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Field effect white and tunable electroluminescence from ion beam synthesized Si- and C-rich SiO₂ layers

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White and tunable electroluminescence has been obtained by field effect injection in 40 nm thick Si- and C-rich SiO₂ layers. The films, synthesized by ion implantation, contain Si and C-rich nanoparticles embedded in SiO₂ which were formed by annealing at 1100 °C. Shifting of the distribution of C-related centers toward the interface region with the substrate allows us to obtain a characteristic white electroluminescence emission under pulsed excitation conditions. Moreover, an evolution of the emission spectrum from white to red is observed by changing the frequency of the pulse. A power efficiency higher than 10⁻³% is estimated. This work opens interesting perspectives on color tunability of field effect electroluminescent devices. © 2007 American Institute of Physics. [DOI: 10.1063/1.2807281]

The development of efficiently emitting silicon-based materials would allow the integration of optoelectronic functionalities and optical interconnects with Si microelectronics. The merging of Si-based photonics and microelectronics is expected to be a revolution, and recent breakthroughs have shown that this field of research is very active and achieving maturity.¹ Regarding light emission, several reports have proposed Si-based materials for electroluminescence (EL) devices, either based on Si nanocrystals (Si-nc) or nanoclusters excitation by hot electrons^{2,3} and optically active defects^{4,5} or field effect EL using pulsed excitation.^{6,7} These devices usually emit in the red for Si-nc in SiO₂ and show a narrow tunable capability depending on Si-nc size. Defects introduced by dopants such as rare earth allow shifting emission until ultraviolet but devices are not reliable for a long time use, and present a narrow emission range.⁴ Additional alternatives to shift the emission to the blue have consisted in replacing the oxide matrix with nitride matrix⁸ or to enrich the silicon oxide matrix with carbon.^{9,10}

We have recently demonstrated that it is possible to obtain white EL from Si- and C-rich silica layers in direct current (dc) excitation.¹¹ For this purpose, 40 nm SiO₂ layers were fabricated and implanted with Si⁺ and C⁺ ions. These layers show an intense white photoluminescence (PL) to the eye, which is due to the convolution of a red PL due to the Si-nc, and a blue-green PL due to the C-rich nanoclusters. Studying the transport mechanisms, we were able to show that EL appears in these samples when the Fowler-Nordheim regime is dominant. This suggests that the excitation of the luminescent centers is produced by impact ionization. A power efficiency of about 10⁻⁴% was reported for a typical voltage of about 40 V. At these voltages, the electric field is very close to the typical breakdown electric field in SiO₂, and the devices show some instabilities leading to a lifetime of the devices that tends to be short.

This work introduces an original solution never explored before for C doped Si-nc, and consists of using the concept

proposed by Walters *et al.* (see Ref. 6) of sequential field effect injection of electrons and holes in pulsed excitation as a method to (i) reduce the working voltage and duty cycle of the device to prevent breakdown, (ii) increase efficiency due to the absence of leakage currents, and (iii) increase the frequency response and possibilities of modulation due to the fast response of the blue-green emission peak due to C doping.¹² This work also demonstrates the presence of a strong dependence of the spectral emission of the EL signal upon implant parameters and also upon excitation conditions. This dependence is related to the different spatial distribution and emission efficiencies of the ion beam synthesized Si and C-rich luminescent nanoparticles. This new device opens interesting perspectives on color tunability in the visible range of field effect EL devices.

The samples were prepared by sequential implantation of Si⁺ and C⁺ in a 40 nm thick SiO₂ layer grown by dry oxidation at 1000 °C on a *p*-type <100> Si wafer. The implantation parameters (energy and dose) were obtained by means of TRIM simulations. Two kinds of samples have been elaborated in this study. The first one (sample 1) has been designed to have a single peak of C in the center of the film and a uniform profile of Si obtained by means of a four step implantation to block the outdiffusion of the C during annealing.⁹ The second one (sample 2) has been designed to have a single peak of C and of Si closer to the substrate. Here, the outdiffusion of C has been prevented by introducing a Si excess double that of C. Details on implantation parameters are given in Table I. Then, the samples were annealed at 1100 °C in N₂ atmosphere for 4 h to form the Si-nc and the C-rich nanophases. Both samples exhibit an intense white PL due to the red emission of the Si-nc and the blue-green emission of the C-rich complexes, as we have already reported on this kind of material.⁹ After implantation, x-ray photoelectron spectroscopy (XPS) measurements show that in sample 1 the peak of C is located at 25 nm from the Si substrate (right in the middle). The C and Si excesses in atomic percentage are around 10%.¹¹ In sample 2, XPS shows that both C and Si peaks are located at less than

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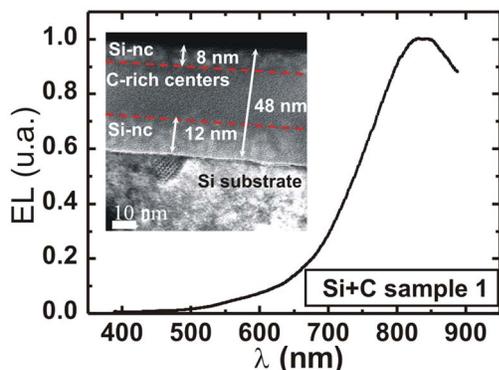
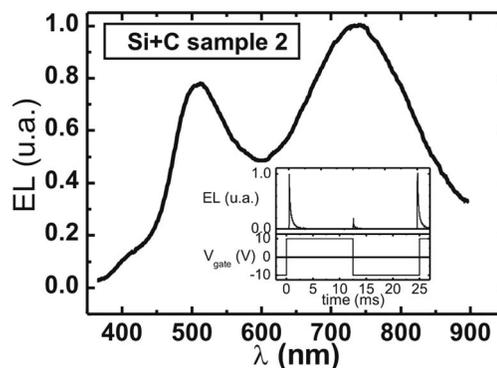
TABLE I. Energies and doses of implantation of Si⁺ and C⁺ in SiO₂ for samples 1 and 2.

	C implants		Si implants	
	Energy (keV)	Dose (cm ⁻²)	Energy (keV)	Dose (cm ⁻²)
Sample 1	6.5	2×10^{16}	1	1.4×10^{15}
			3	3.4×10^{15}
			10	7.5×10^{15}
Sample 2	11	2.7×10^{16}	25	2.7×10^{16}
			27	5.3×10^{16}

15 nm from the substrate, with an excess of 14% and 34% for the C and the Si peaks, respectively. Energy filtered transmission electron microscopy (EFTEM) was performed on a field emission TEM, FEI Tecnai™ F20 microscope operating at 200 kV, equipped with a corrector for spherical aberration and the last generation of the Gatan imaging filter series, the Trididem. The EFTEM images were formed with the electrons that were selected by a slit placed in the energy-dispersive plane of the spectrometer with a width of 4 eV centered at an energy position of 17 eV (Si plasmon energy). Metal-oxide semiconductor structures were fabricated by a standard photolithographic process, with an electrode active area of 9.6×10^{-3} cm². EL excitation was obtained using an Agilent 8114A pulse generator. Collected signal was analyzed with a monochromator and recorded with a charge coupled device camera cooled at 153 K. The spectra were corrected for the spectral response of the system.

Figure 1 shows EL from sample 1 obtained by applying a ± 20 V square signal at a frequency of 40 Hz. It shows clearly a red EL with a maximum of around 840 nm typical of radiative recombination in Si-nc due to quantum confinement effect.¹³ This contrasts with the EL signal measured in the same device under dc excitation conditions, where a broad white emission appears covering the whole visible range from 400 to 900 nm.¹¹ On the other side, white EL is observed on sample 2 under ac excitation conditions, as can be seen in Fig. 2; in this case, the EL is obtained by applying an ac square signal with the same frequency of ± 14 V.

To clarify what happens, an EFTEM picture from sample 1 has been inserted in Fig. 1. It shows that the active layer in this sample is separated in three regions. The two regions at the border are pure Si-nc embedded in SiO₂, and no C has been detected inside. In the region between the two Si-nc layers a C-rich region is detected, and no Si-nc are observed.

FIG. 1. (Color online) EL spectrum of sample 1 obtained by applying ± 20 V at 40 Hz. The inset shows an EFTEM cross section of the sample.FIG. 2. EL spectrum of sample 2 obtained by applying ± 14 V at 40 Hz. The inset shows the decay of the EL intensity with time and the gate voltage.

This result strongly suggests that for pulsed excitation the C-rich nanoparticles are too far from the substrate to be excited. Only the region rich in Si-nc close to the substrate is excited leading to a red EL. However, in sample 2, we have changed the implantation parameters on purpose to put the C-rich nanoparticles closer to the substrate. Consequently, the luminescent centers that are excited in both sample 1 and sample 2 are situated in a region close to the substrate. This strongly suggests that a mechanism of sequential injection of electrons and holes obtained by ac excitation similar to the one first proposed in Ref. 6 occurs in both samples. By applying a negative voltage to the gate, the *p*-type Si substrate enters into the accumulation regime. Due to the applied electric field, the holes are injected from the accumulation region to the Si or C-rich nanoparticles. Changing the polarization, an inversion region rich in electrons is formed in the substrate. These electrons are injected into the nanoparticles, with this mechanism being favored by the Coulomb attraction of the hole that was previously injected in the nanoparticle. The created electron-hole pair forms then an exciton that may recombine radiatively.

Consequently, these data demonstrate the possibility to obtain white field effect EL for films containing ion beam synthesized Si and C-rich nanoparticles. The inset of Fig. 2 shows the decay of EL with time for the whole spectrum, where it can be seen that the light intensity tends to zero after each pulse. This suggests that in the accumulation regime (negative gate voltage), a negligible leakage current flows from the gate because otherwise a continuous EL component would be observed. This supports the interpretation that at this voltage the EL is not due to impact ionization but to a sequential injection of electrons and holes from the substrate.

Let us now compare the efficiency of the white EL, i.e., in dc condition for sample 1 and in ac condition for sample 2. In sample 1, in dc conditions, at 40 V a current of about 10^{-5} A was measured, and a power efficiency of about $10^{-4}\%$ was estimated.¹¹ For sample 2, the estimation of the efficiency is not so evident. Indeed, the current due to the charging of the nanoparticles is hidden by the higher amount of current due to the displacement of charges at the interfaces of the capacitive layer. However, it can be qualitatively shown that a higher efficiency is obtained for sample 2. The power efficiency is defined as the ratio between the received optical power and the given electrical power. The optical power is proportional to the area of the EL spectra, and it can be found that both samples give a similar area, thus a similar

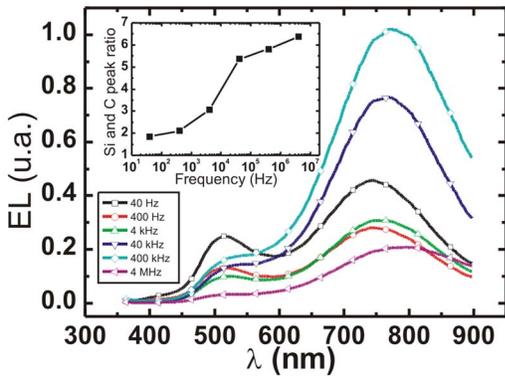


FIG. 3. (Color online) EL spectra of sample 2 at ± 20 V obtained by varying the frequency. The inset shows the evolution of the ratio between the Si and the C peaks.

optical power (the spectra have been taken with the same accumulation time). However, as commented above, it is found that the leakage current in sample 2 is very low, contrary to sample 1 where this leakage current is required to produce EL by impact ionization. Moreover, the applied voltage for sample 2 is four times less than for sample 1. It means that the electrical power consumed to give the same optical power is much less in sample 2 in ac conditions than in sample 1 in dc conditions. Taking into account the above-mentioned considerations, we have estimated a lower bound for the power efficiency of $10^{-3}\%$. Note that in these data, it is important to take into account that the EL devices have been fabricated using a semitransparent 100 nm thick polycrystalline Si layer as gate contact. The optical characterization of these layers shows a significant degradation of transmission in the blue spectral region, with values as low as about 6%. Then, optimization of the transparency of the gate electrode in these devices would allow a significant improvement in their power efficiency. This study thus shows the high interest in these layers to obtain white emitting Si-based LEDs with high efficiency.

Finally, a very interesting effect has been observed on sample 2. Indeed, we have observed that by changing the frequency of the pulse the contribution of both peaks can vary leading to a tunable EL from a white spectrum at high frequencies to a red-dominant one as frequency is increased. This effect is represented in Fig. 3. In this figure the spectra measured from 40 Hz to 4 MHz are shown for the same applied voltage of ± 20 V. It can be seen that a huge variation of the Si-nc peak is observed, with a maximum at 400 kHz. On the purpose to show the tunability from white to red luminescence, the ratio between the peak of the Si-nc and the C has been represented as a function of the frequency in the

inset of Fig. 3. A ratio close to one at low frequency corresponds to white luminescence, and the monotonic increase of the ratio with frequency shows a shift to the red. The increase of the red component with respect to the blue one could be due to the fact that the spatial distribution of the Si-nc is closer to the substrate than the C related one. An increase of the frequency could so decrease the probability of injection inside the C-rich centers. Material morphology investigations are ongoing to understand this point in more detail.

In conclusion, white EL has been obtained on Si- and C-rich 40 nm thick layers by field effect injection. A strong dependence of the EL shape on implant conditions has been shown. A power efficiency higher than $10^{-3}\%$ has been estimated. In addition, it is observed that the contribution of the Si or the C-rich luminescent nanoparticles exhibits a dependence on the frequency of the applied voltage. This opens interesting perspectives on the color tunability of field effect EL devices.

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