Direct lateral epitaxy overgrowth of GaN on sapphire substrates based on a sparse GaN nucleation technique

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(Received 2 February 2000; accepted for publication 1 August 2000)

A sparse nucleation process on sapphire (0001) substrates has been developed for the growth of GaN thin films. The density of nucleation sites is reduced to only $4\times10^4\,\mathrm{cm}^{-2}$. Based on this process, we performed direct lateral epitaxial overgrowth (LEO) of GaN by metalorganic chemical vapor deposition on patterned SiO_2/sapphire (0001) substrates. An aggregate lateral to vertical growth rate ratio of around 2:1 was achieved after the coalescence of the GaN stripes. Cathodoluminescence imaging shows strong and uniform near-band gap luminescence from LEO regions and confirms the improved quality of LEO GaN, which is further supported by atomic force microscopy analysis. © *2000 American Institute of Physics.* [S0003-6951(00)02039-8]

Epitaxially grown III–nitride materials are known to exhibit a high density of dislocations ($10^8-10^{10}\,\mathrm{cm}^{-2}$), owing to the current need to grow on lattice-mismatched substrates.[1] Ordinarily, a low-temperature buffer is deposited prior to the growth of epitaxial GaN at a high temperature. Most of the defects and dislocations in the as-grown GaN film stem from the propagation of defects initially located at the interface of the low-temperature buffer.[1,2] One way to reduce the defect density of the GaN thin film is by the lateral epitaxial overgrowth (LEO) procedure. LEO has been applied to demonstrate marked reductions in extended defect density ($\sim10^4\,\mathrm{cm}^{-2}$) in regions where the lateral epitaxy occurs over a dielectric mask[3–6] and has led to improved performance of optoelectronic devices.[7,8] Another approach to defect reduction is to reduce the density of GaN domains obtained from the low temperature buffer, thereby reducing the defects originating from boundaries of intersecting domains.

In this work, we show that with a special substrate treatment, appropriate low-temperature buffer (LTB) growth conditions, and particular rates of temperature ramping between the LTB growth and the high temperature epitaxial growth, a GaN nuclei density as low as $4\times10^4\,\mathrm{cm}^{-2}$ can be obtained on sapphire substrates. The average distance between the nuclei is ~40 μm. At a GaN growth temperature of 1070 °C, these nuclei have a very high lateral growth rate and coalesce rather quickly. We also successfully performed the sparse nucleation on a sapphire substrate patterned with a SiO_2 mask, thereby promoting all nucleation and lateral growth in the exposed sapphire regions. After LEO growth at ~1070 °C for about 1 h, we have obtained high quality GaN stripes which have coalesced laterally over the masked regions. We have probed the optical and structural properties of the films with cathodoluminescence (CL) imaging and atomic force microscopy (AFM). Again, we note that this direct LEO (D-LEO) approach exhibits a very high lateral-to-vertical growth rate and could impact the established growth procedures for the III–nitride materials, owing to its simplicity.[9]

The sparse nucleation and D-LEO of GaN were performed in a commercial vertical geometry, atmospheric pressure metalorganic chemical vapor deposition reactor with a close-spaced showerhead geometry.[10] Trimethylgallium (TMGa) and NH_3 were used as precursors with H_2 as carrier gas. A 170 nm thick SiO_2 mask was deposited on the sapphire (0001) substrate by *e*-beam evaporation. Conventional photolithography and wet etching were used to form the patterned stripes with 5 μm opening and 30 μm spacing, oriented along the sapphire $\langle 11\bar{2}0\rangle$ direction to ensure the high lateral growth rate.

The sapphire substrates and patterned sapphire substrates with SiO_2 masks were dipped into a buffered oxide etchant (1:10) to remove any possible oxide on the substrate surface before being loaded into the reactor. High-temperature annealing under H_2 was then performed for 20 min, followed by nitridation with NH_3 at a flow rate of 3 slm at 1050 °C for 3 min. The temperature was then lowered to 480 °C, to grow a 5 nm thick GaN buffer using TMGa with a flow of 65 μmol/min and NH_3 with a flow of 1.5 slm. After the temperature was ramped to 1070 °C over a period of 15 min, the LEO was performed with TMGa and NH_3 flow of 65 μmol/min and 5 slm, respectively. The formation of a thin GaN LTB and the proper ramping rate to the LEO growth temperature are important factors that ensure the sparse nucleation and lateral growth of GaN. As the temperature is raised toward the LEO growth conditions, the thin LTB initially formed on the mask desorbs. The LTB on the openings desorbs and redistributes to form the sparse islands. After growth, the samples were cleaved and characterized with Philips XL-30 scanning electron microscopy (SEM). The CL image was taken with a modified JEOL-840A SEM using a 10 keV *e* beam and the temperature was 93 K. The surface topography was measured in tapping mode using a Digital Instruments NanoScope3 scanning probe microscope.

Uncoalesced hexagonal GaN platelets on sapphire substrates were obtained by using the sparse nucleation process

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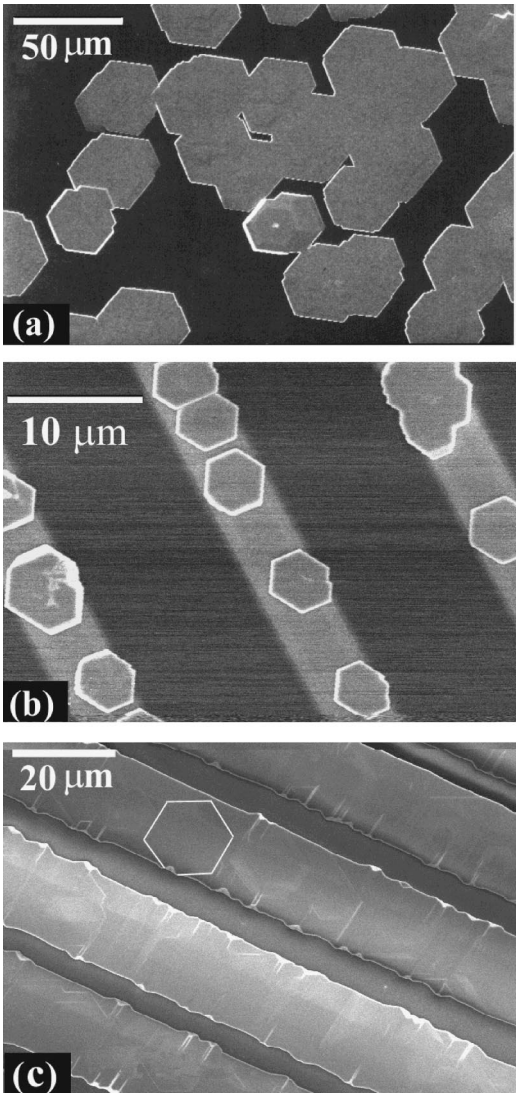

FIG. 1. SEM images of: (a) GaN on sapphire substrate after sparse nucleation and high temperature growth, (b) GaN platelets on patterned sapphire substrate, and (c) GaN stripes directly grown on patterned sapphire substrate with sparse nucleation inside the mask opening region.

followed by a high temperature growth for 20 min, as shown by the SEM image in Fig. 1(a). The size of each platelet is around 40 μm and the thickness is $\sim$2 μm. The average distance between them is also around 40 μm. The density of the platelets is estimated to be $\sim 4\times 10^4\ \mathrm{cm}^{-2}$. The in-plane crystal orientations of the platelets appear to be well aligned. In fact, using x-ray diffraction, these GaN platelets are determined to be aligned with sapphire substrate as $[1\underline{1}00]_{\mathrm{GaN}} \| [11\underline{2}0]_{\mathrm{Sap}}$. The sidewalls of these platelets are vertical $\{10\underline{1}0\}$ planes, as determined by cross-section SEM. These growth conditions appear to favor sparse nucleation, improved grain alignment, and high lateral growth rate at the growth temperatures employed. Our results also agree with the studies of Wickenden *et al.*, who showed that LTB grain alignment is improved by the high temperature nitridation.[11] Sparse nucleation was obtained by growing a 5 nm thick LTB with a low V/III ratio ($\sim$1030) at atmospheric pressure and slowly ramping up to the LEO temperature. We note that a 20–30 nm LTB is usually needed for GaN thin film growth to have high density of nucleation.[10–12] Lateral growth rate was enhanced by higher V/III ratio ($\sim$3400) and relatively

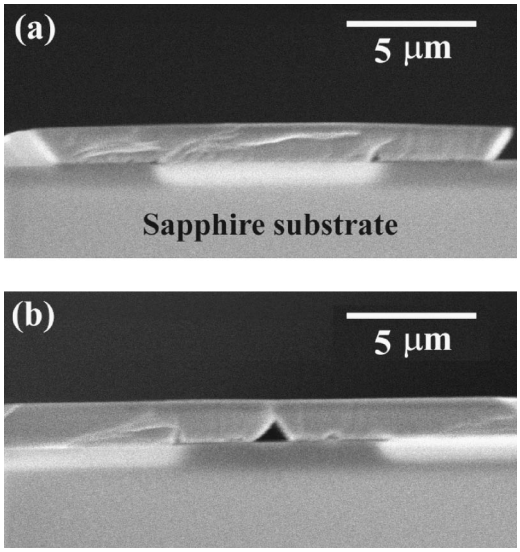

FIG. 2. Cross-sectional SEM images of: (a) uncoalesced LEO GaN stripe and (b) two coalesced LEO GaN stripes on sapphire substrate.

high growth temperature compared to our usual GaN growth temperature. As a result, we have very low nucleation density at the LEO growth temperature ($\sim$1070 °C). These nucleation sites, in turn, have a very high lateral growth rate under our growth conditions and the as-grown GaN platelets are well aligned with each other.

Although we achieved sparse nucleation and high lateral growth at these sites, their random positions on the substrate, as shown in Fig. 1(a), can cause some difficulty in obtaining high quality, smooth, coalesced thin films. The overlapping hexagonal GaN platelets will not only give rise to protrusions in the coalesced GaN thin films, but will also produce threading dislocations.[13] The aim of our growth approach is to deposit a mask material on the sapphire substrates and create stripe openings so as to permit nucleation sites within the mask opening. Our results shown in Fig. 1(b) demonstrate that sparse GaN nucleation can be performed on the patterned sapphire substrate. After about 5 min LEO growth, the nucleation sites form platelets with average size of $\sim$5 μm. These platelets are aligned along the mask opening stripes. GaN stripes with a smooth (0001) surface form from these platelets after 20 min LEO growth as shown in Fig. 1(c). The hexagonal box highlights one of the GaN platelets that coalesced with other GaN platelets and formed the coalesced stripes. The intensity of the SEM image is not uniform because of the surface charging during the *e*-beam scan, as SiO_2 and sapphire are insulators. Nucleation and growth on the SiO_2 mask are not observed.

Cross-sectional SEM images of uncoalesced and coalesced LEO GaN stripes are shown in the Figs. 2(a) and 2(b), respectively. Careful cross-section SEM studies show that sidewalls of the GaN stripe are not smooth on the submicron scale. The sidewalls of GaN stripes could be $\{11\underline{2}L\}$ (L is either $\underline{1}$ or $\underline{2}$). Since $\{11\underline{2}L\}$ are not stable planes on the submicron scale, they are actually facetted by $\{1\underline{1}01\}$ planes.[14] As a result, we expect that the uncoalesced stripes are bound by $\{1\underline{1}01\}$ sidewalls and (0001) top surfaces. This is consistent with the images in Fig. 2(a). The lateral to vertical growth rate is more than 2. We attribute the unique growth of these stripes to the V/III ratio, growth rate, and/or

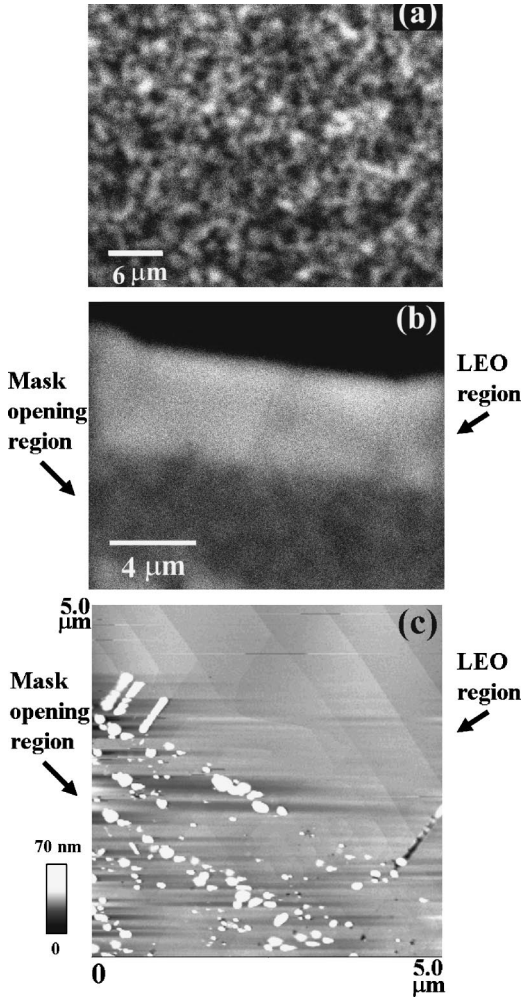

FIG. 3. (a) Monochromatic CL image ($\lambda = 360$ nm) for a conventional GaN buffer grown with a high density of nucleation sites; (b) CL image ($\lambda = 360$ nm) for a LEO GaN stripe directly grown on patterned sapphire substrate using a sparse nucleation process; (c) AFM image of D-LEO GaN stripe showing both the mask opening region and lateral overgrown region.

the surface properties of SiO_2 mask materials used in our experiments. Continued LEO growth results in the coalescence of adjacent GaN stripes, as observed in Fig. 2(b). Small void areas at the coalesced boundaries are observed.

In order to evaluate the optical properties of the LEO growth, CL images of the LEO stripe were acquired for the near-band gap emission with $\lambda = 360$ nm. Figure 3(a) shows a CL image of the conventional GaN layer grown using a thick LTB. The large fluctuation of the luminescence intensity indicates a high density of nucleation sites and also a high density of defects from grain boundaries. In Fig. 3(b), the strong and uniform band gap emission from the lateral overgrown region of GaN indicates the markedly reduced defect density of LEO GaN in comparison to the conventional GaN film. Figure 3(b) also shows that the region in the mask opening is not uniform and dot-like features can be observed. Tapping mode AFM was performed over a 5×5 μm area covering both mask opening and lateral overgrown regions to study the topography of the LEO stripes, as shown in Fig. 3(c). The stripe has a smooth surface on an atomic scale in the lateral overgrown region with a root mean square (rms) roughness of 3 Å, indicating the absence of threading dislocations (TDs).[15,16] However, some dots and bumps of different sizes in the mask opening regions are clearly identified as well as step terminations related to the TDs[15,16] with a rms roughness of 29 Å. Generally, the surface topography of our sample is different from LEO of GaN on patterned GaN/sapphire substrate and AlGaN/Si substrate.[16] The straight and wider growth steps observed in Fig. 3(c) can be attributed to the high lateral growth rate of GaN stripes and the sparse nucleation process employed. The origins of the pits and bumps in the mask opening regions are not clear at the present time, but they can be eliminated by decreasing the growth temperature and the V/III ratio.[13]

In summary, we demonstrate sparse nucleation of GaN on sapphire substrates and subsequent thin film growth using a very thin LTB and optimized growth conditions. We successfully controlled the nucleation site density and location by performing the sparse nucleation on patterned SiO_2/sapphire substrate. After high temperature growth, atomically flat LEO GaN stripes were obtained with unique stripe morphology and therefore D-LEO on sapphire substrates was realized. CL imaging and AFM analysis confirmed the reduced defect density and improved material quality.

This work was supported by ONR and by DARPA through the UCSB GaN consortium.

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