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The origin and mitigation of volcano-like morphologies in micron-thick AlGaN/AIN heteroepitaxy

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We investigated the origin of morphological instability in 2 μm thick Al0.6Ga0.4N/AIN heteroepitaxy. The primary morphology was driven by the residual epitaxial strain, forming hill-like morphologies via surface diffusion. The secondary morphology was driven by the interaction between the primary morphology and dislocation clusters in the epitaxial layers. The difference in the local growth rate yields volcano-like morphologies centering on deep pits. Insertion of multi-stack superlattice transition layers between AlGaN and GaN effectively suppressed the secondary morphologies by simultaneously pre-relaxing the template and filtering threading dislocations. Published by AIP Publishing.

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AlGaN-based UVC (λ = 200 nm–280 nm) light-emitting diodes have attracted much attention for their application in water purification, sterilization, and other bio-medical applications. To enhance the LED performance by improving the epitaxial quality, plenty of efforts have been made to reduce the threading dislocation density (TDD) of the AlN template, cations. To enhance the LED performance by improving the water purification, sterilization, and other bio-medical applications, diodes have attracted much attention for their application in crystals as substrates.1–4 Although the quality of subsequent annealing of buffer layers, and fabrication of bulk AlN single crystals as substrates.1–4 The InGaN and AlGaN alloys,18–20 strain-induced morphological instabilities were reported to be local surface suppression (or “V-pits”) and gliding of prismatic dislocations for non-c-plane epitaxy.12–15 For material systems with very low dislocation densities, for example, SiGe/Si and InGaAs/GaAs, the total free energy could also be lowered by propagation of surface undulation, which is also known as Asaro-Tiller-Grinfield (ATG) instability.16–17 Although the strain-driven composition-pulling effects have been studied in both InGaN and AlGaN alloys,16–20 strain-induced morphological instability in ternary nitrides has not been reported. In this publication, we revealed that the AlGaN morphology was also sensitive to its strain states. The interaction between the strain-induced morphology and its vicinal defect structures was also discussed.

Because of the significant difference between Al and Ga surface properties, it is rather difficult to decouple the parallel strain effects and surface kinetics effects among AlGaN epitaxial layers with different alloy compositions. Therefore, instead of preparing AlGaN epitaxial layers with different Al compositions, we investigate the strain effects on the AlGaN heteroepitaxy with the same composition but different underlying templates. Three samples consisting of 2 μm Al0.6Ga0.4N and 2.5 μm AlN were prepared by Taiyo-Nippon-Sanso SR-2000 metalorganic chemical vapor deposition (MOCVD). The growth conditions of Al0.6Ga0.4N among three samples were identical. The growth temperature and pressure of the top AlGaN were 1090 °C and 20 kPa, respectively, with a growth rate of 0.2 nm/s. The only difference among samples was the transition layer (TL) structure between AlN and Al0.6Ga0.4N,

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as depicted in Fig. 1. Sample A had no transition layer between Al$_{0.6}$Ga$_{0.4}$N and AlN; sample B possessed a 100 nm Al$_{0.85}$Ga$_{0.15}$N layer on top of AlN followed by a 400 nm linearly compositional-graded transition layer (CG-TL) with 85% Al at the bottom and 55% Al at the top of it; sample C also possessed a 100 nm Al$_{0.85}$Ga$_{0.15}$N layer on the top of AlN, but the subsequent transition layer had a more detailed structure. The transition layer on sample C consisted of four stacks of Al$_{0.85}$Ga$_{0.15}$N/Al$_{0.55}$Ga$_{0.45}$N superlattices with the same total thickness but varying thickness ratios. The superlattice near the bottom was dominated by Al$_{0.85}$Ga$_{0.15}$N, while the top one was majorly Al$_{0.55}$Ga$_{0.45}$N. In other words, the multi-stack superlattice transition layer (MS-SL TL) has a gradual variation in the long-range average composition but a drastic compositional variation in a short range. The schematic epitaxial structures are illustrated in Fig. 1.

Three samples were characterized by NT-MDT atomic force microscopy (AFM) and a PANalytical high-resolution X-ray diffractometer (HR-XRD) to investigate the interplay between the strain states and surface morphologies. The defect structures and dislocation dynamics were characterized with a JEOL JSM-7000F scanning electron microscope (SEM) equipped with cathodoluminescence (CL) modules and an FEI TECNAI G2 F20 transmission electron microscope (TEM). TEM samples were prepared by conventional lift-out techniques with a Helios NanoLab 600i focus ion beam.

Figures 2(a)–2(c) illustrate 50 $\mu$m $\times$ 50 $\mu$m three-dimensional AFM images of sample A to sample C. All samples possessed hill-like morphologies on the surface, but the geometry of hills varied from one to one. The height of the hill morphology was larger in sample A and sample B, but its lateral dimension was larger in sample B and sample C. It is worth noticing that some hill morphologies were centered on hundreds-of-nanometer deep pits but some of them was not. Those hills with deep pits were higher than those without them. To distinguish the two types of morphologies, those with deep pits were referred to as “volcano morphology” while those without them were still referred to as “hill morphology” in the following paragraphs. Figure 2(d) plots representative AFM line scans of each sample containing hills and volcanoes. The heights of hills were all around 20 nm $\sim$ 30 nm, while those of volcanoes were $>100$ nm, implying different forming mechanisms between two morphologies.

To correlate the AlGaN morphology with its strain states, asymmetric (105) plane reciprocal space mapping (RSM) was conducted by HR-XRD and is plotted in Figs. 3(a)–3(c). The detailed model and fundamental physical parameters used can be found in the supplementary material and Refs. 21–23.

![FIG. 1. Schematic illustration of AlGaN on AlN heteroepitaxy with (a) no transition layers, (b) compositionally graded transition layer (CG-TL), and (c) multi-stack superlattice transition layer (MS-SL TL). The scanning TEM image of superlattices is shown in the right.](image1)

![FIG. 2. Three dimensional AFM image of (a) sample A with no TL, (b) sample B with CG-TL, and (c) sample C with MS-SL TL. (d) Selected line profiles of each sample denoted with hill/volcano morphologies. The baseline levels were intentionally shifted to distinguish them.](image2)
Their compositions were close, but the degree of relaxation (R) varied. In sample A, the R of AlGaN was 30.2%, whereas that of samples B and C was 40.7% and 39.3%, respectively. The transition layers in samples B and C were both partially relaxed after growth, yielding a template with a larger in-plane lattice constant for subsequent thick Al0.6Ga0.4N deposition. Since the AlGaN was coherently grown on the template in the initial stage, we define the initial strain ($e_i$) as:

$$e_i = \frac{a_{\text{template}} - a_0}{a_0},$$

where $a_0$ is the unstrained lattice constant of the AlGaN layer estimated by Vegard’s Law and $a_{\text{template}}$ is the template lattice constant extracted from the RSM plot. As the AlGaN layer grew thicker, the compressive strain was eventually relaxed. The residual strain ($e_r$) of AlGaN after growth was then estimated by:

$$e_r = \frac{a_{\exp} - a_0}{a_0},$$

where $a_{\exp}$ is the lattice constant of Al0.6Ga0.4N extracted from RSM. As summarized in Fig. 2(d), $e_i$ and $e_r$ of samples B and C were smaller than those of sample A with a pre-relaxed AlGaN template. In a bulk strained layer with an abrupt interface, misfit dislocations (MDs) were deposited at the interface by gliding the TDs. However, as the MD density grew, the resulting strain field around MD impeded further TDs gliding in the orthogonal direction. Therefore, the blockade of TD by MD was also alleviated, which also explained a more effective strain relaxation in sample B. The FWHMs of AlGaN peak in Qx and Qz are summarized in Table I. The narrower AlGaN peak in sample B than in sample A was attributed to the suppressed nucleation of additional TDs. For sample C, the fast-varying strain field in the superlattice bent the threading dislocations, resulting in a relaxed template with a reduced TDD. Although the compositionally graded layer could suppress nucleation of dislocation from the surface, it did not actively reduce the pre-existing threading dislocation density (TDD) as superlattices did. The TDD reduction by the superlattice was consistent with the further narrowed AlGaN Bragg peak width and the stronger CL intensity of sample C in the following paragraph.

Figures 4(a)–4(c) correspond to SEM and CL images of samples A to C. The sizes of volcano morphologies in samples A were smaller than those in samples B and C. The estimated deep pit densities are also summarized in Table I. The CL intensity was strong in the vicinity of volcanoes but very dim in the center. On the contrary, there is no strong CL intensity contrast around the hills. Figure 3(d) shows the point CL spectrum in the volcano vicinity (point A) and in the planar region (point B). The longer CL wavelength indicated a higher Ga

<table>
<thead>
<tr>
<th>Sample</th>
<th>FWHM in Qx ($\times 10^{-3}$ Å$^{-1}$)</th>
<th>FWHM in Qz ($\times 10^{-3}$ Å$^{-1}$)</th>
<th>Pit density ($\times 10^4$ cm$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>8.65</td>
<td>6.93</td>
<td>2.59</td>
</tr>
<tr>
<td>B</td>
<td>7.81</td>
<td>6.76</td>
<td>1.59</td>
</tr>
<tr>
<td>C</td>
<td>7.67</td>
<td>6.31</td>
<td>0.15</td>
</tr>
</tbody>
</table>
composition in the vicinity of volcanoes, which was consistent with previous AlGaN morphology studies.\textsuperscript{7–9} In the integrated CL spectrum, the multiple peaks of sample A implied a strong composition inhomogeneity. Because the excited carriers were prone to relax to the local energy minimum, the intensity was higher for the long wavelength emission. The composition homogeneity was much improved in samples B and C. The integrated intensity of sample C was 1.7 times higher than that of sample B due to the reduced defect density. The difference between Al and Ga surface kinetics might explain this composition fluctuation as the morphology evolves, but it cannot explain the origin of morphology since three samples underwent identical epitaxial process conditions. Therefore, we can conclude in advance that these morphologies were strain-driven phenomena. In continuum mechanic models of ATG instability, the dimension of morphology ($\lambda$) is inversely proportional to the square of misfit strain ($\lambda \sim e_d^{-2}$) in the long wavelength limit.\textsuperscript{17,28,29} In samples B and C, the lateral size of volcano ranged from 10 $\mu$m to 12 $\mu$m, while that in sample A was mostly 6 $\mu$m to 8 $\mu$m. This prediction is in good agreement with the measured residual strain $\varepsilon_r$ ($\varepsilon_r \sim 0.71$ for sample A and $\sim 0.57$ for samples B and C).

In order to further investigate the forming mechanism of these strain-induced morphologies, cross-sectional TEM observation was conducted on two specimens prepared from sample A. One specimen was in the deep pits’ proximity [Figs. 5(a)–5(d)], while the other one was tens of micrometers away from adjacent deep pits [Fig. 5(e)]. Sample A was observed in the dark-field (DF) and weak-beam dark-field (WBDF) with $g = 11\bar{2}0$, and $g = 0002$ for multiple $g$-$b$ analysis. In Figs. 5(c) and 5(d), misfit dislocations pointing out the sample surface populated densely near the AlGaN/AlN hetero-interface, which resulted from the gliding of treading dislocations under epitaxial strain. In multiple $g$-$b$ analysis, most dislocations were only visible under $g = 11\bar{2}0$. Therefore, most dislocations were purely edge-type. The direction of TDs was close to the growth normal in AlN layers. On the contrary, TDs were inclined under the compressive strain of AlGaN heteroepitaxy. It is worth noticing that the dislocation inclination angle was larger in the sample away from the deep pits [Fig. 5(e)].

In Fig. 5. The weak-beam-dark-field (WBDF) images under (a) $g = 11\bar{2}0$ and (b) $g = 0002$ of sample A in the deep pits’ proximity. The dark-field image near the AlGaN/AlN interface under two-beam condition under (c) $g = 11\bar{2}0$ and (d) $g = 0002$. (e) The WBDF image of sample A away from the deep pits under $g = 11\bar{2}0$. Since
the inclination was induced by the epitaxial stress applied on the dislocations, a reduced inclination angle implied a less compressive strain in the proximity of deep pits. Therefore, as a threading dislocation sweeps through the epitaxial layer, it is impeded when it approaches the deep pits’ vicinity. Eventually, dislocations will be accumulating in the deep pits’ proximity and the TDD away from the deep pits will be eventually reduced, which is consistent with our TEM and CL observations. The CL spectrum near the deep pits was not measurable due to a high density of non-radiative recombination centers.

With all the above materials, we can illustrate the origin of morphological instability of AlGaN/AlN heteroepitaxy. In the initial stage, AlGaN was coherently grown on the AlN template and the compressive strain energy accumulated with the increasing thickness. After the thickness exceeded its critical thickness, two strain-relaxation mechanisms occurred in parallel: (1) forming MDs at the AlGaN/AlN interface via gliding TDs and (2) enhancing the surface roughness by surface diffusion, which resulted in the hill morphologies. Because the strain energy near MDs was relaxed, the forces applied on the vicinal threading dislocations were also reduced. As a result, threading dislocations started to cluster, forming a local highly defective area in the vicinity of MDs. If the dislocation cluster coincided with the high chemical potential area in the morphology, it became energetically unfavorable for incoming precursors. Therefore, the growth rate in the center of the cluster became low. On the contrary, the growth rate in its proximity was rather enhanced by harvesting adatoms diffusing from the cluster. As a result, the volcano morphologies were formed eventually.

In conclusion, we revealed the origin of two prominent surface morphologies on the micron-thick AlGaN/AlN heteroepitaxy. We suggested that the hill morphology induced by strain relaxation and the volcano morphology was a secondary morphology due to the interaction between the hill morphology and underneath dislocation cluster. Inserting transition layers between AlGaN and AlN not only suppressed the primary morphology by pre-relaxing the template but also reduced the TDD with a properly designed structure. As a result, the secondary morphology could be significantly reduced. In this study, the pit density was reduced up to 95% after inserting a 500 nm multi-stack superlattice transition layer. The effective suppression of morphological instability of thick AlGaN/AlN heteroepitaxy paved the way for the fabrication of high performance AlGaN-based deep-UV emitters.

See supplementary material for the detailed procedure of how Fig. 3 was constructed. It included the fundamental parameters adopted for calculation, the physical meaning of important reference curves on the plot, how the internal parameters were extracted, and some details about how we labeled those buried peaks.

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