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Properties of GaN epilayers grown on misoriented sapphire substrates

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Three silicon-doped 3 µm thick GaN epilayers were grown simultaneously by metalorganic chemical vapour deposition on (0001) sapphire substrates misorientated by 0°, 4° and 10° toward the m-plane ($10\bar{1}0$). A comparative study of these epilayers was undertaken using photoluminescence (PL) spectroscopy, atomic force microscopy (AFM), scanning electron microscopy (SEM), cathodoluminescence (CL) imaging, CL spectroscopy and Hall effect measurements. Low temperature PL of the 0° and 4° epilayers shows donor bound exciton (BE) emission between 3.47 and 3.48 eV and a low level of yellow band emission. The peak intensities of both emission bands are a factor of 2 higher for the 4° layer. In the 10° epilayer, the BE band is 3x stronger than in the 0° epilayer but there is no discernible yellow band. However, a number of additional bands appear at 3.459, 3.417, 3.362, 3.345, 3.309, and 3.285 eV. These bands, some of which are acceptor related, may be attributed to the presence of structural defects in this epilayer, pointing to an abrupt degradation of its structural quality compared to the others. This degradation is confirmed by AFM studies. On a 20 µm x 20 µm image the 0° and 4° epilayers exhibit smooth surface morphologies, while the 10° epilayer shows a high density of hexagonal pits. Finally, SEM images reveal the surface of the 10° epilayer to be "streaked" and pitted. Low temperature CL images at 3.48 eV (bound exciton region) show random spotty emission, while those at 3.28 eV and 3.41 eV exhibit a streaky appearance similar to the SEM image. This suggests that these luminescence bands are indeed associated with structural defects.

1 Introduction

Since the first GaN epilayers were grown on sapphire substrates in 1971 [1], various techniques have been applied to optimise the quality of the resulting films, such as nitridation [2] of the substrate, or the growth of a low temperature AlN [3] or GaN buffer layer [4]. In this paper we will describe comparative morphological and low temperature luminescence studies of silicon doped GaN epilayers grown on misoriented sapphire substrates. In a number of material systems, growth on misoriented substrates has been shown to improve epitaxial growth. The crystallinity of epilayers may be improved by relieving strain where the epilayer and substrate are mismatched [5], the growth rate may be increased [6] as can the dopant concentration [7]. Previous studies of GaN epilayers grown on misoriented substrates include those reported by Hiramatsu et al [8], Grudowski et al [9] and Ishibashi et al [10]. However, to our knowledge, this is the first report of low temperature luminescence studies of silicon doped GaN epilayers grown on misoriented sapphire substrates.

2 Experimental details

Three silicon-doped 3 µm thick GaN epilayers were grown simultaneously by metalorganic chemical vapour deposition on (0001) sapphire substrates misoriented by 0°, 4° and 10° toward the m-plane ($10\bar{1}0$), see Figure 1. The misoriented substrates were prepared by an *ex-situ* solvent cleaning then an *in-situ* clean at 1200 °C for 10 minutes under hydrogen. The GaN epilayers were nucleated at 480 °C, using a 300Å GaN buffer layer, then the main epilayers were grown at 1050 °C using trimeythylgallium and ammonia as described previously [11]. The samples were characterised by photoluminescence (PL) spectroscopy [12], atomic force microscopy (AFM), scanning electron microscopy, cathodolumines-

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cence (CL) imaging, CL spectroscopy [13] and Hall effect measurements.

3 Results and discussion

Figure 2 shows low temperature (15 K) PL spectra from the three epilayers. They were acquired under identical excitation conditions. The PL is dominated by donor bound exciton (D^0X) emission [14] peaking at between 3.47 and 3.48 eV. We attribute this band to the silicon dopant. The peak intensity of the D^0X is a factor of 2 higher for the 4° epilayer and a factor of 3 higher for the 10° epilayer compared to the 0° epilayer. This increase in PL intensity we ascribe to an improvement in the silicon incorporation with increased misorientation of the substrate. The inset to Figure 2 shows higher spectral resolution temperature dependent PL spectra from the 10° epilayer in the vicinity of the D^0X peak (energy range 3.43 to 3.51 eV), revealing a higher energy peak ≈ 6 meV from the D^0X band. From this energy splitting and from the temperature dependence of the PL shown in the inset, we tentatively attribute this peak to free exciton (FX) luminescence [15].

A low level of yellow band emission is observed from the 0° and 4° epilayers. The peak height of the yellow band increases by a factor of 2 for the 4° epilayer, but there is no discernible yellow band emission from the 10° epilayer. However, as can be seen in Figure 3, which compares PL and CL spectra from the 10° epilayer in the energy range 3.1 to 3.5 eV, a number of additional bands at 3.459, 3.417, 3.362, 3.345, 3.309 and 3.285 eV are observed. As all three epilayers were grown simultaneously, these additional bands are unlikely to be the result of impurities. The origin of these additional bands must be structural defects in the 10° epilayer. This conclusion is supported by previous studies. The 3.459 eV peak we attribute to an acceptor bound exciton (A^0X) transition [15]. The 3.41 eV peak has been assigned to a donor to valence band (D^0h) transition associated with structural defects [16], or to excitons bound to either stacking faults [17] or screw dislocations [18] in GaN films grown on off-axis SiC substrates. The 3.362 eV peak has been attributed to an excitonic transition bound to dislocations, or has been associated with cubic inclusions [19]. A line at 3.345 eV has been identified as a defect-related donor-acceptor pair (DAP) transition [20]. The 3.309 and 3.285 eV bands we attribute to electron-acceptor (eA) and DAP recombination respectively [21] [15]. The bands peaking at 3.214 and 3.285 eV correspond to eA-LO and DAP(at 3.285 eV)-LO respectively. The presence of these phonon assisted bands is consistent with the designation of the 3.01 and 3.285 eV bands to eA and DAP transitions, and the temperature dependence measurements shown in Figure 4 provide further support. The 24 meV separation of the peaks of the eA and DAP bands is within the range of the currently proposed values of the silicon donor binding energy of between 17 and 30 meV [15].

As noted earlier in this discussion, yellow band emission is not observed from the 10° epilayer. We tentatively put forward two possible mechanisms for this quenching of the yellow band. 1) It has been reported previously that the yellow band is suppressed by doping [22] [23]. We intimated earlier in this discussion that silicon doping is greatest for the 10° epilayer and we have also presented evidence that the 10° epilayer is also p-type doped in that a number of the observed defect bands involve shallow acceptor-related transitions. 2) Reduced yellow band emission was also observed by Sasaki and Zembutsu [24] for MOVPE GaN epilayers grown on the $(01\bar{1}2)$ plane of sapphire. They suggested that this may be due to a suppression of the formation of nitrogen vacancies for this substrate orientation, it is conceivable that we are observing a similar effect with the 10° misoriented substrate.

Hall measurements indicate that all three epilayers are n-type, in agreement with the luminescence data. The carrier concentrations are deduced to be -2.4 x $10^{17}cm^{-3}$, -2.7 x $10^{17}cm^{-3}$ and -2.1 x $10^{17}cm^{-3}$ for the 0°, 4° and 10° epilayers respectively. The carrier concentration increases for the 4° epilayer with respect to the 0° epilayer, consistent with the observed increase in the donor bound exciton emission. The Hall measurements show that the 10° epilayer has a lower carrier concentration than either the 0° or 4° epilayers however, the PL implies that the donor concentration is highest for the 10° epilayer. This apparent contradiction can perhaps be explained if we take into account the presence of acceptors in the 10° epilayer. As previously discussed, a number of the additional luminescence bands observed for the 10° epilayer are acceptor related. Further electrical measurements are planned to verify if there is indeed carrier compensation for the 10° epilayer.

The degradation of the structural quality of the 10° epilayer is confirmed by AFM studies. The 0° and 4° epilayers exhibit smooth surface morphologies, while the AFM image (Figure 5), shows that the 10° epilayer exhibits a rough surface with a high density of hexagonal pits. A similar surface structure is observed when SEM images are acquired from the 10° epilayer. In Figure 6 we compare an SEM image with low temperature (≈50 K) CL images acquired from the D^0X luminescence (3.48 eV), from the 3.41 eV defect related exciton luminescence and from the DAP luminescence (3.288 eV). The contrast and brightness of these images has been slightly enhanced to bring out the brighter emitting

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features to allow gross features to be compared. All CL images show patchy emission, however, they are clearly very different. The D^0X CL image exhibits random spotty emission, while the 3.41 eV defect emission and DAP emission appears orientated with some correlation discernible between these CL images and the SEM image. The alignment of the CL emission lies at ≈ 50° to the scale bar, the same angle as the lines of pits observed in the SEM image. The streaky appearance of the DAP emission we attribute to the lifetime of this emission being longer than the dwell time of the beam at a given point as it scans across the sample. The random nature of the D^0X emission is consistent with it originating from the silicon dopant. The correlation between the SEM image, 3.41 eV defect emission and DAP emission, points to the structural origin of these emissions, i.e., they are related to structural defects. Overlaying the images reveals that at some points there is anticorrelation between the D^0X emission, and the 3.41 eV defect emission and DAP emission. This is particularly marked at the lower half of the left hand edge of the CL images.

4 Conclusions

In summary, we have presented results which indicate that silicon incorporation is improved when silicon doped GaN epilayers are grown on sapphire substrates misoriented towards the m-plane $(10\bar{1}0)$. For a 4° misorientation no discernible degradation of the epilayer is observed. However, at a substrate misorientation of 10°, the epilayer appears rough and pitted and additional defect luminescence, some of which is acceptor related, is emitted from the epilayer. Finally, yellow band luminescence is quenched for a substrate misorientation of 10°.

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FIGURES

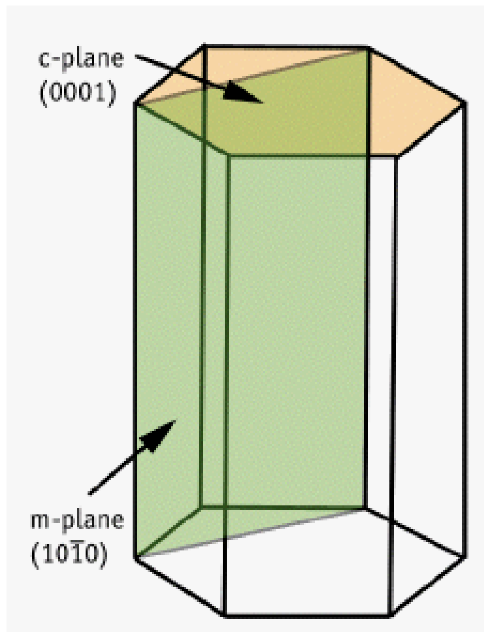

Figure 1. The (0001) and (10$\bar{1}$0) planes of sapphire.

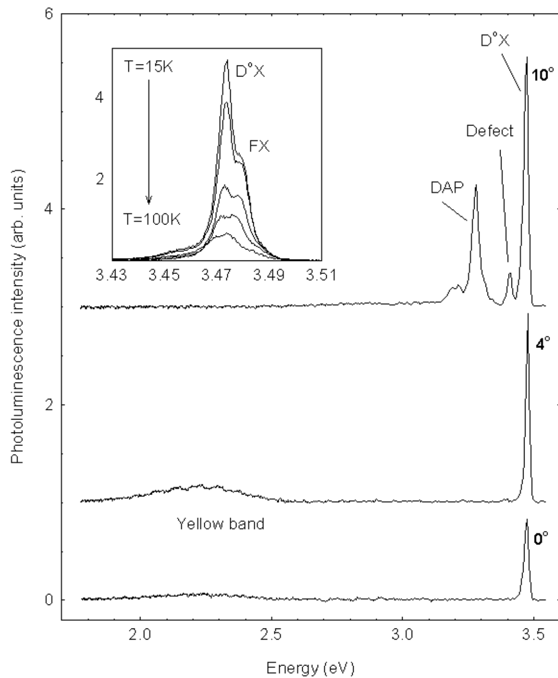

Figure 2. PL spectra from the three epilayers. The spectra are displaced for clarity. The inset shows high spectral resolution temperature dependent spectra in the excitonic region from the 10° epilayer.

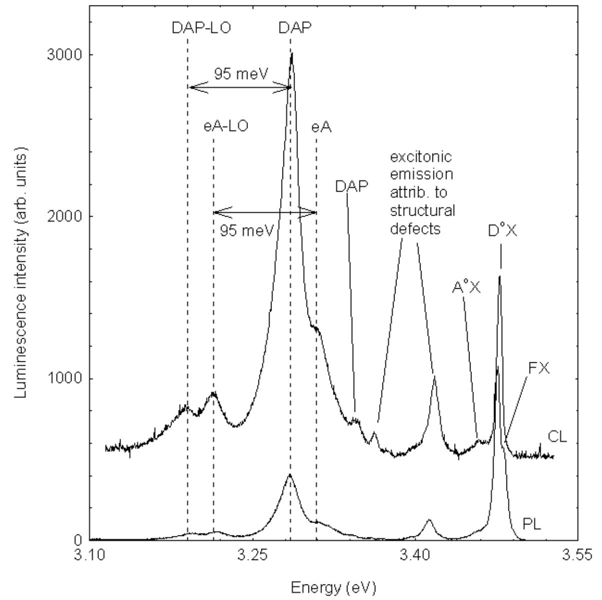

Figure 3. Low temperature PL and CL (15 keV) spectra from 10° epilayer.

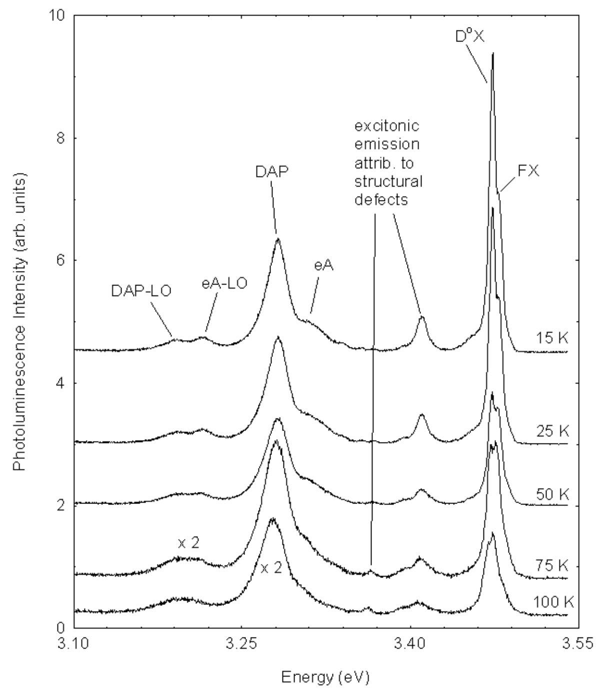

Figure 4. Temperature dependent PL spectra from 10° epilayer.

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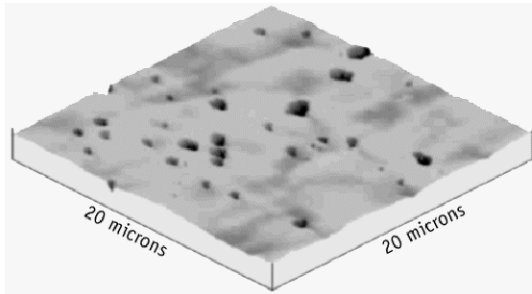

Figure 5. 20 µm x 20 µm AFM image of the surface of the 10° epilayer.

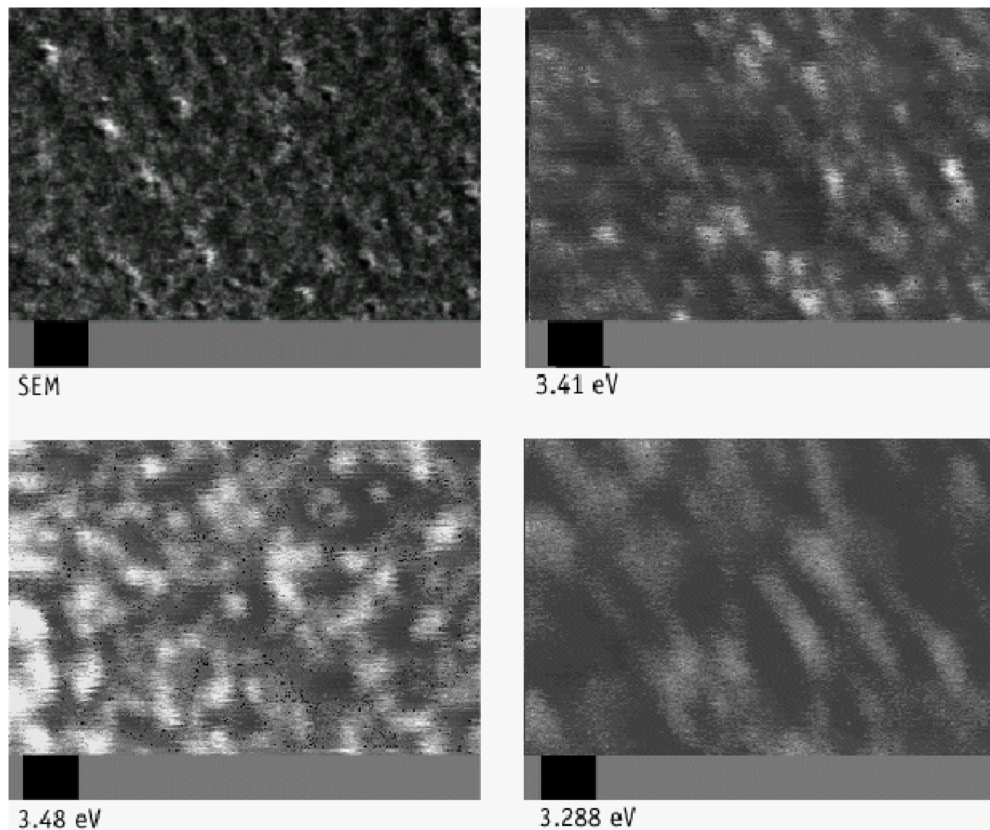

Figure 6. An SEM image and comparative CL images acquired at 15 keV from the 10° epilayer. The image acquired at 3.48 eV corresponds to D^0X emission, where the donor is the silicon dopant. The image acquired at 3.41 eV corresponds to excitonic emission attributed to structural defects. The image acquired at 3.288 eV corresponds to DAP emission. The black scale marker corresponds to 10 µm.

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