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Moon, Y.T., Liu, C., Xie, J., et al. In situ pendeoepitaxy of GaN using heteroepitaxial AlGaN/GaN cracks. Applied Physics Letters, 89, 024103 (2006). Copyright © 2006 AIP Publishing LLC.

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### In situ pendeoepitaxy of GaN using heteroepitaxial AlGaN/GaN cracks

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(Received 11 April 2006; accepted 31 May 2006; published online 11 July 2006)

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  $\langle 1\bar{1}00 \rangle$  directions on the amorphous silicon nitride mask. The threading dislocation density by this method has been reduced from  $2 \times 10^9$  cm<sup>-2</sup> in control samples to  $2 \times 10^8$  cm<sup>-2</sup> in some parts and  $5 \times 10^7$  cm<sup>-2</sup> 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.<sup>1</sup> 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  $10^9$  cm<sup>-2</sup> in vapor phase epitaxial  $GaN^2$  and up to  $10^{10} cm^{-2}$  in molecular beam epitaxial GaN. A great deal of effort directed towards reduction of the threading dislocation (TD) density has been reported.<sup>2-6</sup> Currently, the most commonly used method to reduce TD density significantly is the epitaxial lateral overgrowth (ELO) technique<sup>3</sup> or a further improvement (so-called pendeoepitaxy).<sup>5</sup> 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).<sup>7,8</sup> 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  $\mu$ m thick Al<sub>0.34</sub>Ga<sub>0.66</sub>N epilayer was deposited on a 2  $\mu$ m 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 °C and 200 Torr for 5 min, after which an amorphous silicon nitride thin layer (referred to as SiN<sub>x</sub> 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<sub>x</sub>. 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  $\langle 11\overline{20} \rangle$  crystallographic directions with  $\{1\overline{100}\}$  cleavage planes. Cracks com-



FIG. 1. A schematic of the crack-assisted pendeoepitaxy (CAPE) using *in* situ deposited amorphous  $SiN_x$ .

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FIG. 2. SEM micrographs of (a) the cracks in an as-grown AlGaN/GaN heterostructure, (b) microwirelike GaN seeds formed along the crack lines, (c) a network of microwirelike GaN seeds, and (d) a magnification of area A marked in Fig. 2(c).

mence from the surface of the AlGaN and proceed toward the AlGaN/GaN interface. The plastic relaxation is accompanied by the introduction of misfit dislocations from the crack edges in the AlGaN/GaN interface.<sup>9</sup> These, in turn, enlarge the width of the AlGaN crack trench and expose the GaN along the cracks. Full relaxation of lattice mismatch induced stress in the heterostructure includes the propagation of cracks into the GaN template along the exposed interface when the template is sufficiently thick.' Cracking would finally lead to production of a network of V-shaped crack trenches in the AlGaN/GaN heterostructure. The second step in CAPE is the *in situ* thermal etching of the crack trenches by interrupting the source flows of Ga and Al for a short time. AlGaN is thermally more stable than GaN because the atomic bonding energy of Al–N (11.52 eV) is higher than that of Ga-N (8.92 eV). The *in situ* thermal annealing under hydrogen and ammonia ambients would preferentially etch GaN at the edges of the V trenches, producing deep distorted U-shaped crack trenches as shown in Fig. 1. The third step is deposition of a few monolayers-thick amorphous SiN<sub>x</sub> layer on the cracked heterostructure by flowing silane as the silicon source under hydrogen and ammonia ambients. The amorphous SiN<sub>y</sub> would deposit on the top surface of the AlGaN as well as on the bottom of the U trenches [Fig. 1]. Any deposition of amorphous  $SiN_r$  on the sidewalls of the U trenches would expectedly be impeded by the trench geometry. Thus, in the fourth step of CAPE, the overgrowth of GaN would be initiated from the sidewalls of the U trenches. The initial GaN wings from both sidewalls of the trench would then grow laterally along the  $\langle 1100 \rangle$  directions as well as vertically along the [0001] direction and then meet, resulting in the formation of a void due to the limitation of mass transport into the trench. After coalescence, microwirelike GaN seeds would grow vertically and laterally, as shown in Fig. 1.

Based on the concept of the CAPE method proposed in Fig. 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



FIG. 3. SEM micrographs of (a) the overall network of microwirelike GaN seeds with a nominal thickness of 0.5  $\mu$ m and (b) the cross section of the wirelike GaN network.

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<sub>x</sub> 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<sub>x</sub> 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 wirelike 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 crosssectional 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<sub>x</sub> 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 wirelike GaN network by enhancing the lateral growth of the wirelike GaN seed regions and the hexagonal GaN islands. The sidewalls of the wirelike GaN seeds and the hexagonal GaN islands would mainly be composed of {1101} pyramidal or {1100} prismatic planes. Thus, in order to enhance the lateral growth rate of the  $\{1\overline{1}00\}$  facets, the following growth conditions would be desirable: a Ga-rich ambient by ammonia flow modulation<sup>10</sup> or a lower ammonia partial pressure<sup>11</sup> with Mg doping<sup>12</sup> at a relatively high temperature and low pressure.<sup>6</sup> In order to investigate the morphological evolution of the overgrown GaN layer on the network of wirelike GaN seeds, we grew a 4  $\mu$ m thick GaN on the wirelike 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 wirelike GaN seed network grow in the lateral directions and finally converge at the centers of



FIG. 4. SEM micrographs of (a) 4  $\mu$ m thick, and (b) 12  $\mu$ m thick regrown GaN overlayers on the wirelike GaN seed networks at 200 and 76 Torr, respectively.

the cells in the network. When a 12  $\mu$ m 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  $\mu$ m 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<sup>-2</sup> while another region showed a density of  $\sim 2 \times 10^8 \text{ cm}^{-2}$ . This compares with a density of low 10<sup>9</sup> cm<sup>-2</sup> characteristically obtained in control samples (not shown here). This anisotropy in threading dislocations can be explained as follows. The distribution of wirelike GaN seeds is not uniform as can be seen in Fig. 3(a). The GaN overgrown on the regions where the seed networks are dense would coalesce mainly by the lateral overgrowth of the sidewalls of wirelike GaN seeds. However, the GaN overgrown on the regions where the networks are sparse would coalesce mainly by the GaN overgrowth process on hexagonal GaN islands formed in the open pores in amorphous  $SiN_x$ , resulting in the relatively defective regions. It should be mentioned that either case is better than uniform growth as was done for the control sample. The dislocation density of low 10<sup>8</sup> cm<sup>-2</sup> is consistent with that of GaN overlayer regrown on GaN template without AlGaN using in situ deposited porous SiN<sub>x</sub>,<sup>13</sup> while the mid 10<sup>7</sup> cm<sup>-2</sup> 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  $\mathbf{g}=(11\overline{2}0)$ , showing some edge ( $\mathbf{b}=1/3[11\overline{2}0]$ ) and mixed-type dislocations. Figure 5 clearly shows that dislocations which penetrate through the amorphous SiN<sub>x</sub> are bent from the crack side toward the  $\langle 1\overline{1}00 \rangle$  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  $SiN_x$  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  $SiN_x$  during the overgrowth process. The penetration of dislocations would be suppressed by increasing the thickness of the *in situ* deposited SiN<sub>x</sub> 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<sub>x</sub> 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.

This work was supported by grants from ONR (Dr. C. E. C. Wood) and AFOSR (Dr. G. L. Witt and Dr. K. Reinhart). The authors acknowledge collaboration and discussions with Professor R. M. Feenstra and Professor T. S. Kuan and Dr. A. Sagar during the early stages of *in situ* deposited silicon nitride development.

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