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

L. Macht, a P. R. Hageman, S. Haffouz, and P. K. Larsen

Experimental Solid State Physics III, Institute for Molecules and Materials, University of Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands

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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. [DOI: 10.1063/1.2042546]

Despite extensive research into the growth of gallium nitride (GaN), there are still no industrially available freestanding 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 10¹⁰ 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 SiNₓ 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 × 10⁷ 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 (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 C₄F₈ plasma, and, subsequently reactive ion etching-oxygen plasma etching was used to remove the organic resi-

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.
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. Figure 1(a) shows a micrograph at 45° inclination of the GaN layer after PEC etching. Single whiskers represent threading dislocations. Finally, the actual etching of Si substrate was carried out in SF6/O2 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. 8

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™ 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.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 luminescence10 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 15×15 µm area, the parameter mapped is intensity at the position of the peak. The position of the peak shifts depending on the location. (b) A µ-PL map of a 15×15 µm area, the parameter mapped is the spectral position of the peak. Emission wavelength is presented in Å.
15 × 15 μm area. This peak position is not constant and it
varies from spot to spot, the intensity of luminescence is
taken at the actual spectral position of the peak for each point
and not at an arbitrary energy. Thus, the measured intensity
of overgrown areas is, on average, five times higher than
intensity measured in the areas directly over the substrate. In
Fig. 3(a), the bright circular areas represent over-the-void
layer.

A corresponding map of the same area shown in Fig.
3(b) shows peak emission energy. The luminescence from
the overgrown areas of the GaN layer have markedly lower
energy than those from areas above the Si substrate and the
values are 3.400 eV and 3.405 eV, respectively. Figure 4
shows PL spectral data from two points designated as A
and B in Fig. 3. The scans show the difference between
luminescence intensity as well as the spectral position of
the peaks. The latter value is consistent with value of 3.406 eV
reported elsewhere for GaN/Si layers. For comparison,
GaN/sapphire which is believed to be in small compressive
strain at room temperature emits at 3.424 eV, while com­
pletely unstrained GaN emits light at 3.420 eV.

GaN layers grown on Si substrates are believed to be in
tensile strain at room temperature resulting in the emission
wavelength shifted toward lower energies. This is also the
case for our samples as can be seen by lower emission energies
than those for relaxed GaN. It is also believed that layers
grown over a mask in the ELO process or, in this case, over
a void should be relaxed and therefore have emission at com­
parably higher energy. Figure 3(b) shows exactly the oppo­
site 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 lumines­
cence intensity and a slight shift of luminescence peak to­
ward lower energies. This redshift indicates higher levels of
tensile stress which is caused by a locally lower density of
dislocations.

μ-PL mapping has been used to study local optical prop­
erties of the samples and it is proved to be a suitable tool for
local strain mapping of GaN layers.

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