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Microphotoluminescence mapping of laterally overgrown GaN layers on patterned Si (111) substrates

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Spatial distribution of optical properties of GaN layers grown on patterned Si (111) substrates by maskless metalorganic chemical vapor deposition has been investigated. The Si substrates were prepared with a pattern of 1.5 μm diameter holes at a 3.5 μm distance from each other. The holes were overgrown by GaN until coalescence, creating GaN areas with no substrate underneath. Microphotoluminescence mapping measurements with 0.8 μm lateral resolution show a five-fold increase in luminescence intensity coming from the overhang areas as compared to the layer directly over the substrate. This is accompanied by a slight redshift of the luminescence peak wavelength. Photoelectrochemical etching shows that the dislocation density is much lower in those areas while the photoluminescence redshift is attributed to lesser strain relaxation resulting from a lower dislocation density. © 2005 American Institute of Physics.

Despite extensive research into the growth of gallium nitride (GaN), there are still no industrially available free-standing GaN wafers. This fact enforces the use of foreign substrates and, therefore, results in layers exhibiting large numbers of dislocations. Among the possible substrates, silicon offers some potential advantages as a substrate for GaN; these include: Low cost, large-scale availability, and well-established processing technology. Additionally, growth of nonpolar r-plane GaN layers can be realized on Si (001) by careful control of the nucleation layer. However, silicon's large lattice mismatch with GaN (17%) and additionally their difference in thermal expansion coefficients makes it very challenging to grow GaN layers of high enough quality to ensure proper operation of devices. Apart from a high density of dislocations (in the order of $10^{10}$cm$^{-2}$) created at the substrate-layer interface, the thermal expansion coefficient mismatch leads to the generation of cracks for layers thicker than 1 μm. Several methods of growth optimization can be used to improve these unfavorable characteristics, for instance, various modifications to the AlN nucleation layer. An additional SiN$_x$ layer, or AlGaN/GaN superlattice structure also serves to release strain and lower the overall dislocation density. A significant improvement can be obtained by using the epitaxial lateral overgrowth (ELO) technique. In fact, dislocation densities as low as $5 \times 10^7$cm$^{-2}$ have been obtained. However, these techniques require complicated and costly ex situ processing during an interruption of the GaN growth.

The present letter reports on the photoluminescence (PL) characteristics of ELO GaN epilayers on (111) Si substrates patterned with hole openings. The results are discussed in terms of strain distribution and dislocation density as shown by photoelectrochemical etching (PEC) etching. According to PL results, these layers exhibit characteristics favorable for the growth of low cost light-emitting diode structures and are therefore industrially feasible.

Prior to the growth of the GaN layers, the Si substrates were etched in a hole pattern creating 4 μm deep circular holes with a diameter of 1.5 μm and center to center distance of 3.5 μm, arranged in a hexagonal plan. The etching was done in a series of dry etching steps: Inductively coupled plasma (ICP) etching followed electron cyclotron resonance (ECR) plasma etching. First, an oxide mask was deposited in an ICP C$_4$F$_8$ plasma, and, subsequently reactive ion etching-oxygen plasma etching was used to remove the organic resid-

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FIG. 1. (a) SEM micrograph at 45° inclination of the GaN layer overgrowing the pits in the substrate. Complete coalescence can be observed, however, the growth run was too short for complete smoothing of the surface. (b) Schematic drawing of the overgrowth process. The arrows indicate lateral and vertical growth directions and solid line indicates the sample surface at the end of the growth run. The dotted lines show growth surfaces during the growth and the dashed line shows projected surface if the growth run had been continued.
dues. Finally, the actual etching of Si substrate was carried out in SF$_6$/O$_2$ ECR plasma. Under these conditions, the etch rate is estimated to be 300 nm/min. The depth of 4 μm is chosen to prohibit nucleation of the GaN layer inside the holes or at the ridges. A more extensive description of the etching process is provided by Haffouz et al.\textsuperscript{8}

The GaN layers have been grown by metalorganic chemical vapor deposition on the patterned (111) Si substrates. Trimethylgallium, trimethylaluminum, and ammonia were the precursors for gallium, aluminum, and nitrogen, respectively. An optimized AlN nucleation layer—needed for the correct growth—is deposited first at 850 °C. Later, the sample is heated to 1170 °C to commence the growth of the main GaN layer of Ga polarity.\textsuperscript{8} Figure 1(a) shows a scanning electron microscopy (SEM) micrograph of the cleaved GaN layer on top of the patterned Si substrate. It can be clearly seen that nucleation occurred on the unetched surfaces and that the growth then proceeded laterally and vertically. Lateral growth resulted in the closing of the holes etched in the substrate creating crystallographically oriented inverted pyramidal shapes with the slopes made of <1011) planes. A schematic drawing of the growth process is presented in Fig. 1(b); the arrows indicate both lateral and vertical growth directions, the solid line represents samples’ surface at the end of the growth run, dotted lines represent sample surface during growth, and the dashed line corresponds to the expected surface if the growth had been continued. At the growth thickness of 2-2.3 μm, the holes are on the threshold of being completely overgrown, however the surface exhibits hexagonally shaped pits. This sample, as shown in Fig. 1, has been used in all of the PL measurements.

Micro-PL (μ-PL) mapping measurements were performed at room temperature on a custom-built setup. HeCd laser provided excitation at 325 nm with power of 20 mW; the laser beam was routed into a diffraction-limited aspheric lens by means of a dichroic mirror. The same lens was used to collect PL which after passing through the same dichroic mirror was collected into a Coherent\textsuperscript{TM} liquid light guide. The luminescence was analyzed by a 1-m Spex 1704 monochromator equipped with a back illuminated ultraviolet (UV)-enhanced charge coupled device camera by Jobin Yvon. The spectral resolution was measured to be less than 0.3 Å, and the spatial resolution was measured to be 0.8 μm, which is quite satisfactory taking into account that the theoretical diffraction limit for the lens used (numerical aperture 0.6) is 0.66 μm.

PEC etching was employed to visualize the threading dislocations and to give an estimate of the dislocation density. It was performed in a stirred aqueous KOH solution (0.004 M) at room temperature. The UV illumination was provided by a 450W UV-enhanced Xe lamp. The nature and physical background of PEC etching can be found elsewhere.\textsuperscript{9}

Figure 2 shows the GaN layer after PEC etching. Due to its nature, PEC etching reveals dislocations of edge, mixed, and screw type as well as narrow inversion domain boundaries. Because the sample exhibits Ga-polarity, formation of inversion domains does not seem probable. Therefore, all the whiskers visible on the figure are attributed to dislocations and the density is calculated to be ~8 × 10$^9$ cm$^{-2}$. No whiskers can be seen in the overgrown areas. There are two reasons for that. First, as shown in Ref. 8, the dislocations which nucleated at the edge of a hole in the Si substrate tend to bend toward the center and either annihilate or become basal plane dislocations. Second, no new dislocations are generated in the overhanging GaN layer. The dislocations are known to suppress band edge luminescence\textsuperscript{10} and this is clearly reflected in Fig. 3. Figure 3(a) shows a map of luminescence intensity at the spectral position of the peak for a
Fig. 3(a), the bright circular areas represent over-the-void layers of dislocations. There are two major reasons for this behaviour, as spots corresponding to areas grown over a void should be relaxed and therefore have emission at comparably higher energy. Figure 3(b) shows exactly the opposite behavior, as spots corresponding to areas grown over the voids show even lower emission energy than those directly over the substrate.

The reason for such an effect can be explained by the dislocation formation mechanism and the strain relaxing properties of dislocations. There are two major reasons for the existence of dislocations. One is the fact that growth is usually started as three-dimensional growth and the layers coalesce at a later time which induces the creation of low angle grain boundaries at the spots of coalescence, creating rows of dislocations. The second reason is the differences in thermal expansion coefficients which induce stress upon cooling of the sample and dislocations appear postgrowth as a means of relaxing that stress. Differences between the density of GaN nucleation islands result in the general differences of statistical dislocation density between GaN layers grown on different substrates.

In the case of this sample, the PL results shown in Fig. 3(b) suggest that GaN areas grown over a void have higher tensile stress than the ones over Si substrate. This can be explained by the fact that the areas over Si have very high in-grown dislocation density. Those dislocations relax strain, whereas a much lower dislocation density in the areas over the voids does not present such an opportunity. The mismatch in the thermal expansion coefficients between Si and GaN induces tensile strain when the sample is cooled down to room temperature, which can be partially relaxed by the in-grown dislocations directly over substrate but will be much less relaxed over the void due to the lack of dislocations.

In summary, it has been shown that GaN layers can be grown on patterned Si substrates exhibiting locally much lower dislocation density and better structure quality. This enhancement is accompanied by a large increase in luminescence intensity and a slight shift of luminescence peak towards lower energies. This redshift indicates higher levels of tensile stress which is caused by a locally lower density of dislocations.

\[ \mu \text{-PL mapping has been used to study local optical properties of the samples and it is proved to be a suitable tool for local strain mapping of GaN layers.} \]

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