

Selective Area Growth of GaN Directly on (0001) Sapphire by the HVPE Technique

Raj Singh¹, Richard J. Barrett¹, John J. Gomes¹, Ferdynand P. Dabkowski¹ and T.D. Moustakas²

¹Laser Diode Manufacturing and Development, Polaroid Corporation,

²Department of Electrical and Computing Engineering and Center for Photonics Research, Boston University,

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In this paper, we report on the selective area growth (SAG) of GaN directly on patterned c-plane sapphire substrates by hydride vapor phase epitaxy (HVPE). A number of researchers have reported that the HVPE growth technique, unlike the MBE and MOCVD methods, is capable of producing device quality GaN films without the need for any low temperature nucleation/buffer layers. The density of edge dislocations in these HVPE films decreases dramatically as the film thickness is increased, and the dislocation density values for thick films ($> 10\mu\text{m}$) are comparable to those reported for the best GaN films grown by other methods on c-sapphire. These advantages of the HVPE growth technique makes it possible to achieve high quality selective area growth of GaN directly on c-sapphire substrates.

C-plane sapphire substrates were coated with PECVD SiO_2 and photolithographically patterned with different size and shape openings. Subsequently, these patterned substrates were introduced in a horizontal, hot-wall quartz reactor for the GaN growth. It was observed that single crystal GaN growth was preferentially initiated in the openings in the oxide layer. This selective area growth was followed by epitaxial lateral overgrowth (ELO), leading to the formation of hexagonal GaN prisms terminated in smooth, vertical (1100) facets. We have been successful in shearing these pyramid structures from the sapphire substrates as individual devices, which do not require any post-growth etching for feature definition. This procedure allows for the dramatic reduction of the process complexity and the duration and expense for GaN growth for device applications. Stimulated emission results on these self-formed optical cavities are also presented.

1 Introduction

In recent years, there has been renewed interest in the Hydride Vapor Phase Epitaxy (HVPE) method for the growth of GaN layers. This technique is capable of producing device quality GaN films without the need for any low temperature nucleation/buffer layers. The density of edge dislocations in the HVPE grown GaN films decreases dramatically as the film thickness is increased ($> 10\mu\text{m}$), and the dislocation density values ($\sim 10^8\text{ cm}^{-2}$) are comparable to those reported for the best GaN films grown by other methods [1]. A number of recent studies have indicated that the crystalline quality of heteroepitaxial GaN films can be further improved by epitaxial lateral overgrowth (ELO) on patterned GaN substrates [2-5]. This technique involves the heteroepitaxial growth of a GaN layer on sapphire or SiC substrate. This step is followed by the deposition of a

dielectric (SiO_2) film on the GaN layer. The dielectric layer is patterned appropriately and then used as the substrate for the ELO growth of GaN. Edge dislocation densities as low as 10^4 cm^{-2} can be obtained by using this improved growth technique. The ELO-GaN layers have been used to make commercially viable GaN lasers with an estimated lifetime in excess of 10,000 hours [2]. Although the majority of ELO-GaN results have been reported for the MOCVD growth method, there have been a few reports of the ELO-GaN prepared by the HVPE method [3] [4]. However, all these ELO studies have involved the growth of two separate layers of GaN interspersed with a patterned dielectric mask layer. This can prove to be a fairly tedious and lengthy growth procedure due of the multiple growth steps involved.

In this paper, we report the selective area growth (SAG) of GaN directly on patterned (0001) sapphire

substrates by the HVPE technique, thus eliminating an entire GaN growth sequence. The selective area growth leads to epitaxial lateral overgrowth (ELO) of the GaN on the patterned substrate. Bare sapphire substrates were coated with a silicon dioxide layer. The SiO₂ was photolithographically patterned and etched to make selective area patterns on the sapphire substrate. These substrates were used for the ELO-GaN growth. The pattern was chosen in such a manner that coalescence between the individual GaN islands nucleated in the openings was not possible in the growth times employed for these samples. This allowed for the growth of segregated GaN structures, in the form of hexagonal pyramids, on these patterned substrates. These structures were studied by optical and scanning electron microscopy. Photoluminescence and optical pumping studies were also performed to characterize the ELO-GaN layers.

2 Experimental Methods

Figure 1 shows a schematic of the HVPE growth system used in these studies. It consisted of a fused quartz growth chamber, heated in a three zone resistive furnace. The Group III source was gallium chloride gas produced by passing hydrogen chloride over gallium metal at 900 °C. Ammonia gas, diluted in nitrogen, was used as the group V source. Nitrogen gas was used as the gallium chloride diluent and also as the main carrier gas.

ELO-growth of GaN was carried out on patterned sapphire substrates. C-plane sapphire wafers were coated with 5000 Å of PECVD deposited SiO₂. Subsequently, photolithography and wet etching were used to pattern the SiO₂/sapphire wafers. A schematic of one of the mask patterns used in this study is shown in Figure 2. It consisted of 100 µm diameter circles centered on grid spacing of 500 µm. The oxide layer was etched to reveal the surface of sapphire under the circles. After chemical degreasing, the patterned substrates were mounted on a quartz holder and introduced in the growth chamber. They were thermally degassed at the growth temperature of 1050-1070 °C in a nitrogen ambient. After half an hour of degassing, ammonia was introduced for surface nitridation for 15 minutes. Subsequently, hydrogen chloride gas flow was started to initiate the growth of GaN. The duration of the growths was typically between 15 to 60 min. No intentional doping was used during the growth of the GaN layers reported in this study. The growth parameters were first optimized for GaN deposited on bare sapphire (unpatterned) substrates. Under these conditions, uniform coverage of the entire substrate by highly transparent GaN was obtained. The growth rate was controlled by the

HCl flow rate and was typically between 15-25 µm/hr on unpatterned sapphire substrates.

After the growth, the samples were cooled to room temperature in a nitrogen stream. Visual inspection of the samples revealed that GaN had preferentially nucleated in the openings in the oxide layer. Polycrystalline deposits of GaN on the remaining portion of the masking dielectric layer were also observed. These samples were subsequently etched in a buffered oxide etch solution, resulting in the removal of the SiO₂ masking layer as well as the polycrystalline GaN deposits.

3 Experimental Results and Discussion

Figure 3 shows the post-growth surface of the ELO-GaN/sapphire substrate after the removal of the masking SiO₂ layer. This substrate was patterned with 100 µm holes on 500 µm centers in the oxide layer. The circular openings were completely covered with the hexagonal shaped GaN pyramids as a result of lateral overgrowth on top of the oxide mask. These individual GaN pyramids were all oriented in the same direction on a wafer and their edges were terminated in the m-planes (01 $\bar{1}$ 0).

Figure 4 shows the edge and top view of one of these hexagonal GaN islands. The mask pattern used for this particular sample consisted of 25 µm square openings on 500 µm centers. In plan view, two surface morphologies were observed on the top of the GaN pyramid. In the center region directly above the opening in the silicon dioxide layer, the surface morphology was rough and showed signs of coalescence of individual nuclei, which initiated on the sapphire surface. On the other hand, the laterally overgrown material had a much more smoother morphology. However, the edge view revealed that the top of the epitaxial overgrowth region was not completely flat. The differences in the surface morphologies between the laterally overgrown and directly grown GaN were expected due to the difference in the defect structure. The edge view showed that the walls of the hexagonal structure were very smooth and perpendicular. No features were observable on the wall surfaces at higher (>100,000) magnification of the SEM.

Room temperature photoluminescence for a GaN pyramid structure is shown in Figure 5. The sample was excited with a 10mW He-Cd laser incident normally to the substrate surface. The emission was dispersed through a 0.5m spectrometer to obtain the wavelength scan. The room temperature PL spectrum showed the near band edge emission at 361 nm with a FWHM of 12.5 nm. No yellow luminescence was observed under these excitation conditions, attesting to the good material quality of the pyramid structure. The background electron concentration, for thick GaN films grown under

similar conditions was determined by Hall effect measurements to be between 10^{17} to 10^{18} cm^{-3} .

Room temperature optical pumping studies on these GaN hexagon structures were carried out using a focussed nitrogen laser pump at 337 nm with a peak power of 40 kW (10 ns pulse). Stimulated emission, as indicated by linewidth narrowing and red shift of the luminescence, was observed. The threshold of stimulated emission was at an incident power density of 3.4 MW/cm^2 . This value of threshold is significantly lower than that previously reported (30 MW/cm^2) for similar structures prepared by the MOVPE method [5]. The peak of the stimulated emission was observed at 377.8nm with a FWHM of 1.2 nm, at twice the threshold pump intensity. Line width narrowing and the output emission power intensity as a function of incident power density are shown in Figure 6. The slope of the peak intensity versus power density increased by a factor of 172 above threshold. We expected to observe longitudinal lasing modes for these structures with smooth and parallel walls. However, the hexagonal structure does not have simple Fabry-Perot cavity modes and the power density of the pump varied greatly ($> 10\%$) over spectral collection time. Thus no longitudinal modes were observed.

4 Conclusion

In conclusion, we have demonstrated the selective area growth of GaN directly on patterned sapphire substrates by the HVPE method. The growth of GaN on patterned c-plane sapphire resulted in hexagonal shaped GaN pyramids as a result of lateral overgrowth. The surface morphology of the overgrown regions was significantly improved as compared to the GaN grown directly on sapphire. With proper selection of the mask pattern, individual GaN pyramids with smooth and vertical walls were directly grown on the substrates without the need for any post-growth etching. Optically pumped stimulated emission was observed from these as-grown individual pyramid structures at room temperature. The threshold power density for stimulated emission was found to be 3.4 MW/cm^2 .

Further improvements in the growth of ternary alloys and p-type doping of GaN by the HVPE growth technique, coupled with the ELO-GaN growth will make it possible to fabricate complete device structures without post-growth etching. This will make it possible to fabricate atomically smooth laser mirror facets in-situ, resulting in improved device performance. Specifically, the growth of a-plane GaN ($11\bar{2}0$) on r-plane sapphire ($011\bar{2}$) will result in rectangular features. This may be more suitable for the definition of in-situ rectangular laser cavities.

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FIGURES

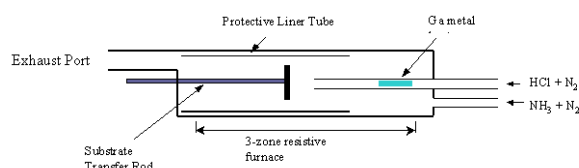


Figure 1. Schematic of the HVPE growth system for GaN selective area growth



Figure 2. Schematic of the pattern used for the selective area coverage of the sapphire substrates. Textured area represents the SiO_2 layer while the clear circles show the exposed sapphire substrate.

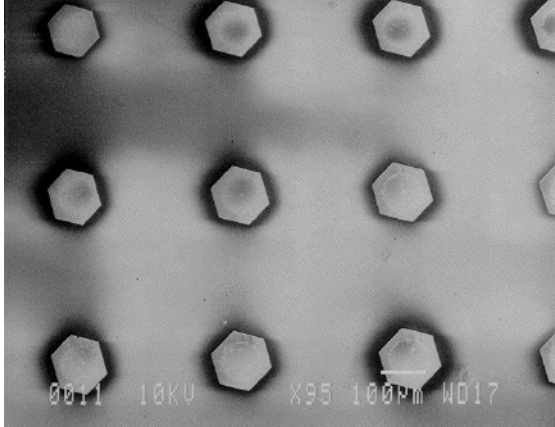


Figure 3. SEM image of the selective area grown GaN pyramid structures.

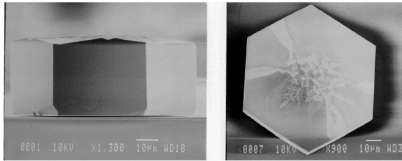


Figure 4. Edge and top view of one of the GaN hexagonal pyramid structures showing the texture of the top surface and the sidewalls. The edges of the square openings were oriented along the m -plane $(01\bar{1}0)$.

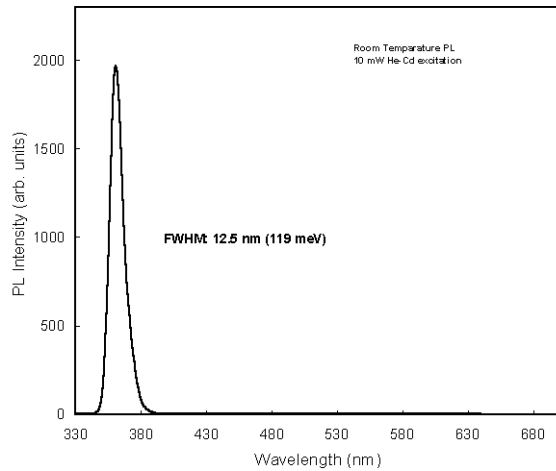


Figure 5. Room temperature PL from a GaN film (unintentionally doped) grown by HVPE

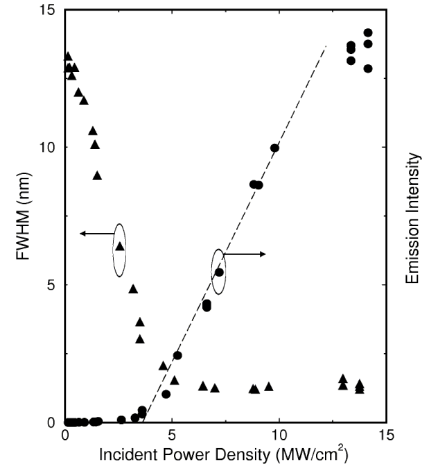


Figure 6. Stimulated emission from the GaN hexagonal pyramid structure. Peak intensity and emission linewidth as a function of incident power density.