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In situ pendeoepitaxy of GaN using heteroepitaxial AlGaN/GaN cracks

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Pendeoepitaxy on patterned templates has been proven to be efficient for reducing threading dislocation densities in GaN thin films. In this letter, we report on in situ crack-assisted pendeoepitaxy of GaN using spontaneously formed cracks in AlGaN/GaN heterostructures. Our approach involves the growth of an AlGaN/GaN template followed by in situ thermal etching and deposition of an amorphous silicon nitride mask in a low pressure metal organic chemical vapor deposition system. Microwirelike GaN seeds are then formed along the crack lines during the initial stage of GaN overgrowth, which act as nucleation stripes for epitaxial lateral overgrowth. Transmission electron microscopy revealed that the lateral overgrowth of the wirelike GaN seeds effectively bends threading dislocations toward (1100) directions on the amorphous silicon nitride mask. The threading dislocation density by this method has been reduced from $2 \times 10^8 \text{cm}^{-2}$ in control samples to $2 \times 10^6 \text{cm}^{-2}$ in some parts and $5 \times 10^7 \text{cm}^{-2}$ in other parts of the GaN layer as determined by plan-view transmission electron microscopy which is very encouraging. © 2006 American Institute of Physics. [DOI: 10.1063/1.2219093]

Advances in GaN-based semiconductor thin film technology have paved the way for the production of high-power ultraviolet to visible light-emitting diodes, laser diodes, ultraviolet detectors, and field-effect transistors. However, the lack of native GaN substrates in large quantity and of sufficient size remains a serious problem. Consequently, current GaN-based optoelectronic devices are fabricated by heteroepitaxy of III-nitride thin films on foreign substrates such as sapphire, SiC, and Si. The heteroepitaxy of III-nitrides intrinsically suffers from high dislocation density due to large lattice mismatch, on the order of low to mid $10^9 \text{cm}^{-2}$ in vapor phase epitaxial GaN and up to $10^{10} \text{cm}^{-2}$ in molecular beam epitaxial GaN. A great deal of effort directed towards reduction of the threading dislocation (TD) density has been reported. Currently, the most commonly used method to reduce TD density significantly is the epitaxial lateral overgrowth (ELO) technique or a further improvement (so-called pendeoepitaxy). However, these methods require additional ex situ processing, resulting in high production costs. Another issue germane to nitride heterolayers is that the epitaxy of (0001) AlGaN on GaN templates generates cracks that relax the lattice mismatch-induced tensile strain when the thickness of the AlGaN is above the critical thickness because the introduction of misfit dislocations is very difficult due to the lack of available slip systems (as in the case of GaN on Si and SiC substrates). In the present work, the cracks in AlGaN/GaN heterostructures have been utilized as self-formed mesa structures by in situ thermal etching and deposition of an amorphous silicon nitride mask. In this letter, we report the in situ crack-assisted pendeoepitaxy of GaN using spontaneously formed cracks in AlGaN/GaN heterostructures.

A 0.3 µm thick $\text{Al}_{0.34}\text{Ga}_{0.66}\text{N}$ epilayer was deposited on a 2 µm thick GaN template. The formation of cracks in the AlGaN epilayers was confirmed by scanning electron microscopy (SEM). The AlGaN/GaN heterostructure was then reloaded into the growth system and thermally annealed at 1020 ºC and 200 Torr for 5 min, after which an amorphous silicon nitride thin layer (referred to as SiN$_x$ because of its unknown stoichiometry) was deposited on the heterostructure.

Figure 1 shows a schematic of the crack-assisted pendeoepitaxy (CAPE) using in situ deposited amorphous SiN$_x$. Wurtzite AlGaN heteroepitaxy along the (0001) direction on GaN would be expected to generate cracks during growth to relax the lattice mismatch induced tensile stress when the AlGaN layer thickness is thicker than the critical thickness because the introduction of misfit dislocations is hampered by the lack of available slip systems. The cracks lie mainly along the energetically favored (1120) crystallographic directions with (1100) cleavage planes. Cracks com-

\[\text{FIG. 1. A schematic of the crack-assisted pendeoepitaxy (CAPE) using in situ deposited amorphous SiN}_x\]
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Figure 1. We deposited an initial GaN seed layer using in situ deposited amorphous SiN$_x$ on an AlGaN/GaN heterostructure with a network of film cracks. Figure 2(a) shows a SEM micrograph of the cracks in an as-grown AlGaN/GaN heterostructure. The AlGaN/GaN heterostructure with a network of cracks was reloaded into the growth chamber and a nominally 0.3 \( \mu \)m thick initial GaN seed layer was deposited for a period of 9 min after the deposition of an amorphous SiN$_x$ mask layer. Figures 2(b)–2(d) show SEM micrographs of the initial GaN seed layer, clearly showing the formation of microwirelike GaN seeds along the crack lines in the AlGaN/GaN heterostructure. Figure 2(d) is a magnified SEM micrograph of area A in Fig. 2(c) which reveals a segment of a crack line between the GaN seeds. It should be noted that truncated hexagonal GaN islands are formed on the amorphous SiN$_x$ mask layer when the crack-free area between the crack lines is relatively wide [Fig. 2(c)].

Figure 3(a) shows a low magnification SEM micrograph of a nominally 0.5 \( \mu \)m thick initial GaN seed layer deposited for 15 min on an AlGaN/GaN heterostructure with a network of cracks. The deposition time was increased from 9 to 15 min to obtain a full network of microwirelike GaN seed regions along the crack network. Figure 3(a) clearly shows that the overall network of microwirelike GaN seed regions is affected by the surface geometry of the cracks on the AlGaN/GaN heterostructure. Figure 3(b) shows a cross-sectional SEM micrograph of the microwirelike GaN seed network, revealing the formation of a buried void that extends into the GaN template. The trapezoidal cross section of a microwirelike GaN seed also shows that the height of the trapezoid is much higher than the nominal thickness of 0.5 \( \mu \)m. It should be noted that the nominal thickness was determined by the growth rate of the GaN film. This indicates that some Ga adatoms on the amorphous SiN$_x$ effectively diffuse to the microwirelike GaN seed regions and incorporate.

The next step in the CAPE method is to produce a coalescent flat GaN overlayer on the microwirelike GaN network by enhancing the lateral growth of the microwirelike GaN seed regions and the hexagonal GaN islands. The sidewalls of the microwirelike GaN seeds and the hexagonal GaN islands would mainly be composed of \{0001\} pyramidal or \{1100\} prismatic planes. Thus, in order to enhance the lateral growth rate of the \{1100\} facets, the following growth conditions would be desirable: a Ga-rich ambient by ammonia flow modulation\cite{10} or a lower ammonia partial pressure\cite{11} with Mg doping\cite{12} at a relatively high temperature and low pressure.\cite{13}

In order to investigate the morphological evolution of the overgrown GaN layer on the network of microwirelike GaN seeds, we grew a 4 \( \mu \)m thick GaN on the microwirelike GaN network [Fig. 4(a)] at 1040 °C and 200 Torr while the seed network was grown at 1020 °C and 300 Torr. Figure 4(a) shows a SEM micrograph of the overgrown GaN, indicating that the sidewalls of individual cells with triangular, trapezoidal, or parallelogram shapes in the microwirelike GaN seed network grew in the lateral directions and finally converge at the centers of
the cells in the network. When a 12 µm thick GaN regrowth was performed on the wirelike GaN network at 76 Torr, the surface coalesced as shown in Fig. 4(b) with an AFM surface rms roughness of 0.28 nm.

Plan-view TEM analysis was performed to investigate the threading dislocation distribution of the 12 µm thick overgrown GaN. The analysis revealed two different regions with different threading dislocation densities. One region showed a dislocation density of about $5 \times 10^7$ cm$^{-2}$ while another region showed a density of $\sim 2 \times 10^8$ cm$^{-2}$. This compares with a density of low $10^6$ cm$^{-2}$ characteristically with that of GaN overlayers regrown on GaN templates with defective regions. It should be mentioned that either case is the threading dislocation distribution of the 12 surface coalesced as shown in Fig. 4 was performed on the wirelike GaN network at 76 Torr, the another region showed a density of $10^7$ cm$^{-2}$ is comparatively lower.

Cross-sectional TEM analysis was performed to investigate the dislocation behavior near the crack lines in the overgrown GaN. Figure 5 shows a TEM micrograph taken under two-beam diffraction conditions with $g=\bar{1}20$, showing some edge ($b=\frac{1}{3}(120)$) and mixed-type dislocations. Figure 5 clearly shows that dislocations which penetrate through the amorphous SiN, are bent from the crack side toward the (100) direction, as indicated by the arrow, implying that the lateral growth of the sidewall of wirelike GaN seed effectively caused dislocation bending. The micrograph also shows that some dislocations are terminated at the amorphous SiN, layer deposited on the AlGaN layer.

Basically, threading dislocations would be expected to penetrate from the GaN template into the overgrown GaN layer through open pores in the few monolayers-thick amorphous SiN, during the overgrowth process. The penetration of dislocations would be suppressed by increasing the thickness of the in situ deposited SiN, mask and by enhancing the lateral growth rate of the sidewalls of GaN seeds at an early stage of the overgrowth. However, a reduction in pore density in the porous SiN would decrease the density of hexagonal GaN islands which assist in coalescence where the line density of the cracks is relatively low. If the crack density were uniform and controllable, the reduction efficiency of TD density would be maximized by minimizing the formation of hexagonal GaN islands in the open pores. Cracking of heteroepitaxial AlGaN would be dependent on the composition and thickness of the AlGaN as well as the thickness and quality of the GaN template. It should be mentioned that a further significant reduction in threading dislocation density is possible if the growth parameters for the CAPE method are optimized. Moreover, the CAPE method would also be applicable to the growth of thick (0001) GaN films on SiC and Si substrates where thermal mismatch leads to tensile strain and severe cracking.

In conclusion, we have demonstrated in situ CAPE of GaN using self-formed cracks in AlGaN/GaN heterostructures. We believe that the CAPE method provides an additional avenue for developing high quality III-nitride heteroepitaxial films with a low threading dislocation density at an affordable cost, compared to the conventional ex situ pendeoepitaxy and epitaxial lateral overgrowth techniques.

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