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Defect diffusion and strain relaxation in epitaxial GaN laterally overgrown on (0001) sapphire under low energy electron beam irradiation

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Abstract. *In situ* cathodoluminescence experiments have been performed to follow the time dependence of the UV luminescence in epitaxial lateral overgrowth GaN specimens. The decrease of the observed intensity and red-shift of the UV peak are interpreted in terms of non-radiative defect introduction and diffusion. This leads to strain relaxation of the GaN epilayer, which is initially under compressive strain. Monochromatic UV and yellow CL images show that dislocations act as efficient non-radiative recombination centres, that they are not at the origin of the yellow band and that they do not move under the electron beam.

1. Introduction

III–V semiconductors of the nitride family are attracting more and more interest because of their very many potential applications in the fields of optical and electronic devices. Their chemical stability allows also their use in a hostile environment. Blue light emitting diodes have been already commercialized, and blue laser diodes (LDs) with working life of about 10 000 hours under continuous wave operation were demonstrated, at room temperature, by Nakamura and coworkers in 1999 [1]. The LDs consisted of an InGaN/GaN/AlGaN structure elaborated on epitaxially lateral overgrown GaN (ELOG) layers, which, despite their high mismatch with the sapphire substrate, do not exhibit the mosaic structure of standard epilayers, and have a low dislocation density (10^7–10^8 cm^{-2}). Since the commercial development of nitride-based LDs requires the stability of their optical properties under photon or electron injection, we have found it of great importance to undertake an *in situ* study of the luminescence evolution, with time, of bulk GaN epilayers under the injection of a low energy (10 keV) electron beam. ELOG specimens have been preferred to standard ones since they allow us to study independently large areas free of dislocations as well as dislocation contrasts. In this paper, we present results which have been obtained in the cathodoluminescence mode (CL) of a scanning electron microscope.

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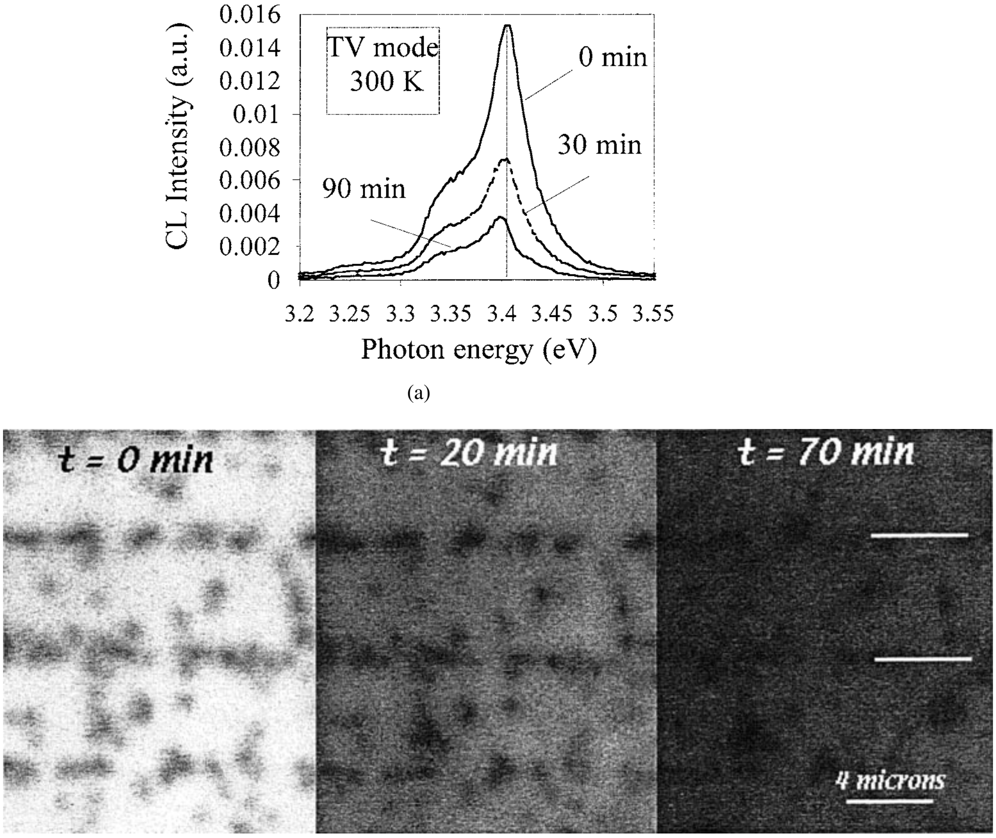


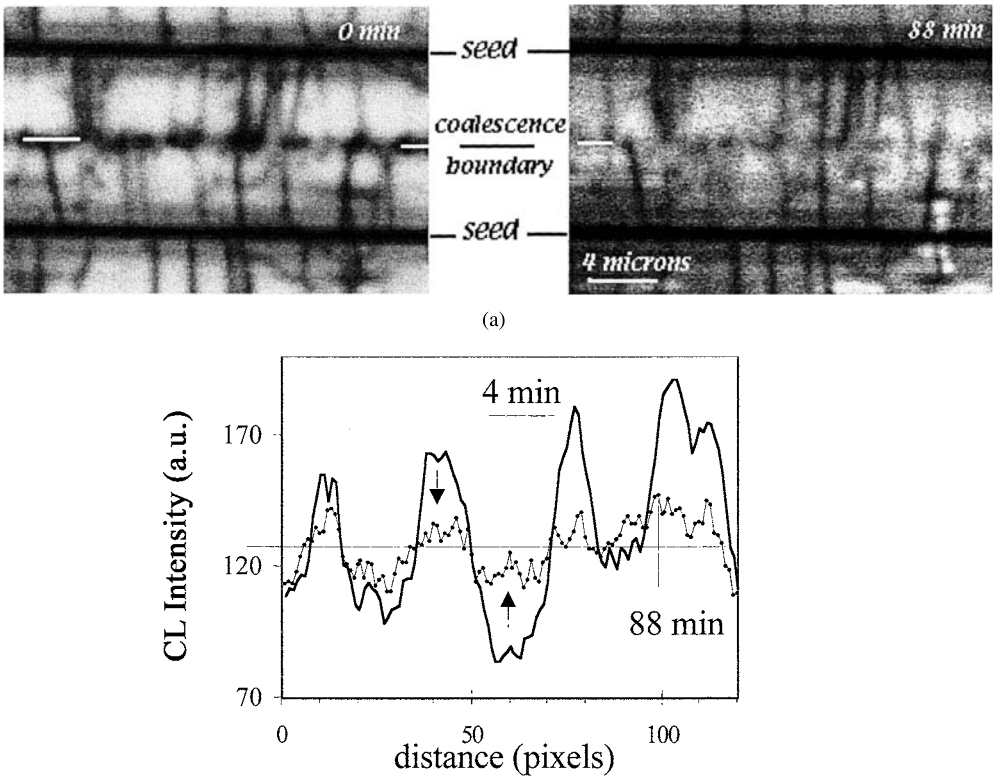
(a)

(b)

Figure 1. (a) CL spectrum recorded at 300 K in TV mode on the ELOG-2T specimen. The UV intensity decreases by about a factor of five within 90 min of beam injection. The low energy shoulder corresponds to the first phonon replica of the UV peak. (b) ELOG-2T specimen. Evolution of plan-view UV CL pictures with beam injection time. The numerical images are recorded at 90 K with the same brightness and contrast settings. The white lines show the coalescence boundaries.

2. Experimental details

The ELOG epilayer has been prepared by MOVPE on a (0001) sapphire substrate. The growth is achieved with a two step process in a home-made vertical reactor at atmospheric pressure using trimethyl species and NH_3 [2]. A GaN epilayer is first grown at 1080 °C on the sapphire substrate. Then, a silicon nitride mask is deposited and stripe patterns are opened by photolithography to form a grating with a 10 μm period and 5 μm wide stripes. The stripes are aligned along $[10\bar{1}0]_{GaN}$. In the first step, the temperature is fixed at 1040 °C, which favours the growth of GaN seeds in the form of pyramids. In the second step, the growth temperature is raised to 1120 °C to favour lateral growth from the seeds. When the structure is fully coalesced, a 2 μm thick GaN cap layer is finally deposited. This two step method reduces the dislocation density to a few 10^7 cm^{-2} even above the seeds [2]. This is due to the bending of dislocations of the seeds, by the image force, over the mask, in the (0001) plane. The specimen is nominally undoped, and the residual electron concentration in the cap layer is in the range 5×10^{16}–10^{17}cm^{-3} (specimen named ELOG-2T). The extrinsic yellow



(a)

(b)

Figure 2. (a) Plan view UV CL images of the etched ELOG-2T specimen. The images have been recorded at 90 K at the beginning (left) and after 88 min (right) of beam injection. The vertical dark lines located between the seed and the coalescence boundary correspond to dislocations which have been bent in the (0001) plane during the lateral overgrowth. (b) Evolution of the UV profile along the coalescence boundary shown in figure 2(a). The intensity of the brightest areas decreases whereas that of the darkest areas increases.

band (YB) being absent in the cap layer, we have studied a piece of the specimen which was thinned from 9 μm to about 7 μm by ionic etching, in order to record the evolution of both the UV and yellow bands (specimen named etched ELOG-2T). The cathodoluminescence (CL) experiments have been performed at 90 K and 300 K in a Cambridge Stereoscan 250 scanning electron microscope (SEM) equipped with an Oxford CL collecting mirror and home-made liquid nitrogen specimen holder. The luminescence has been detected with a R 636 Hamamatsu photomultiplier. A monochromator with 1200 grooves per mm of grating was used to record CL spectra and monochromatic plan-view CL numerical images. The spectral resolution is equal to 2 nm. The specimen has been also studied at 6 K in a Cambridge Stereoscan 360 SEM equipped with an Oxford CL mirror and an Oxford liquid helium specimen holder. The accelerating voltage and beam current were equal to 10 kV and 10 nA respectively. The ELOG specimens have been irradiated in TV and spot modes. In the TV mode, an area of 50 μm $\times$ 40 μm was scanned at the TV rate for 90 min. A CL spectrum and a monochromatic image were recorded about every 10 min. In the spot mode, the position of the electron beam remained unchanged for about 2 min.

3. Decrease of CL intensities

The first important result of our study concerns the decrease of the near band edge ultra-violet (UV) intensity with electron beam injection, whatever the temperature in the range 6 K–300 K. The UV luminescence corresponds, at 90 K and 300 K, to the recombination of the A free exciton. The decrease of its intensity is clearly seen in both CL spectra (figure 1(a)) and monochromatic CL images (figure 1(b)). It is faster when the temperature increases. As proposed by Toth *et al* [3], it could result from carbon contamination of the free surface. But, in that case, all contrasts in monochromatic pictures should decrease identically. Figure 2 shows that this assumption can be ruled out in our experiments: the UV intensity recorded along coalescence boundaries of the etched ELOG-2T specimen increases in dark areas, and decreases in bright areas with beam injection. The second important result is that there is no correlation between the intensity variations of both UV and yellow bands recorded on the etched ELOG-2T specimen (figure 3). Furthermore, no extra luminescence band or peak could be detected after beam injection. Since the electron beam energy used in our experiments (10 keV) is too low to create Frenkel pairs [4, 5], we can conclude that electron beam injection leads to the introduction of non-radiative defects, and then to the observed decrease of the UV and yellow luminescence intensities.

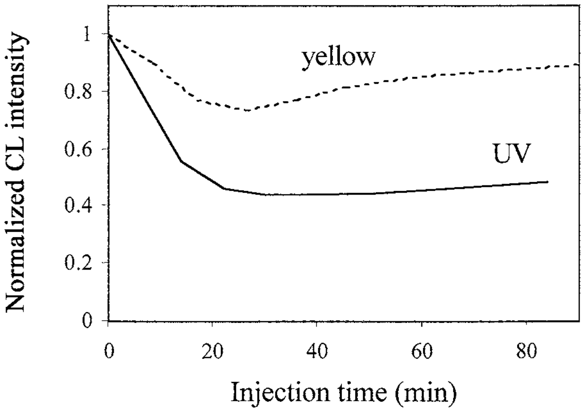


Figure 3. Etched ELOG-2T specimen. Evolution of the normalized maximal UV and yellow CL intensities.

Table 1. Evolutions of the UV band observed on the ELOG-2T specimen after a 90 min beam injection.

	TV mode—90 K	Spot mode—6 K	TV mode—300 K	Spot mode—90 K
Broadening	Yes	Yes	No	No
Red-shift	8 meV	No	10 meV	3–4 meV

4. Spectral evolution of the free A exciton related band

Three types of CL spectral evolution have been observed in the ELOG-2T specimen (table 1). When the beam injection is made at 90 K in the TV mode, the UV band broadens towards low energies, and its maximum red-shifts by 8 meV (figure 4). The full-width at half maximum

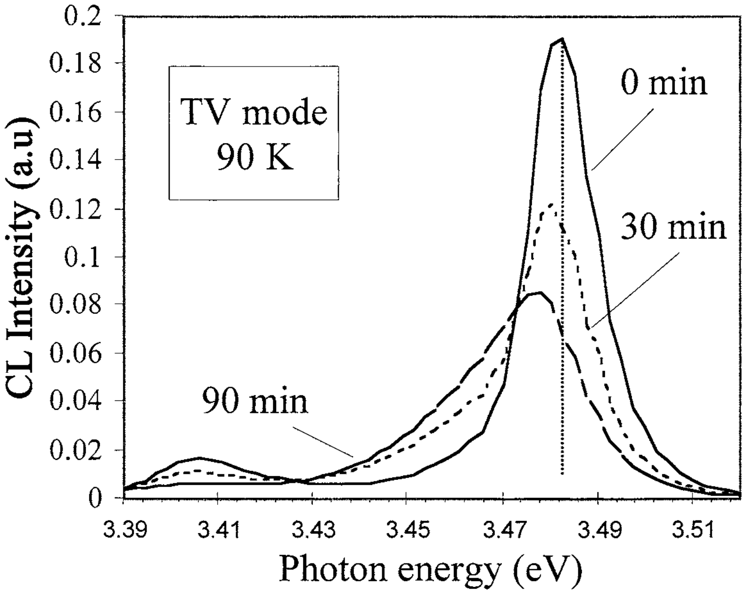


Figure 4. UV band evolution of the ELOG-2T specimen when the beam injection is made at 90 K in the TV mode. The FWHM increases from 17 meV to 30 meV, and the red-shift is equal to 8 meV.

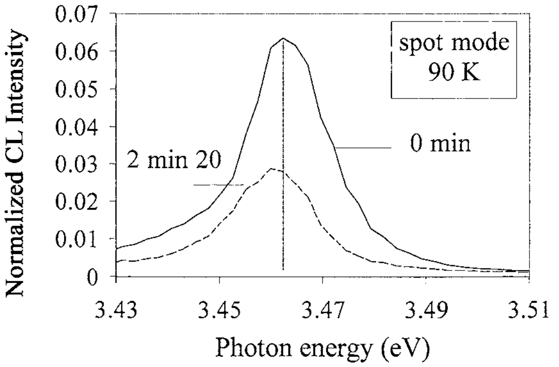


Figure 5. UV band evolution of the ELOG-2T specimen when the beam injection is made at 90 K in the spot mode. The red-shift is equal to 3.5 meV and no broadening is observed.

(FWHM) increases from an initial value of 17 meV to 30 meV after 90 min of beam injection. The same kind of broadening is also observed in the spot mode at 6 K, but without any red-shift. When the beam injection is made at 300 K in the TV mode, or at 90 K in the spot mode, no broadening occurs but the maximum is red-shifted by 10 meV (Figure 1(a)) and 3.5 meV (figure 5) respectively.

The red-shift of the UV peak may indicate strain relaxation of the GaN epilayer, which is under basal compressive strain when it is grown on (0001) sapphire [6]. From the initial position of the A exciton peak in the ELOG-2T specimen, the strain ε_{xx} is found equal to -0.1921% [7, 8]. The percentage of strain relaxation, estimated from the red-shift of the UV peak (table 1) varies from a minimum value of 18% (spot mode, 90 K), to a maximum value of 53% (TV mode, 300 K). The broadening of the CL spectra observed in the TV mode at

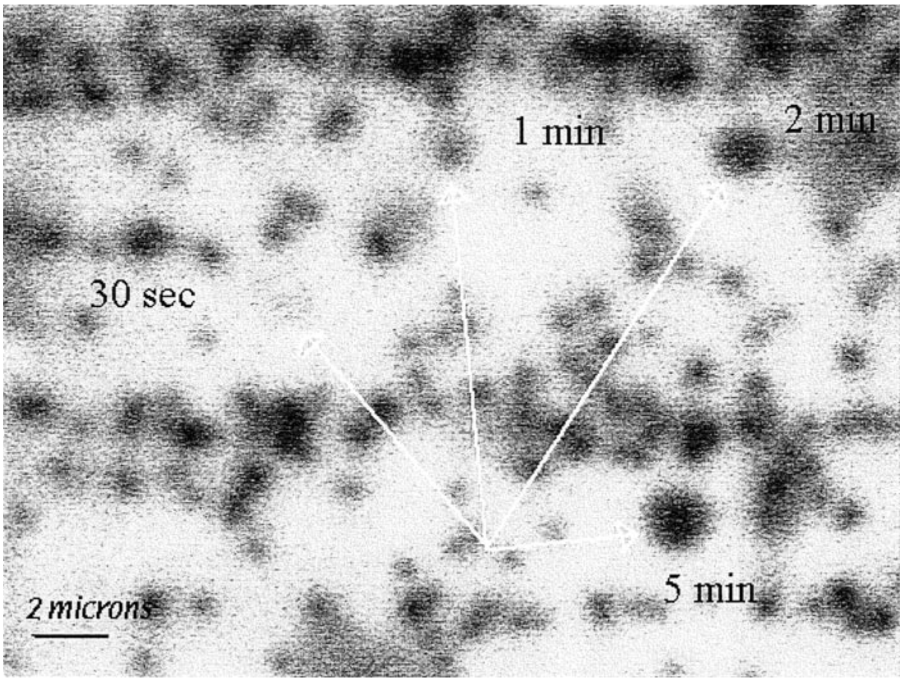


Figure 6. UV images of the ELOG-2T specimen showing the dark spots induced by the electron beam injection in spot mode. The diameter of the dark spots increases with injection time.

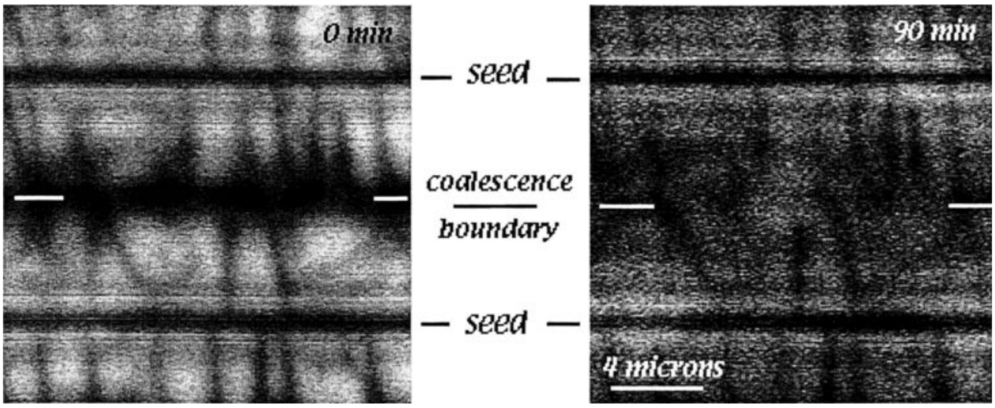


Figure 7. Plan view yellow (2.25 eV) CL images of the etched ELOG-2T specimen. The images have been recorded at 90 K at the beginning (left) and after 90 min (right) of beam injection.

90 K and in the spot mode at 6 K, shows that the electron beam injection leads to fluctuations of the band gap induced by *strain relaxation disorder* (i.e. spatially inhomogeneous strain). Since it is not observed in all experiments, it could be due to a slow diffusion of non-radiative defects which induces some defect disorder in the volume scanned by the electron beam. The diffusion of non-radiative defects is validated by the appearance of dark spots, in spot mode at 90 K, whose diameter increases with time: it doubles between 1 and 5 minutes (figure 6). The absence of any broadening in the CL spectra recorded at 300 K in the TV mode, and

at 90 K in the spot mode, shows that the diffusion of non-radiative defects is, under these conditions, rapid enough to lead to a spatially homogeneous distribution of defects, and then to strain relaxation. In contrast, when the temperature is very low (6 K), the diffusion is too slow to lead to strain relaxation. This shows that the diffusion process of non-radiative defects, which can be enhanced by non-radiative recombinations initially present in the material, is also thermally activated. The resulting strain relaxation increases with temperature. The defects involved in the relaxation process should obviously only be of the vacancy type. The vacancies can originate from the destruction of complexes, and may coalesce to form more elaborate defects such as dislocation nano-loops, as already observed by Bonard *et al* [8]. They can be issued from the free surface. In the case of the etched ELOG-2T specimen, they could also originate from the coalescence boundaries, which are probably made up of void regions, threading dislocations and impurities. Indeed, their reconstruction under electron beam injection is shown in UV and yellow CL images (figures 2(a) and 7). We suggest that pipe diffusion along bent dislocations accelerates the diffusion of vacancies from the coalescence boundaries to the bulk, whose luminescence decreases with beam injection [9].

5. CL imaging of dislocations

No extra dark spot or dark line has been detected during electron beam irradiation. Furthermore, within the CL spatial resolution, no dislocation motion was observed. These experimental facts are opposite to Maeda's observations by transmission electron microscopy [10]. The CL spatial resolution of dislocation images decreases under electron beam injection, indicating an evolution of their Cottrell atmosphere.

In all UV images recorded on the ELOG-2T specimen, emerging dislocations appear as dark spots surrounded by impurities (figures 1(b) and 6). From quantitative CL line scans profiles, their contrast was evaluated in the range 20 to 50%. This shows that dislocations in GaN act as efficient recombination centres. These large contrast values are of the order of those usually displayed by growth induced dislocations in III–V compounds such as GaAs. In all UV and yellow images recorded on the etched ELOG-2T specimens, bent dislocations appear as dark lines (figure 2(a) and 7). We have observed a one to one correspondence between the UV and yellow images of dislocations. It can therefore be concluded that dislocations are not, as often proposed in the literature [11–15], at the origin of the yellow band.

6. Conclusion

In situ CL experiments performed on ELOG specimens have shown that low energy beam injection leads to the decrease of both UV and yellow luminescences, as well as to the relaxation of the compressive strain. CL imaging of dislocations has shown that dislocations act as non-radiative recombination centres, and that they are not at the origin of the yellow band.

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