

Lateral epitaxy overgrowth of GaN with NH₃ flow rate modulation

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We demonstrate the effects of NH₃ flow modulation on the lateral growth rate and morphology of GaN stripes employing lateral epitaxial overgrowth (LEO) by metalorganic chemical vapor deposition. The self-limiting growth mechanism, enhanced Ga diffusion on the (0001) plane, and Ga lateral supply are used to explain our observations. A lateral overgrowth rate to a vertical growth rate ratio of 2.1 and fully coalesced LEO GaN stripes after 1 h growth have been achieved. © 2000 American Institute of Physics. [S0003-6951(00)04136-X]

Lateral epitaxial overgrowth (LEO) of GaN on sapphire,¹ SiC,² and Si(111) (Refs. 2, 3, and 4) has demonstrated marked reductions in defect density in regions where the lateral overgrowth occurs over the dielectric mask. This has led to much improved performance of optoelectronic devices.^{5,6} In terms of efficiency as well as device application, it is desirable to maximize the lateral growth rate for fast coalescence of the LEO stripes with smooth top surfaces and sidewalls. The proper stripe orientation ($\langle 1\bar{1}00 \rangle$), growth temperature,⁷ V/III ratio, and fill factor⁸ were reported to be important factors to inhibit the slow $\{1\bar{1}01\}$ growth facets and to favor smooth vertical $\{1\bar{1}20\}$ facets that have a fast growth rate. The effects of Mg doping on the LEO of GaN were also reported in Ref. 9 in order to obtain fully coalesced LEO GaN stripes.

In this work, we demonstrate the effects of NH₃ flow modulation (FM) on the lateral growth rate and morphology of LEO GaN stripes, which is called FM-LEO in this study. During growth, we continuously inject trimethylgallium (TMGa) into the reactor but interrupt NH₃ flow for 5–15 s for every 20 s of GaN growth. It was found that the lateral growth rate was greatly enhanced and that the morphology of LEO GaN stripes changed from a triangular cross section to a rectangular cross section. Self-limiting growth and enhanced surface migration of Ga on the (0001) plane of LEO GaN stripes are used to explain our observations.

The FM-LEO of GaN was carried out in a commercial vertical geometry, low-pressure (76 Torr) metalorganic chemical vapor deposition (MOCVD) reactor with a closely spaced showerhead.¹⁰ TMGa and NH₃ were used as precursors with H₂ as the carrier gas. A 170 nm thick SiN_x mask was deposited on a 2 μm thick conventional GaN buffer layer by plasma enhanced chemical vapor deposition. Standard photolithography and reactive ion etching were used to form patterned stripes, with 5 μm wide openings and 15 or 40 μm spacings, oriented along the GaN $[1\bar{1}00]$ directions to ensure a higher lateral growth rate. The LEO GaN was grown at 1080 °C with TMGa and NH₃ flows of 38 μmol/min and 1 slm, respectively. The total gas flow in the reactor

is 6 slm. The as-grown samples were cleaved and characterized with a Philips XL-30 scanning electron microscope (SEM).

Figures 1(a)–1(d) show time charts of the NH₃ flow rate in the FM-LEO for samples A, B, C, and D, respectively. Since a TMGa flow of 38 μmol/min was always injected into the reactor during growth, we did not plot its flow rate in Fig. 1. Except for the NH₃ flow rate, the growth conditions are otherwise the same for all four samples. The most important aspect of our LEO growth is the periodic NH₃ interruption during growth. For every 20 s of GaN growth when both TMGa and NH₃ are injected into the reactor, we interrupted the NH₃ flow for 5, 10, and 15 s for samples B, C, and D respectively.

The cross-sectional SEM images of samples A, B, C, and D are shown in Figs. 2(a)–2(d), respectively. The growth time is 20 min, which includes both the GaN growth time and NH₃ interruption time. These samples have 5 μm mask openings and 40 μm spacing. Figure 2(a) shows that without the NH₃ interruption, the GaN stripe has almost no lateral overgrowth. The slow growth rate of the $\{1\bar{1}01\}$ fac-

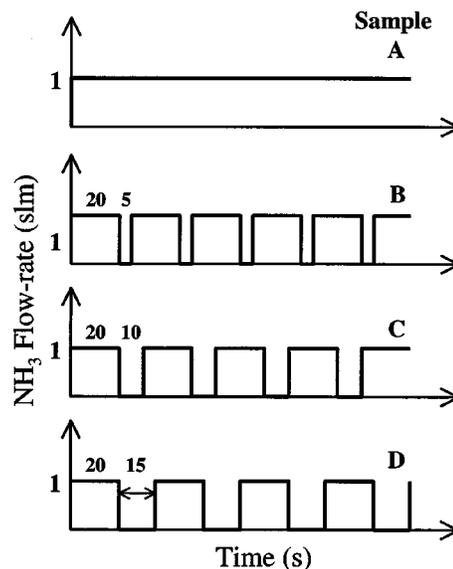


FIG. 1. Time charts of the NH₃ flow rate in FM-LEO (a) without NH₃ interruption and with (b) 5, (c) 10, and (d) 15 s periodic NH₃ interruptions for samples A, B, C, and D, respectively.

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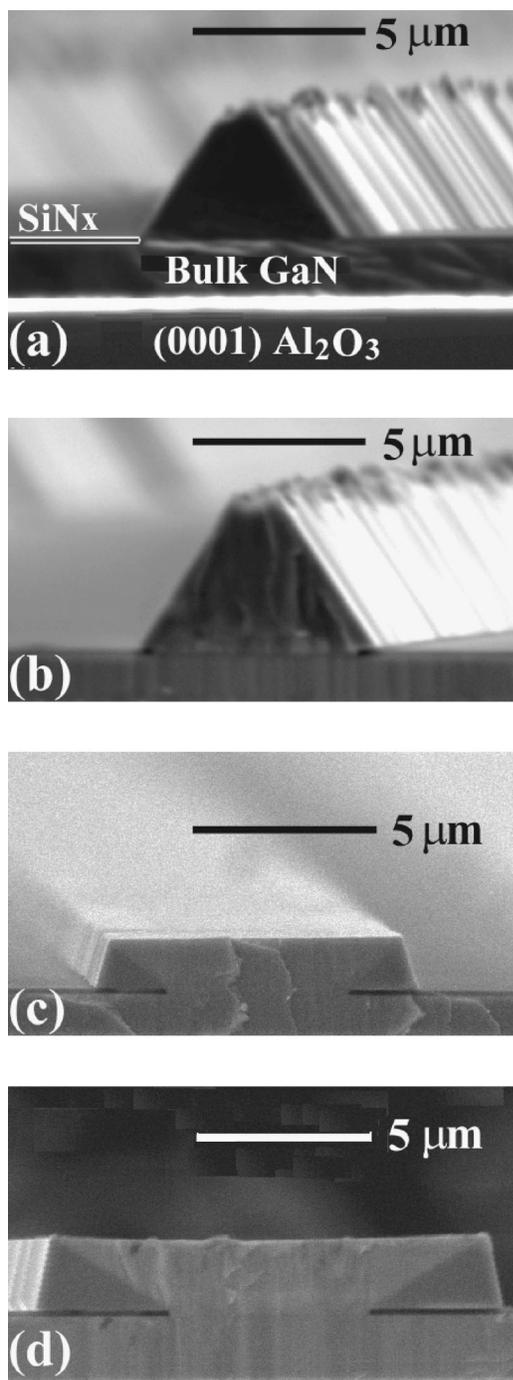


FIG. 2. Cross-sectional SEM images of sample (a) A, (b) B, (c) C, and (d) D after 20 min growth.

ets and fast growth rate of the $\{0001\}$ facets lead to the inclined sidewalls seen on sample A. Further growth leads to completely pinched off stripes (not shown here). The top of the GaN stripes is not flat and has a high density of pits.^{11,12} The exact mechanism of pit formation is not clear and our observations are not sensitive to the growth temperature or V/III ratio. However, the polarity of the GaN buffer may play an important role, since the polarity and surface diffusion of Ga can profoundly change the GaN thin film morphology.^{13,14}

With the introduction of a periodic 5s NH_3 interruption, the lateral growth rate was increased, as shown in Fig. 2(b). Within 20 min, the lateral overgrowth is about $0.5 \mu\text{m}$ but

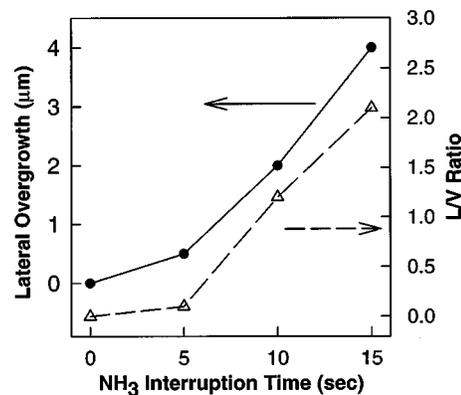


FIG. 3. Lateral overgrowth and L/V ratio as a function of NH_3 interruption time.

the stripe still holds the inclined $\{1\bar{1}01\}$ sidewall and rugged surface consisting of a high density of pits. The lateral to vertical growth rate ratio (L/V ratio) is only ~ 0.1 . Figure 2(c) shows that a dramatic change in stripe morphology occurs when the periodic NH_3 interruption time was increased to 10 s for every 20 s of GaN growth. The LEO stripe has a rectangular cross section and very smooth top surface and sidewall. The L/V ratio was increased from ~ 0.1 to 1.2, a factor of 12 increase. Although the sidewall is still not completely vertical, the proper choice of fill factors (ratio of mask opening to the periodicity of the patterning), V/III ratio, and NH_3 interruption time will result in vertical walls, as will be discussed later. The increase of NH_3 interruption time to 15 s for sample D further increased the lateral overgrowth to $4 \mu\text{m}$ in 20 min and the L/V ratio to 2.1 as shown in Fig. 2(d). The lateral overgrowth and L/V ratio as a function of NH_3 interruption time are shown in Fig. 3.

Our observations of a morphology change and enhanced lateral overgrowth rate of GaN stripes can be explained by the self-limiting mechanism induced by the NH_3 interruption and the enhanced Ga diffusion on the (0001) plane. The concept of self-limiting growth has been widely used in the atomic layer epitaxy of III-V compound semiconductors and flow rate modulation epitaxy of GaAs to obtain good thickness control and high quality photonic devices.^{15,16} In our case, the Ga sites of the GaN (0001) plane will be saturated by the continuous injection of the TMGa during the appropriate NH_3 interruption period (10–15 s). The growth of the (0001) plane of GaN stripes is thus inhibited and, at the same time, Ga diffusion on the (0001) plane will occur more easily, since the reduced N supply can greatly enhance the diffusion of Ga adatoms.¹⁷ As a result, the vertical growth rate of the (0001) plane is suppressed and a smooth surface is obtained. The lateral supply of TMGa (both vapor-phase diffusion and surface diffusion) to the sidewall will provide additional sources of Ga in the next GaN growth period when both TMGa and NH_3 are injected into the reactor, resulting in a higher lateral overgrowth rate. In fact, the Ga diffusion on top of the stripe could also play an important role in enhancing the lateral growth rate, which will be discussed next.

In order to study the effects of fill factor on the stripe morphology, we made two different stripe patterns, $5/15$ and $5/40 \mu\text{m}$ (opening/spacing), on one GaN wafer covered with

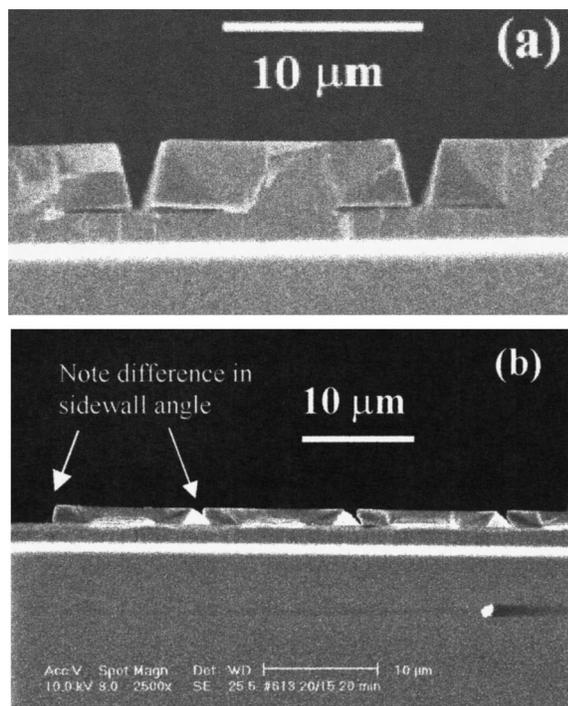


FIG. 4. Cross-sectional SEM images of (a) sample E, 1 h growth with 10 s periodic NH_3 interruption and (b) sample F, 20 min growth with 15 s periodic NH_3 interruption

SiN_x . One piece of sample E from this wafer was grown for 1 h with a 10 s periodic NH_3 interruption (the same conditions as those for sample C except the growth time). The remaining space between the closely spaced LEO stripes is around $1 \mu\text{m}$ as shown in Fig. 4(a). In this case, the sidewall is not completely vertical but shows a retrograde (inward) incline. Since the close spacing between the stripes limits the lateral supply of reactants, it will take a long time (more than several hours) to obtain fully coalesced LEO stripes for sample E unless the growth conditions are changed.¹⁸ Figure 4(b) shows sample F from the same wafer grown for 20 min with 15 s periodic NH_3 interruption (same growth conditions as those for sample D). The stripes of sample F have $40 \mu\text{m}$ spacing on the left side of the marker and $15 \mu\text{m}$ spacing on the right side of the marker. Compared with sample E, the lateral growth rate of sample F is much higher and the sidewalls of sample F incline outward. This observation supports the argument that enhanced Ga diffusion on the (0001) plane enhances the lateral growth rate of LEO stripes even with a limited or no lateral supply of reactants from the mask surface. On the left side of the marker in Fig. 4(b), however, the lateral supply of growth nutrients is not limited since the spacing between stripes is $40 \mu\text{m}$, and the stripe has a vertical sidewall. Therefore, the interplay of Ga surface diffusion on the (0001) GaN surface and the lateral supply of the reactants during the LEO growth is important to determine the morphology of LEO stripes. Continued growth of sample F leads to the continuous $10 \mu\text{m}$ thick GaN layer by lateral overgrowth within 1 h, as shown in Fig. 5. Further studies of

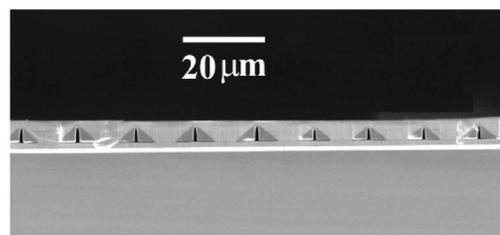


FIG. 5. Fully coalesced LEO GaN film in 1 h. The mask opening is $5 \mu\text{m}$ and the spacing is $15 \mu\text{m}$.

the influence of growth temperature, pressure, and V/III ratio on the morphology of LEO stripes and their optical and structural properties are ongoing.

In summary, we demonstrated the effects of NH_3 flow modulation on the morphology of LEO GaN stripes. The self-limiting growth mechanism and enhanced Ga diffusion on the (0001) plane as well as the lateral supply of Ga from the mask region are used to explain our observations. A L/V ratio of 2.1 and fully coalesced LEO GaN stripes within 1 h growth have been achieved.

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