ORGANOMETALLIC VAPOR PHASE LATERAL EPITAXY OF LOW DEFECT DENSITY GaN LAYERS

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ABSTRACT

Lateral epitaxial overgrowth (LEO) of GaN layers has been achieved on 3 μm wide and 7 μm spaced stripe windows contained in SiO_2 masks on GaN/AlN/6H-SiC(0001) substrates via organometallic vapor phase epitaxy (OMVPE). The extent and microstructural characteristics of lateral overgrowth were a complex function of stripe orientation, growth temperature and triethylgallium (TEG) flow rate. A high density of threading dislocations, originating from the interface of the underlying GaN with the AlN buffer layer, were contained in the GaN grown in the window regions. The overgrowth regions, by contrast, contained a very low density of dislocations. The second lateral epitaxial overgrowth layers were obtained on the first laterally grown layers by the repetition of SiO_2 deposition, lithography and lateral epitaxy.

INTRODUCTION

Conventional heteroepitaxial growth of GaN on low temperature GaN or AlN buffer layers previously deposited on Al_2O_3 and SiC substrates results in films containing high dislocation densities (10^8-10^{10} cm^{-2}) resulting from the mismatches in lattice parameters between the buffer layer and the film and/or the buffer layer and the substrate. This is believed to limit the performance of selected types of devices. Several groups[1-3] are investigating the growth of bulk GaN crystals from which would be derived substrates for growth of GaN and related films. However, at present, the size and growth rate of these crystals are limited.

The initial results of research regarding the lateral epitaxial overgrowth (LEO) of GaN layers on amorphous SiO_2 have been reported by the authors[4,5]. Additional results from studies[6] concerning the selective growth of GaN hexagonal pyramids for field emitters showed that lateral overgrowth unintentionally occurred over the SiO_2 mask layers under selected growth conditions. Transmission electron microscopic study also revealed that these overgrown regions of pyramids contained a very low density of dislocation (<10^4 cm^{-2})[7].

Following the reports of our research[4,5], Nakamura[8] reported a substantially increased laser diode life time of approximately 10,000 hours at room temperature in devices fabricated using the LEO technique. As a result, the LEO technique has received considerable interest from the III-Nitride research community.

The first lateral epitaxial overgrowth and coalescence of GaN stripes patterned in SiO_2 masks deposited on GaN film/AlN buffer layer/6H-SiC(0001) substrate assemblies and the second lateral epitaxial overgrowth on the first laterally overgrown GaN layer have more recently been accomplished and are described in the following sections.

EXPERIMENTAL PROCEDURES

Substrates for the lateral epitaxy studies were prepared, as shown schematically in Figure 1, via growth of 1.5-2.0 μm thick GaN films at 1000°C on high temperature (1100°C) AlN buffer

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layers previously deposited on 6H-SiC(0001) substrates in a cold-wall, vertical and RF inductively heated organometallic vapor phase epitaxy (OMVPE) system. Additional details of the growth experiments are given in Ref. 9. An SiO_2 mask layer (thickness = 1000 Å) was subsequently deposited on each GaN/AlN/6H-SiC(0001) sample via low pressure chemical vapor deposition at 410°C. Patterning of the mask layer was achieved using standard photolithography techniques. Etching was accomplished with a buffered HF solution. The pattern contained 3 μm wide, parallel stripe openings spaced 7μm from edge-to-edge and oriented along the $<11\bar{2}0>$ and $<1\bar{1}00>$ directions in each GaN film. Prior to lateral overgrowth, the patterned samples were dipped in a 50% buffered HCl solution to clean the underlying GaN layer.

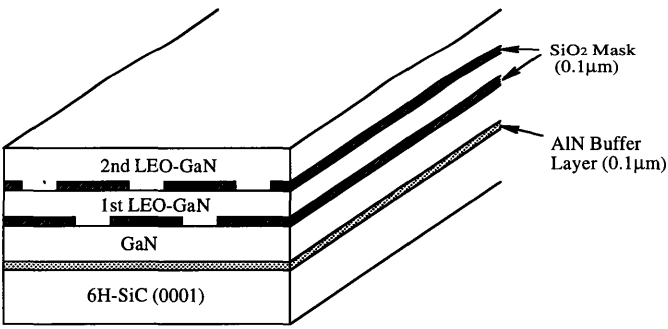
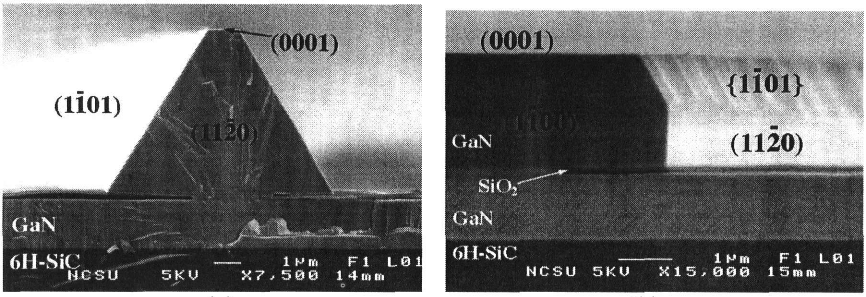


Figure 1. Schematic diagram showing the first and second lateral epitaxial overgrowths of GaN layers from within stripe openings and over the top surfaces of two SiO_2 masks.

The lateral overgrowth of GaN was achieved at 1000 - 1100°C and 45 Torr. Triethylgallium (13 - 39 μmol/min) and NH_3 (1500 sccm) precursors were used in combination with a 3000 sccm H_2 diluent. The second lateral epitaxial overgrowth was conducted on the first laterally grown layer via the repetition of SiO_2 deposition, lithography and lateral epitaxy, as shown in Figure 1. The samples were characterized using scanning electron microscopy (SEM-JEOL 6400 FE), atomic force microscopy (AFM-Digital Instrument NanoScope III) and transmission electron microscopy (TEM-TOPCON 002B, 200KV).

RESULTS AND DISCUSSION

Figure 2 shows the representative cross-sectional morphologies of two GaN stripes selectively grown for 60 min along $<11\bar{2}0>$ and $<1\bar{1}00>$. Truncated triangular stripes having $(1\bar{1}01)$ slant facets and a narrow (0001) top facet were observed for window openings along $<11\bar{2}0>$. Rectangular stripes having a (0001) top facet, $(11\bar{2}0)$ vertical side facets and $(1\bar{1}01)$ slant facets developed in samples grown along $<1\bar{1}00>$. Observations via SEM of GaN stripes grown for different times up to 3 min revealed similar morphologies regardless of stripe orientation. The stripes subsequently developed into different shapes if the growth was continued. The amount of lateral growth exhibited a strong dependence on stripe orientation. Results obtained under various growth conditions showed that the lateral growth rate of the $<1\bar{1}00>$ oriented stripes was much faster than those along $<11\bar{2}0>$. We believe that the different morphological development as a function of window orientation is related to the stability of the crystallographic planes in the GaN structure. Stripes oriented along $<11\bar{2}0>$ always had wide $(1\bar{1}01)$ slant facets and either a very



(a) (b)

Figure 2. Scanning electron micrographs showing the morphologies of GaN layers grown on stripe openings oriented along (a) <11$\bar{2}$0> and (b) <1$\bar{1}$00>.

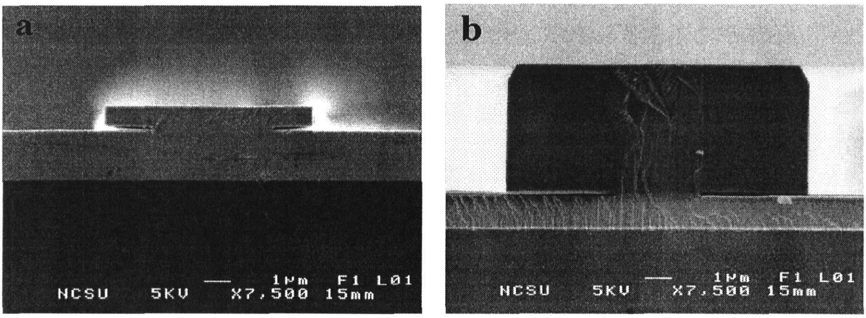


Figure 3. Scanning electron micrographs of <1$\bar{1}$00> oriented GaN stripes grown at TEG flow rates of (a) 13 µmol/min and (b) 39 µmol/min for 60min.

narrow or no (0001) top facet depending on the growth conditions. The primary reason for this is that (1$\bar{1}$01) is the most stable plane in the GaN wurtzite crystal structure, and the growth rate of this plane is lower than that of others. As shown in Figure 2 (b), the {1$\bar{1}$01} planes of the <1$\bar{1}$00> oriented stripes were wavy, which implies the existence of more than one Miller index. It is believed that competitive growth of selected {1$\bar{1}$01} planes occurs during the deposition which causes these planes to become unstable and which causes their growth rate to increase relative to that of the (1$\bar{1}$01) plane of stripes oriented along <11$\bar{2}$0>[4].

The morphological development of the GaN stripes also depends on the flow rate of the TEG, as shown in Figure 3. An increase in the supply of TEG increased the growth rate of the stripes in both the lateral and the vertical directions. However, the lateral/vertical growth rate ratio decreased from 1.7 at the TEG flow rate of 13µmol/min to 0.86 at 39 µmol/min. This increased influence on growth rate along <0001> relative to that of <11$\bar{2}$0> with TEG flow rate is believed to be related to the type of reactor employed in this research, wherein the reactant gases flow vertically and perpendicular to the substrate. The considerable increase in the concentration of the Ga species on the surface may sufficiently impede their diffusion to the {1$\bar{1}$01} planes such that chemisorption and GaN growth occur more readily on the (0001) plane. The morphologies of the GaN layers were also a strong function of the growth temperatures[4]. Stripes grown at 1000°C possessed a

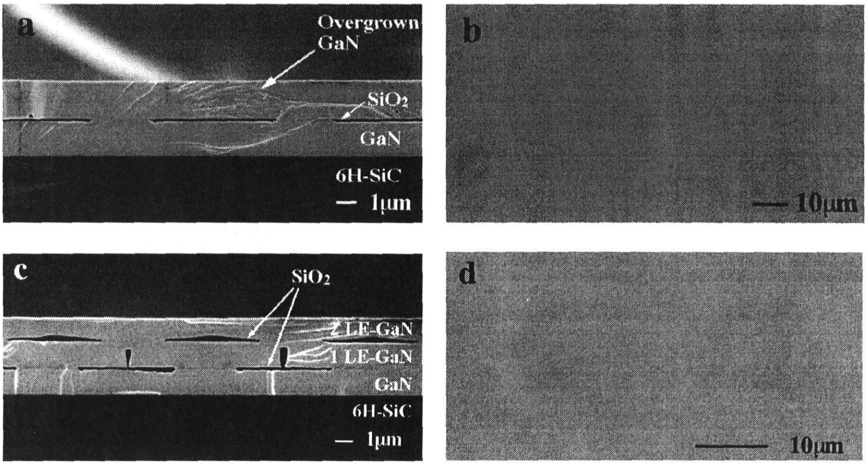


Figure 4. Cross-section and surface SEM micrographs of the first, (a) and (b), and second, (c) and (d), coalesced GaN layers, respectively, grown on 3 μm wide and 7 μm spaced stripe openings oriented along $<1\bar{1}00>$.

truncated triangular shape. This morphology gradually changed to the rectangular cross-section as the growth temperature was increased.

The first lateral epitaxial overgrown GaN layers with thickness of 2μm were obtained using 3 μm wide stripe openings spaced 7 μm apart and oriented along $<1\bar{1}00>$ (Figure 4 (a)). The growth parameters were 1100°C and a TEG flow rate of 26 μmol/min. A plan view of the first lateral epitaxy layer revealed a microscopically flat and pit-free surface (Figure 4 (b)). Atomic force microscopy showed the surfaces of the laterally grown GaN layers to consist of a terrace structure having an average step height of 0.32 nm. The average RMS roughness values of the regrown and overgrown areas were 0.23 nm and 0.29 nm, respectively; these are similar to the values obtained for the underlying GaN films.

The second lateral epitaxial overgrowth of GaN layers on the first lateral epitaxy layers has also been achieved as shown in Figure 4 (c). This achievement was made through the repetition of SiO_2 deposition, lithography and lateral epitaxy. Each black spot in the overgrown GaN layers shown in Figure 4 (a) and (c) is a subsurface void which forms when two growth fronts coalesce. These voids were most often observed using the lateral growth conditions wherein rectangular stripes having vertical $\{11\bar{2}0\}$ side facets developed[5]. Surface morphology of the second overgrown layer was comparable to the first layer, as shown in Figure 4 (d). Cracks were occasionally observed along the coalesced interface under selected growth conditions, probably due to the thermal mismatch between the GaN layers and the SiO_2 mask.

The micrograph obtained via cross-sectional TEM and presented in Figure 5 shows a typical laterally overgrown GaN. Threading dislocations, originating from the GaN/AlN buffer layer interface, propagate to the top surface of the regrown GaN layer within the window regions of the mask. The dislocation density within these regions, calculated from the plan view TEM micrograph is approximately $10^9 cm^{-2}$. By contrast, there were no observable threading dislocations in the overgrown layer[7]. Additional microstructural studies of the areas of lateral growth obtained using various growth conditions have shown that the overgrown GaN layers contain only a few

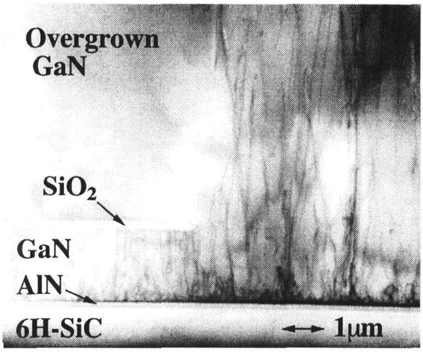


Figure 5. Cross-section TEM micrograph of a section of a laterally overgrown GaN layer on an SiO_2 mask region.

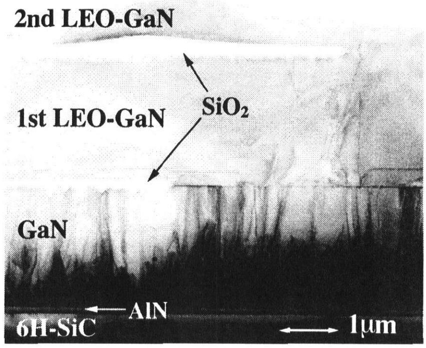


Figure 6. Cross-section TEM micrograph of a section of the second lateral epitaxial overgrown GaN layers.

dislocations. These dislocations formed parallel to the (0001) plane via the extension of the vertical threading dislocations after a 90° bend in the regrown region[7]. Plan view TEM observation revealed that these dislocations never propagate to the top surface of the overgrown GaN layers.

Cross-sectional TEM observation of the second LEO sample in the micrograph presented in Figure 6 shows that a very low density of dislocations parallel to the (0001) plane, formed via bending of threading dislocation, exist in the first and second LEO-GaN layers on the SiO_2 masks. The second SiO_2 mask is slightly misaligned relative to the first. These results suggest that very low defect density GaN layers can be fabricated by precise alignment of the mask in the second lithographic process.

Preliminary photoluminescence and cathodoluminescence results indicated that yellow emission (2.25eV) originates from the regrown regions having high dislocation densities; but only strong band edge emission has been observed from the overgrown layers. Donor-acceptor pair emission was also observed from the lateral overgrown layers. Studies including photoluminescence, cathodoluminescence and micro-Raman scattering are underway to determine correlations between the optical and the microstructural characteristics of the laterally overgrown GaN layers.

CONCLUSIONS

Lateral epitaxial overgrowth of GaN on SiO_2 masks containing stripe windows oriented along different orientations has been achieved via OMVPE. The morphological development of these stripes depended strongly on the stripe orientation and growth conditions. The first and second lateral epitaxy GaN layers with both extremely low densities of dislocations and smooth and pit-free surfaces has been obtained for the first time along $<1\bar{1}00>$ at 1100°C and a TEG flow rate of 26 μmol/min.

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REFERENCES

1. S. Porowski, M. Bockowski, B. Lucznik, M. Wroblewski, S Krukowski, I Grzegory, M. Leszczynski, G. Nowak, K. Pakula and J. Baranowski, Mat. Res. Soc. Symp. Proc. **449**, 35 (1996).
2. S. Kurai, Y. Naoi, T. Abe, S. Ohmi and S. Sakai, Jpn. J. Appl. Phys., **35**, L77 (1996).
3. C.M. Balkas, Z. Sitar, T. Zheleva, L. Bergmann, I.K. Shmagin, J.F. Muth, R. Kolbas, R J. Nemanich and R.F. Davis, Mat. Res. Soc. Symp. Proc. **449**, 41 (1996).
4. O.H. Nam, M.D. Bremser, T.S. Zheleva and R.F. Davis, The 39th Electronic Materials Conference, Fort Collins, Colorado, June 25-27 (1997).
5. O.H. Nam, M.D. Bremser, T.S. Zheleva and R.F. Davis, Appl. Phys. Lett., **71**, 2638, (1997).
6. O.H. Nam, M.D. Bremser, B.L. Ward, R.J. Nemanich and R.F. Davis, Jpn. J. Appl. Phys., **36**, L532 (1997).
7. T.S. Zheleva, O.H. Nam, M.D. Bremser and R.F. Davis, Appl. Phys. Lett., **71**, 2472 (1997).
8. S. Nakamura, The 2nd Int. Conf. Nitride Semiconductors, Tokushima, Japan, October 25-31, (1997)
9. T.W. Weeks Jr., M.D. Bremser, K.S. Ailey, E.P. Calson, W.G. Perry and R.F. Davis, Appl. Phys. Lett., **67**, 401 (1995).