layer could be found in [5].

Current–voltage ($I$–$V$) characteristics are recorded with an Agilent B1500 A semiconductor device analyzer. The device is electrically driven by applying a bias to the n-type polysilicon top contact while grounding the p-type substrate (i.e. negative voltage values correspond to forward bias while positive to reverse bias). A function generator, Tektronix AFG 3252, is used to drive the device in ac. EL spectra are collected by a fiber bundle and analysed with a Spectra-Pro 2300i monochromator coupled with a nitrogen cooled charge-coupled device (CCD) camera. Time-resolved EL signal is collected with a single photon counting module, PerkinElmer SPCM-AQRH 16, and recorded via a multichannel scaler, Stanford Research System SR 430. Spectral and time resolved measurements are performed by placing near-infrared (NIR) interferential filters between the fiber bunch and the detector. The measurements are performed at room temperature in the dark.

The optical power is recorded with a large area p-i-n photodiode (UDT PIN-10DF), whose output current signal is measured by a Keithley 6485 picoamperometer. The photodiode is placed within a few millimeters above the devices. A simple correction for the collection solid angle is taken into account.

3. Results and discussion

Under reverse bias the LEDs do not show any measurable EL signal. In forward bias uniform and orange EL was observed from the LED surface (figure 1). EL spectra collected at different forward bias voltages are shown in figure 2. The spectra are peaked at 900 nm, which is almost independent from the voltage. Rising the voltage, a band emerges at 800 nm. In the literature this lineshape has been interpreted as due to recombination from two distinct recombination centres [6]. In addition a small undulation is superimposed to the spectra. To clarify the origin of this peculiar lineshape, we measured the reflectivity at normal incidence focusing the spot on the active area of the LED (see figure 2, left axis). Interference fringes observed are due to multiple reflections from the polysilicon electrode interfaces (perfect matching between experimental surface reflectivity and simulated surface reflectivity). We note that the emission from the active gate dielectric, where Si-NCs are formed, is modulated by this interference. However, the surface reflectivity alone cannot explain the EL lineshape. Indeed, the undulation on the EL lineshape has a higher frequency than the one measured by the surface reflectivity. The reason is due to the fact that the fibre bundle, which collects
the EL, collects the emission from the whole LED emitting area. Therefore, emission is also directed to the periphery of the active area where the thickness of the polysilicon gate is thicker (figure 1). Indeed, when we model\(^4\) also this effect (total reflectivity line in figure 2, left axis) we are able to reproduce the emission lineshape (diamonds in figure 2, right axis) by the product of a Gaussian lineshape centred at 900 nm and the modelled reflectivity spectrum. Thus, no different recombination centres are required to explain the broad and structured EL lineshape, which, in our case, is due to recombination in the size dispersed Si-NCs which are excited by the electrical injection.

Figure 3 shows the time resolved current signal when the bias is a square waveform between −4 and +4 V at a frequency of 50 kHz. A current pulse less than 1 \(\mu\)s long is observed at each bias transition (higher when passing from forward to reverse bias). While no current is observed under reverse bias, a constant current is measured in forward bias. The rectifying \(I-V\) characteristic of this LED is shown in the inset to figure 3. It is noted that dc or ac bias yields the same injected current in stationary condition. The current pulse, which is manifested at each bias transition is significantly larger than the current injected in stationary condition under forward bias. As we have already demonstrated [4], in forward bias we do have bipolar injection of electrons and holes into the Si-NCs. When both carriers are injected into the same Si-NCs, EL occurs (see the model in figure 1 and the EL spectra in figure 2). In forward bias, holes and electrons are injected into the gate dielectric. A few of them are trapped into Si-NCs near the gate dielectric interfaces; others move

\(^4\) The basics needed to perform numerical simulations on multi-layer films in optics has been taken from [7].
lineshape characterized by longer time constants for longer wavelength [10]. Figure 4 (right, bottom) shows the wavelength resolved decay lineshapes with, in the inset, the time constants obtained by a fit to the decays with stretched exponential line-shape [11]. The lengthening of the lifetime with wavelength is typical of quantum size effects in Si-NCs [10].

The experimental results can be explained by a combination of Si-NCs charging followed by carrier migration between Si-NCs (see sketch in figure 1).

In forward bias, there is a continuous charge injection. The injected carriers drift through the device as a function of applied field. The carriers will move through the device within a characteristic time (transient-time) or recombine radiatively or non-radiatively. Therefore, the EL depends on transient-time and lifetime (radiative and non). When the bias is inverted there is a kind of dead-time where the transient-time is zero, with plenty of charges in the active layer. In this moment much more EL is observed, since there is just diffusion or migration of carriers and no electric-field dependent drift. In fact, at the forward to reverse bias transition, electrons and holes trapped separately in Si-NCs near the gate dielectric interfaces can either tunnel in the electrodes (this originates the current pulse) or diffuse in the dielectric and reach Si-NCs where opposite-sign carriers are trapped (this originates the EL overshoot). With no current flow in the device—as with no or reverse bias, the electron-hole pairs in the Si-NCs recombine with the typical recombination dynamics observed in usual photoluminescence experiments [10]. This condition is enhanced by the absence of the externally applied electric field, which would force the charge carriers to tunnel across the dielectric layers. The EL overshoot is also enhanced by the presence of the current peak due to the release of carriers accumulated in the device under steady state conditions, and which are released when the external biasing voltage is switched off [8]. The EL overshoot is not present during reverse-to-forward bias transitions because of the lack of injection in reverse which prevents charge accumulation in the gate dielectric.

The recorded power efficiency of the analyzed device is reported in table 1.

<table>
<thead>
<tr>
<th>Optical power density [µW cm⁻²]</th>
<th>Current density [mA cm⁻²]</th>
<th>Gate voltage [V]</th>
<th>Power efficiency [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.51 ± 0.02</td>
<td>0.59 ± 0.01</td>
<td>2.00 ± 0.01</td>
<td>0.043 ± 0.002</td>
</tr>
</tbody>
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Figure 4. (Left) Time resolved electroluminescence (EL) for three bias conditions. Top-red lines, the voltage switches between [+3 V, −3 V]; central-blue lines [0 V, −3 V]; bottom-green lines [+3 V, 0 V]. The frequency of the square waveforms is 100 Hz. (Right, top) Contour plot of the EL under square waveform excitation [+3 V, −3 V] when resolved in time, x-axis, and in wavelength, y-axis, respectively. Colour codes are continuously tuned from low EL intensity-blue to high EL intensity-red. (right, bottom) The EL lineshape relative to the forward-to-reverse bias transition for some selected wavelengths from 950 to 700 nm. The decay times extracted from a stretched exponential decay lineshape fit are reported in the inset. Different frequencies of the square wave bias are used in this figure due to the different information that we aim to show.

4. Conclusions

In conclusion, the injection dynamics of electrons and holes in Si-NCs is strongly dependent on the bias waveforms. Under forward bias conditions electrons and holes tunnel in the gate dielectric and are either separately trapped in Si-NCs or reach
concurrently the same Si-NCs where they recombine. At the switch off of the injection, a rapid diffusion of the trapped charge occurs towards large Si-NCs, which causes the appearance of an EL overshoot peaked at longer wavelength that decays with the characteristics recombination dynamics of unbiased Si-NCs.

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References