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Temporal dependence of gallium nitride quantum dot cathodoluminescence under weak electron beam excitation

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Abstract
In situ cathodoluminescence experiments on GaN quantum dots (QDs) show that the area concerned with the degradation is much larger than that where electron–hole pairs are created. It is also shown that an increase in the beam current density leads to a slower degradation of the QDs' optical properties.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Previous studies have shown that gallium nitride (GaN) epilayers are sensitive to electron beam (e-beam) injections [1–4]. The degradation of the ultra-violet (UV) luminescence is partly attributed to the spatial delocalization of excess carriers. Therefore, the radiation induced damage is thought to be reduced in GaN quantum dots (QDs) which exhibit three-dimensional carrier confinement, as shown in other semiconductor systems [5–9]. Of course, an increased resistance to beam damage is crucial for the development of UV devices such as laser diodes. To the best of our knowledge, no data on the reliability of GaN QDs to electron beam irradiations have been reported to date. Therefore, we have undertaken an *in situ* study of the temporal dependence of the UV luminescence GaN QDs under stationary e-beam irradiation at low excitation level.

2. Experimental details

The nitride based semiconductor structures studied in this work are separate or graded index separate confinement heterostructures (SCH).

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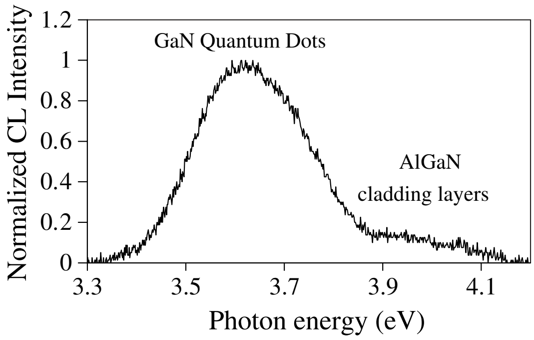

Figure 1. Room temperature CL spectrum recorded at 10 keV. The absorbed beam current is 0.18 nA. The spectrum has been recorded in the TV mode from a 10 μm × 10 μm area.

The SCH mainly studied in this work, and named SCH-1 in the following, has been grown at 730 °C by plasma assisted molecular beam epitaxy on a 6H (0001)-SiC substrate. A 1 μm AlN epilayer was first deposited on the substrate. The SCH region consisted of five layers of GaN QDs embedded in 15 nm $Al_{0.27}Ga_{0.73}N$ cladding layers deposited between two 45 nm $Al_{0.27}Ga_{0.73}N$ optical waveguide layers *coherent* on AlN. The GaN QDs, formed in the Stranski–Krastanov growth mode [10], were 2 nm high and 10 nm wide. A 120 nm AlN cladding layer was grown on the top of the structure.

A few experiments were performed on a second SCH (SCH-2) grown in similar conditions: the active region was made of three layers of GaN QDs embedded in 12 nm $Al_{0.5}Ga_{0.5}N$ cladding layers deposited between two 86 nm $Al_{0.5}Ga_{0.5}N$ optical waveguide layers. The AlN cladding layer grown on the top of the structure was 50 nm thick. The low value of the AlGaN barrier cathodoluminescence (CL) signal obtained from the SCHs at weak excitation level prevented us from studying its temporal evolution. Therefore, in order to compare the behaviour of GaN QDs with that of an epilayer, we performed the same experiments on a few microns thick epitaxial laterally GaN layer overgrown on (0001) sapphire (ELOG-GaN) [11] which exhibited a large enough polychromatic CL signal.

In situ plane-view CL experiments have been performed at room temperature in a Hitachi 4700 cold field emission scanning electron microscope (FESEM) operating at 10 keV and equipped with a Gatan CL collecting mirror. A Jobin-Yvon H20 UV monochromator and a Perkin-Elmer photomultiplier were used to record CL spectra. The beam current absorbed by the sample was varied in the range of 40 to about 300 pA. The beam current to the absorbed current ratio was constant; the beam current was measured with a Faraday cup. The evolution of the polychromatic intensity was recorded while the electron beam was kept stationary for 30 min (SCH-1, ELOG-GaN) and 50 min (SCH-2).

3. Results and discussion

Figure 1 shows a typical 300 K CL spectrum recorded at 10 keV and low beam current on SCH-1 before beam injection. The maximum of the depth dose function is located in the AlN buffer layer. The luminescence intensity of the AlGaN cladding layers, which occurs close to 3.94 eV, is about one-tenth of that of the GaN QDs whose peak is located at 3.62 eV. The full width at half maximum is as large as 260 meV, due to the distribution in the QDs' size [12]. Similar results hold for heterostructure SCH-2.

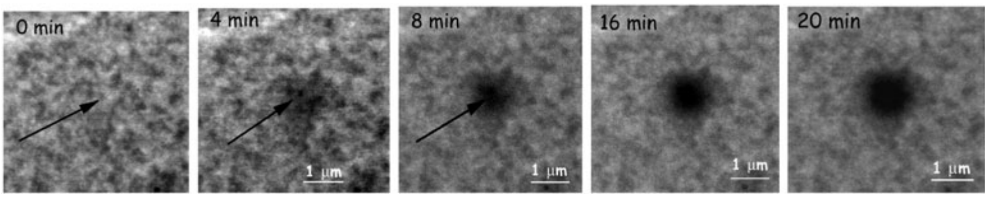

Figure 2. CL polychromatic images of SCH-2 under e-beam injection performed at 10 keV and recorded at different times between 0 and 20 min. The absorbed current is 0.2 nA.

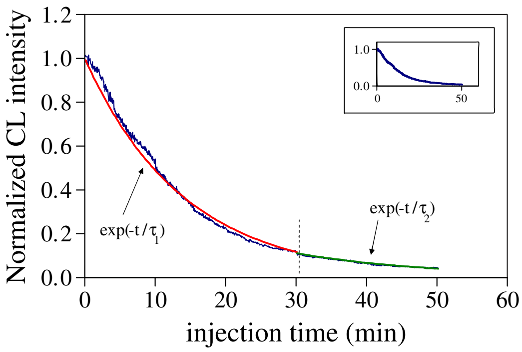

Figure 3. Time evolution of CL intensity SCH-2 structure under e-beam injection performed at 10 keV for 50 min. The absorbed beam current is 0.2 nA. The experimental curve, which corresponds to the CL images in figure 4, is fitted with two exponentials: $\tau_1 = 14$ min and $\tau_2 = 20$ min.

The spectrum in figure 1 shows that the CL polychromatic signal can be used to study the influence of e-beam injection on the optical properties of GaN QDs since it mainly originates from them. Polychromatic CL images recorded at different times of e-beam injection on SCH-2 at 0.2 nA evidence the appearance of a 'dark spot' after 4 min of injection (figure 2). The diameter of the dark spot, which does not increase between 4 and 20 min, is equal to 1 μm. This is much larger than the electron beam spot in a FESEM, than the lateral extension R of the 10 keV generation volume of electron–hole pairs (e–h pairs) and also than $R + L$ with L the minority carrier diffusion length: in the AlGaN layers of SCH-1, L was found equal to 7.5 nm [13]. Of course R can be calculated by Monte Carlo simulations [13]. But, in the case of our structures in which the QD density is large enough (5×10^{11} cm^{-2}) we can also experimentally estimate R from the spatial resolution of the CL images. Figure 2 shows bright small spots which correspond to the light emission of QD clusters. The smallest bright spot which can be detected is 200 nm in size. So we take this value as corresponding to the lateral extension R of the generation volume.

After 20 min of e-beam injection, the remaining CL intensity at the location of the e-beam, i.e. in the middle of the dark spot, is only 20% of the initial one, as shown in figures 3 and 4. It can be seen that during the first 30 min the CL intensity decreases by a factor of ten, whereas the dark spot diameter remains equal to 1 μm. The CL curve could be best fitted with two independent exponential functions. It first decays exponentially with a time constant of 14 min. After 30 min of e-beam injection, the dark spot diameter starts to increase from 1 μm at 30 min to 1.3 μm at 50 min. This corresponds to a slower decay of the CL curve: the CL intensity decreases only by a factor of 2.5 (from 10% to 4%) during these next 20 min (figure 3). The time constant of the second exponential function is equal to 20 min. These results evidence

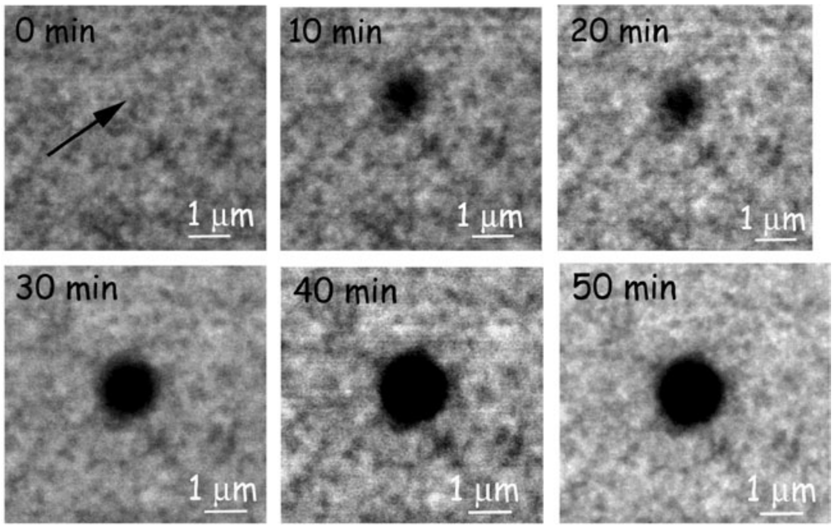

Figure 4. CL polychromatic images of SCH-2 under e-beam injection performed at 10 keV and recorded at different times between 0 and 50 min. The absorbed current is 0.2 nA.

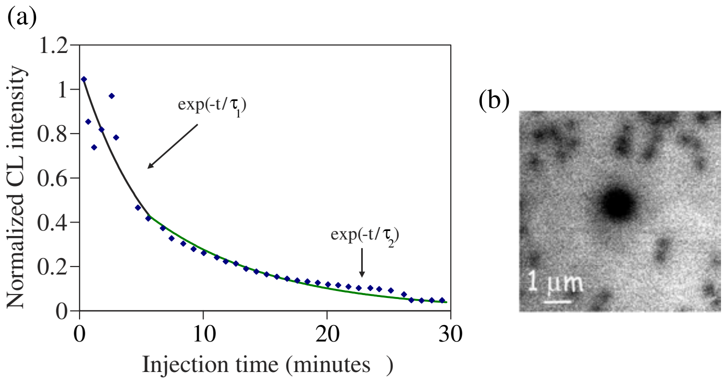

Figure 5. 10 keV stationary e-beam injection for 30 min on a ELOG-GaN epilayer performed at room temperature: (a) time evolution of the polychromatic CL intensity, $\tau_1 = 6$ min and $\tau_2 = 10$ min; (b) resulting 1.4 μm large dark spot. The smaller dark spots are dislocations. The absorbed current is 0.22 nA.

that, despite the three-dimensional confinement of carriers, the luminescence efficiency of QDs can be reduced by e-beam injection. But, preliminary results seem to indicate that ELOG-GaN epilayers degrade more rapidly than QDs and that a larger area is concerned: under similar e-beam injection conditions, the UV luminescence after 30 min is only 4% of the initial one in the GaN epilayer (figure 5(a)) and the resulting dark spot is 1.4 μm wide (figure 5(b)). The CL curve recorded on the ELOG-GaN epilayer can, as in the case of the QD case, be fitted by two independent exponentials but on a different timescale, both serial mechanisms leading to a faster degradation. The difference in the dark spot diameter in QDs and epilayers has also been observed on the SCH-1 heterostructure: it is 0.8 μm in QDs and 1 μm in AlGaN cladding layers (figure 6).

Together with the absence of recovery of the initial optical properties, these results suggest that a recombination enhanced diffusion of defects is responsible for the degradation, as has

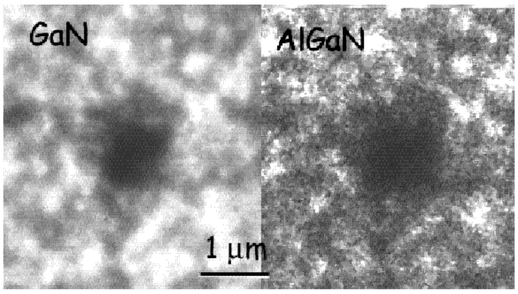

Figure 6. Dark spots resulting from a 30 min e-beam injection on SCH-1. The dark spot in the AlGaN cladding layer is larger (1 μm) than in the GaN QDs (0.8 μm). The absorbed current is 0.27 nA.

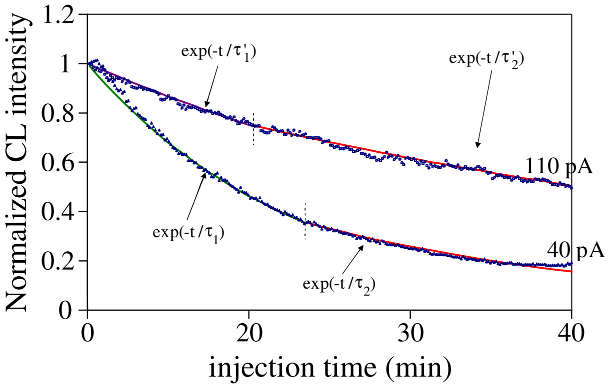

Figure 7. Time evolution of CL intensity for the SCH-1 structure for two values of the absorbed beam current. The experimental curves (rectangular and triangular points) are fitted by exponential decay curves with the following parameters: 40 pA curve, $\tau_1 = 13$ min, $\tau_2 = 20$ min; 110 pA curve, $\tau_1 = 35$ min, $\tau_2 = 50$ min.

been observed very frequently in semiconductors exposed to either electrons or ions [1–9]. In the case of QDs, the nonradiative recombinations which are needed to promote the diffusion of nonradiative defects could occur at dislocations, whose density has been measured to be as high as 3×10^{10} cm^{-2} in the AlN buffer of the SCH-1 heterostructure [13]. Point defects present at AlGaN/GaN interfaces could also be involved in nonradiative recombinations.

The last result of this paper concerns the effect of the electron beam current density. The experiments were carried out by keeping constant the values of the condenser lenses of the FESEM, and by changing only the current of emission, in order not to vary the FESEM beam spot. Thus, an increase of the absorbed beam current leads to an increase of the absorbed beam current density J (A cm^{-2}). We derive J from the value of R (lateral extension of the generation volume of e–h pairs, equal to 200 nm) and not from the FESEM beam spot diameter on the AlN cap layer; J is therefore more meaningful since it scales directly with the density of e–h pairs created within the QDs. In the case of figures 7 and 8, J is equal to 0.15 and to 0.35 A cm^{-2} for an absorbed beam current of 40 and 110 pA respectively.

Contrary to what could be expected, the CL intensity remaining after 30 min of e-beam injection increases with beam current density: 18.6% at 0.15 A cm^{-2} (40 pA) and 50% at 0.35 A cm^{-2} (110 pA) (figure 7). This could be due to a saturation of the nonradiative recombination centres with increasing beam current density. At the same time, the diameter of

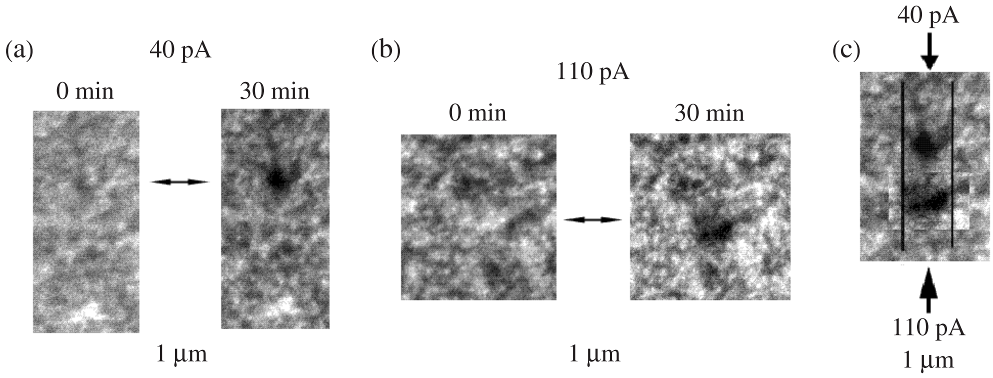

Figure 8. CL polychromatic images recorded on the SCH-1 heterostructure before and after the e-beam injections of figure 7. Influence of the absorbed current on the dark spot diameter: (a) 40 pA; (b) 110 pA; (c) the dark spot is larger after e-beam injection at 110 pA (1.2 μm) than at 40 pA (0.4 μm).

the e-beam induced dark spot also increases with beam current density: for instance, it is equal to 0.4 μm and 1.2 μm for J equal to 0.15 and 0.35 A cm^{-2} respectively (figure 8). By the same time, the time constant values derived from the fits of the CL curves by two independent exponential functions both increase with beam current density (figure 7).

Use of the beam current density instead of the beam current allows us to compare the results obtained on both heterostructures. Since in SCH-2 the remaining CL intensity as well as the dark spot diameter are smaller after injection at a larger absorbed beam current density of $J = 0.6$ A cm^{-2} (figures 2 and 3) it is highly probable that it contains a larger density of nonradiative recombination centres.

4. Conclusion

We have shown that the luminescence of GaN QDs is degraded by e-beam injection at low excitation level. The e-beam induced dark spot diameter is much larger than the lateral extension R of the generation volume of electron–hole pairs, as a result of a recombination enhanced diffusion of defects. The observed increase of the remaining CL intensity with increasing beam density could be due to a saturation of nonradiative recombination centres.

Acknowledgments

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