The role of surface kinetics on composition and quality of AlGa_N

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Abstract

Metal-polar, Al-rich AlGa_N films were grown on both single crystalline AlN and sapphire substrates. The role of surface morphology and surface kinetics on AlGa_N composition is presented. With the reduced dislocation density of the films grown on AlN substrates, atomically smooth bilayer stepped surfaces are achieved with RMS roughness of less than 50 pm for a 5x5 μm² AFM scan area. By controlling the surface supersaturation through adjusting the growth rate, a transition from 2D nucleation to step flow was observed. The critical misorientation angle for step-bunching in nominal Al_{0.70}Ga_{0.30}N grown with a growth rate of 600 nm/hr on AlN substrates was found to be 0.4°. The composition of bilayer stepped AlGa_N was strongly dependent on substrate misorientation angle, where a compositional variation by a factor of two for a change in misorientation angle from 0.05 to 0.40° was observed; this is explained by the different surface diffusion lengths of Ga and Al. Step-bunching resulted in strong compositional inhomogeneity as observed by photoluminescence and scanning transmission electron microscopy studies.

Keywords: A3. Metalorganic chemical vapor deposition; B1. Nitrides; A1. Growth models; A1. Surface kinetics

I. Introduction

Al-rich AlGa_N alloys are ideal for deep-UV optoelectronic devices due to their direct wide bandgap.^{1, 2} To operate at their full potential, these devices require epitaxial layers with low dislocation densities and smooth interfaces. To achieve low dislocation density Al-rich AlGa_N thin films, the growth on single crystalline native AlN substrates is mandatory. High quality Al-rich AlGa_N thin films have been demonstrated on these substrates.³ By realizing low dislocation density films, the number of non-radiative events and leakage paths is decreased, therefore improving the performance of the device. High quantum efficiencies can be obtained by maintaining atomically smooth interfaces in quantum well structures.⁴ Also, the importance of thickness control on an atomic level has been demonstrated in quantum cascade lasers, where the thickness non-uniformity caused by surface roughness hindered the overall performance of the devices.⁵ For these reasons, a control scheme is needed to consistently achieve atomically smooth Al-rich AlGa_N surfaces with desired composition. To properly formulate such a scheme, an understanding of the surface kinetics during growth is needed.

The compositional uniformity of ternary alloys was shown to be directly related to the surface roughness.⁶ This was due to the fact that the surface kinetics of different adatom species of the alloy intrinsically differed from each other and, in addition, it depended on local energetic barriers associated with the steps.^{7, 8} In particular, step-bunched surface morphology contained seemingly abrupt and undesirable lateral compositional gradients that were correlated with the differences in step densities across such a surface.^{6, 9, 10} The local variations in alloy composition observed on rough surfaces are not desirable since these variations can scatter carriers and broaden or add additional emissions in optoelectronic devices. Therefore, in order to achieve a

uniform composition in alloy films and ultimately improve device performance, controlling the surface morphology during AlGaIn growth is critical.

This work builds upon and is a logical extension of the work by Bryan et al. that presented an all-inclusive model for controlling the surface morphology in the homoepitaxial growth of AlN on vicinal surfaces.¹¹ It was shown that the surface kinetics and morphology could be controlled by properly managing the vapor supersaturation and substrate misorientation angle. By understanding the relationship between these two parameters, as they related to the surface supersaturation, one could predictably and reproducibly achieve a transition between 3D island growth, step-flow, or step-bunching in a variety of complementary parameter spaces. Since this framework was developed without a specific material in mind, it was suggested to be universally applicable to other materials.¹¹

In this study, we followed the same framework to understand the surface kinetics and to control surface morphology and composition of Al-rich AlGaIn. Al-rich AlGaIn films were grown by metalorganic chemical vapor deposition (MOCVD) on vicinal c-plane AlN and sapphire substrates to determine the role of surface steps and dislocations on the resulting surface morphology. The compositional uniformity and Al content in AlGaIn as it related to the surface morphology was also studied. The end result of this work was a scheme for controlling the surface, composition, and compositional uniformity of Al-rich AlGaIn thin films in MOCVD growth.

II. Experimental Procedure

Physical vapor transport deposition was implemented to grow the single crystalline boules used to generate the AlN substrates.¹²⁻¹⁴ These boules were cut to fabricate substrates with vicinal (0001) oriented surfaces to grow on. The substrates had a chemo-mechanically polished,

atomically smooth surface, free of surface defects based on inspection by optical and atomic force microscopies (AFM). Prior to growth, the AlN substrates were treated following the procedure that included solvent cleaning, acid etch and nitridation of the surface before epilayer deposition.^{11, 15} All of the films studied here were grown in a rf-heated, vertical, cold-walled, MOCVD reactor.¹⁶ For films grown on sapphire substrates, first an AlN template was grown. These AlN templates had atomically smooth bilayer stepped surfaces with RMS roughnesses of less than 200 pm for 5x5 μm^2 AFM scan areas.¹¹

For the results discussed in this study, all surfaces shown are of films that were either grown directly on the AlN substrates or on AlN templates on sapphire. The following growth conditions were used for these films: H_2 was used as the diluent gas with a flow rate of approximately 450 mmol/min during the entire growth; the trimethylaluminum (TMA) flow was varied from 7 to 28 $\mu\text{mol/min}$, the triethylgallium (TEG) flow was between 2 and 12 $\mu\text{mol/min}$, while the ammonia (NH_3) was fixed at around 13 mmol/min. All AlGaIn epitaxial films were grown at 1100°C and 20 Torr total pressure.

The Al content of the AlGaIn films was determined using reciprocal space mapping by high resolution x-ray diffraction (HRXRD) and a technique developed by Tweedie et al. that allows for separation of strain and composition effects in XRD.¹⁷ The HRXRD measurements were conducted using a Philips X'Pert Materials Research Diffractometer with a $\text{Cu } K_\alpha$ x-ray source. Conventional transmission electron microscopy (TEM) was implemented to study extended defects in these films using a JEOL 2000FX operating at 200 kV. TEM samples were prepared by mechanical wedge polishing followed by Ar^+ ion milling with a Fischione Model 1010 Ion Mill. For the films grown on the AlN substrates, TEM analysis confirmed that no new dislocations were introduced in the epitaxial layers, which indicated that the high quality of the

substrate was maintained throughout the MOCVD growth and all AlGa_N films were pseudomorphic. For films grown on sapphire, similar TEM analysis provided a consistent dislocation density around $1 \times 10^{10} \text{ cm}^{-2}$. High angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) was performed using a probe corrected FEI Titan G2 60-300 kV S/TEM at 200 kV for Z-contrast imaging of AlGa_N epitaxial layers. Photoluminescence (PL) spectra were acquired using a pulsed ArF excimer laser ($\lambda = 193 \text{ nm}$) along with a Princeton Instruments Acton SP2750 0.75 m monochromator, with a 150 grooves/mm grating, and a PIXIS:2KB_{UV} cooled charge-coupled device camera. All measurements were taken with an excitation and detection angle of 45° to the surface normal. An Asylum Research MFP-3D atomic force microscope in tapping mode was used to study the surface morphology of all the films. Before growth, both AlN and sapphire substrate surfaces were characterized by AFM to determine the misorientation angle relative to the [0001] direction. These measurements were further supported using standard HRXRD techniques.

III. Results and Discussion

Although the RMS roughness of all Al-rich AlGa_N films was below 1 nm, those grown on AlN templates on sapphire were relatively rough compared to those grown on the native AlN substrates. The larger dislocation density present in the films grown on sapphire substrates resulted in step-pinning and a high degree of spiral growth. This spiral growth mechanism resulted in overall rougher surfaces. Figure 1 shows the surface of an AlN and Al_{0.60}Ga_{0.40}N film grown on sapphire. The AlN surface had a much smoother surface relative to the Al_{0.60}Ga_{0.40}N surface, with over an order of magnitude difference in the RMS roughness. Step-pinning and spiral growth made it difficult to directly observe the long-range surface kinetics as related to

different growth modes. Therefore, to properly study the surface kinetics of Al-rich AlGa_N, it was necessary to grow films on low dislocation density substrates.

The amount of Ga in the alloy films directly affected the spirals as predicted by the Burton, Cabrera, and Frank (BCF) theory, i.e. the terrace width about a dislocation spiral was inversely proportional to the natural logarithm of the vapor supersaturation:¹⁸

$$\frac{1}{\lambda_0} \propto \ln(1 + \sigma), \quad (1)$$

where λ_0 and σ are the average terrace width and vapor supersaturation, respectively. A 2x2 μm^2 AFM image of the growth spirals is given in Figure 2. These spirals formed due to dislocations which had a component of the displacement vector normal to the growth surface, i.e. mixed and screw type dislocations. In particular, the spirals observed in Al-rich AlGa_N and AlN were double spirals comprising of two interlocking spiral ramps, each dislocation pinning two steps.¹⁹ By introducing Ga, the supersaturation was reduced since the equilibrium vapor pressure of Ga is much larger than that of Al. Studies on the 60–100% Al content AlGa_N films grown on sapphire indicated a direct correlation of increasing terrace width about a spiral with an increase of Ga content. Most importantly, for any further reduction in the RMS roughness of AlGa_N films using a sapphire substrate, the dislocation density must be reduced.

For the 60–100% Al content compositional range studied, the observed types of morphology were similar to those observed in AlN homoepitaxy.¹¹ Because of the low dislocation density of $< 10^3 \text{ cm}^{-2}$, spiral growth did not interfere with the steps, allowing for a detailed study of surface kinetics of Al-rich AlGa_N. Depending on growth conditions and substrate misorientation, macro-sized 3D islands, bilayer steps, and step-bunches were observed.

Figure 3 shows two $\text{Al}_{0.80}\text{Ga}_{0.20}\text{N}$ films grown on AlN substrates with different growth rates. The substrate misorientation angle, α , was kept constant at 0.17° . A 3D growth mode appeared for a growth rate of 1200 nm/hr. The islands were slightly tilted due to the misorientation angle. By reducing the growth rate to 600 nm/hr, a purely step flow growth mode was achieved and resulted in an atomically smooth bilayer step morphology with an RMS roughness < 50 pm for a $5 \times 5 \mu\text{m}^2$ AFM scan area. This demonstrated the transition from the 3D island growth to step flow growth mode by reducing the vapor supersaturation, as predicted by the BCF theory applied to the growth of III-nitrides.¹¹ By reducing the vapor supersaturation, the surface supersaturation was reduced, making the nucleation less likely. The relative relationship between the vapor supersaturation and the substrate misorientation determined the surface supersaturation, which had to be lower than the critical supersaturation for spontaneous nucleation in order to maintain atomically smooth bilayer step morphology, as discussed previously.¹¹

Figure 4 shows the step height and terrace width as a function of the misorientation angle for nominal $\text{Al}_{0.70}\text{Ga}_{0.30}\text{N}$ grown with a growth rate of 600 nm/hr on AlN substrates. Similar to the observations in AlN homoepitaxy,¹¹ for given growth conditions, a critical substrate misorientation angle, α^* , above which step-bunching occurred, was observed.

For the above growth conditions, the α^* was determined to be around 0.4° , as indicated by an abrupt and simultaneous increase in the step height and terrace width. In this analysis, the entire step-bunch was considered for the step height and width. An increase in α resulted in an increase in the step-bunch height while the terrace width remained relatively constant in the range from 0.4 to 0.55° misorientation and started to decrease gradually thereafter. Below this critical misorientation angle, only bilayer step morphology was observed with step heights

measured to be around $2.5 \pm 0.1 \text{ \AA}$. The measured terrace widths coincided with the calculated values for a given misorientation angle: $\lambda_0 = h/\tan(\alpha)$, where h is the step height. Similar trends were observed in AlN homoepitaxial films.¹¹ Figure 4 includes films misoriented toward [1-100], [11-20], and variations in between, which shows that the general trends for step height and terrace width were not affected by the misorientation azimuth.

It is important to note, that the critical misorientation angle was determined by given growth conditions. Since it depended on the surface diffusion length relative to the terrace width, it changed in the same direction as the change in the surface supersaturation. Therefore, in order to increase α^* , the diffusion length had to be reduced by an adjustment in the vapor supersaturation. At growth conditions for which the diffusion length was much greater than the terrace width, step-bunching was unavoidable.¹¹

It has been suggested in the literature that step-bunching was related to strain relaxation.²⁰ Since pseudomorphic AlGa_{0.3}N is strained to AlN and the total strain energy increases with AlGa_{0.3}N thickness, it is an ideal model system to test this hypothesis, as at some thickness, one should have accumulated enough strain energy to invoke a transition from bilayer growth to step-bunching. To test this idea, Al_{0.70}Ga_{0.30}N of various thicknesses (up to 700 nm) was deposited at the same conditions on AlN substrates. The resulting AlGa_{0.3}N surface showed perfect bilayer steps for all thicknesses without any evidence of step-bunching. HRXRD confirmed that the films were fully strained to the AlN substrate and no detectable relaxation had occurred. The same experiment was repeated on step-bunched AlN surfaces and the resulting AlGa_{0.3}N surfaces showed the same bilayer morphology as for the substrates with bilayer steps. This behavior led us to make two important conclusions: (1) step-bunching is controlled primarily by the surface kinetics while biaxial strain has a minor or no effect, and (2) a surface with preexisting step-

bunches can be recovered to bilayer steps by an appropriate adjustment of the surface supersaturation regardless of strain.

While it is intuitive that one should avoid step-bunching for any devices that utilize flat interfaces or quantum structures, it is less intuitive what effect step-bunching would have on a ternary alloy itself. Figure 5 shows AFM images of AlGaIn films grown during the same run but on two substrates with different misorientation angles: one below α^* and the other above. Clearly, the sample with offcut $< \alpha^*$ was smooth and showed bilayer steps (Figure 5 (left)), while the sample with offcut $> \alpha^*$ showed step-bunches (Figure 5 (right)). HRXRD did not show any noticeable differences in the crystalline quality between the two films, however, there was an indication of two distinct AlGaIn compositions for the step-bunched film. To further examine this phenomenon, low temperature (3K) PL spectra were taken. As shown in Figure 5 (left), the bilayer stepped film showed a single emission peak as expected for a uniform AlGaIn film with 81% of Al, whereas the step-bunched film showed two distinct peaks corresponding to two different AlGaIn compositions straddling the one shown in Figure 5 (left). Since AlGaIn forms a complete range of solid solutions without a miscibility gap and since the simultaneously grown film on a low offcut substrate showed single emission, the observed apparent phase separation had to be related to surface morphology. This compositional inhomogeneity was observed also in quantum well structures grown on step-bunched surfaces, and as a consequence, the electroluminescence exhibited multiple emission peaks. In addition, gain measurements by Guo et al. indicated that a lower energy peak of AlGaIn PL emission can possess significantly higher gain, pointing to an efficient energy transfer between the two levels.²¹ The presence of the double emission from that work is now attributed to the step-bunched morphology of the AlGaIn film.

In order to better understand the mechanism of the apparent phase separation in AlGaIn, HAADF STEM imaging was conducted. Ga-rich AlGaIn streaks were observed in Z-contrast STEM when step-bunching occurred, as seen in Figure 6 (right). However, these streaks were not present when bilayer steps were present, as indicated in Figure 6 (left). The streaks were approximately 2-5 nm in width and their angle changed as a function of the substrate misorientation angle, indicating a correlation between the step fronts and the Ga incorporation. These streaks were spaced relatively uniformly and could be thought of as quantum wells, explaining the observed high gain values.²¹

Indeed, a more detailed TEM study of the hetero-interface revealed that the Ga-rich streaks emanated from the step-bunches where high step density existed while Ga-poor material emanated from the flat terraces separating the step-bunches, as shown schematically in Figure 7. The direction of the streaks was consistent with the direction of step flow.

From the evidence presented above, it is clear that bilayer stepped AlGaIn is mandatory to achieve atomically smooth surfaces and for the growth of quantum structures. However, the above results suggest that AlGaIn composition is a function of the step density and this should be true even for the bilayer growth mode. Figure 8 shows the Ga content for AlGaIn films grown on substrates with different misorientation angles but at identical growth conditions, where bilayer steps were maintained throughout the growth. With an increase of the misorientation angle from ~ 0.05 to 0.40° there was an increase in the Ga content of more than 10%. This is a significant change in the composition that results in fluctuations in the emission wavelength by over 20 nm. As a consequence, if one is to achieve the targeted emission in an LED or LD grown on native substrates, one needs to control the substrate misorientation angle very precisely, or

appropriately change surface kinetics to compensate for the variation in terrace width from substrate to substrate.

As with growth type, this effect can also be understood and predicted by considering surface kinetics.¹⁸ For AlGaN, both the Al and Ga adatoms must be considered. The arrival and desorption fluxes will not necessarily be the same for Ga and Al, as different species have different surface mobilities and residence times.

Following the BCF-based model presented for pure AlN, the growth rate, R , is directly related to the surface diffusion flux, J_s , at the step edge, $x=\lambda_0/2$.¹¹ Knowing the diffusion flux at the step edge for both species allows for a rough estimate of the composition, assuming that Ga and Al do not interact on the surface. In this case, the Al content in AlGaN can be expressed as:

$$x_{Al} = \frac{J_{s,Al}\left(\frac{\lambda_0}{2}\right)}{J_{s,Al}\left(\frac{\lambda_0}{2}\right) + J_{s,Ga}\left(\frac{\lambda_0}{2}\right)} = \frac{R_{Al}}{R_{Al} + R_{Ga}}, \quad (2)$$

where

$$R_{Al} = \frac{2h\lambda_{s,Al}}{n_0\lambda_0} \left(J_{Al} - \left(\frac{n_{s0}}{\tau_s} \right)_{Al} \right) \tanh\left(\frac{\lambda_0}{2\lambda_{s,Al}} \right) \quad (3)$$

and

$$R_{Ga} = \frac{2h\lambda_{s,Ga}}{n_0\lambda_0} \left(J_{Ga} - \left(\frac{n_{s0}}{\tau_s} \right)_{Ga} \right) \tanh\left(\frac{\lambda_0}{2\lambda_{s,Ga}} \right). \quad (4)$$

Here, λ_s , n_0 , J , n_{s0} , and τ_s are the surface diffusion length, density of adatom sites on the surface, adatom arrival flux from the gas phase to the surface, equilibrium adatom density at the step edge and mean residence time for appropriate species, respectively.

When λ_s is much greater than λ_0 , the hyperbolic tangent term tends to $\lambda_0/2\lambda_s$. Conversely, if λ_s is much smaller than λ_0 , the hyperbolic tangent term tends to 1, which gives the following relationships for the two limiting conditions:

$$R \approx \begin{cases} \frac{2h\lambda_s}{n_0\lambda_0} \left(J - \frac{n_{s0}}{\tau_s} \right), & \lambda_0 \gg \lambda_s \\ \frac{h}{n_0} \left(J - \frac{n_{s0}}{\tau_s} \right), & \lambda_0 \ll \lambda_s \end{cases} \quad (5)$$

The only thing that is changing across the surface in **Figure 6 (right)** (see also Figure 7) is the terrace width λ_0 , which influences the incorporation of the metal species: short terraces incorporate more Ga with respect to Al while long terraces incorporate less Ga. This observation suggests that $\lambda_{s,Ga} < \lambda_{s,Al}$, which is consistent with the model presented above. This is easily seen in the case when $\lambda_0 > \lambda_{s,Ga}$ and $\lambda_0 < \lambda_{s,Al}$, which gives the composition deviation as:

$$x_{Ga} = \frac{A}{A+B\lambda_0}, \quad (6)$$

where A and B are taken as constants which include all the terms from Equation 5 that remain fixed under the assumed conditions. Thus, the longer the terrace width the lower the Ga incorporation for given growth conditions. The experimental results and model indicate that the residence time and diffusion length of Ga must be lower than that of Al. This is contrary to the common assumption that the Ga adatoms have a much longer diffusion length than the Al adatoms.²² This work clearly shows that the control of the substrate misorientation and growth mode are crucial for growing uniform AlGaN with predictable Ga content.

IV. Conclusions

It has been demonstrated that the growth mode and composition of AlGaN strongly depend on the initial surface condition and surface kinetics. Copious mixed and screw-type dislocations present in the growth on sapphire-based templates promoted spiral growth, which roughened the surface and led to short-range compositional variation in AlGaN. Atomically smooth, Al-rich AlGaN surfaces required low dislocation densities offered by pseudomorphic growth on native AlN substrates, however, while this was a necessary condition it was not sufficient. For desired growth mode and composition, surface kinetics needed to be controlled as well. This could be achieved through the control of the surface supersaturation and substrate misorientation angle.

By growing pseudomorphically on native AlN substrates with dislocation density $< 10^3 \text{ cm}^{-2}$, parallel bilayer steps, free of step-pinning and spiraling, with RMS roughness of less than 50 pm for a $5 \times 5 \text{ }\mu\text{m}^2$ AFM scan area could be maintained. AlGaN composition depended on the terrace width and surface supersaturation of the Al and Ga species, which was explained through a BCF theory-based model. Apparent phase separation of AlGaN was determined not to be an intrinsic property of AlGaN and was explained within the same model as a result of step-bunching. The presented experimental results clearly show the importance of controlling the surface kinetics in AlGaN growth while the developed model offers a convenient control scheme for achieving atomically-smooth, bilayer-stepped surfaces of desired AlGaN composition.

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References

1. J. Xie, S. Mita, Z. Bryan, W. Guo, L. Hussey, B. Moody, R. Schlessner, R. Kirste, M. Gerhold and R. Collazo, *Applied Physics Letters* **102**, 171102 (2013).
2. J. R. Grandusky, S. R. Gibb, M. C. Mendrick, C. Moe, M. Wraback and L. J. Schowalter, *Applied Physics Express* **4**, 082101 (2011).
3. R. Dalmau, B. Moody, R. Schlessner, S. Mita, J. Xie, M. Feneberg, B. Neuschl, K. Thonke, R. Collazo and A. Rice, *Journal of the Electrochemical Society* **158**, H530 (2011).
4. T. Akasaka, H. Gotoh, Y. Kobayashi and H. Yamamoto, *Advanced Materials* **24**, 4296 (2012).
5. J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson and A. Y. Cho, *Science* **264**, 553 (1994).
6. G. Kusch, H. Li, P. R. Edwards, J. Bruckbauer, T. C. Sadler, P. J. Parbrook and R. W. Martin, *Applied Physics Letters* **104**, 092114 (2014).
7. R. L. Schwoebel, *Journal of Applied Physics* **40**, 614 (1969).
8. R. L. Schwoebel and E. J. Shipsey, *Journal of Applied Physics* **37**, 3682 (1966).
9. P. Venezuela and J. Tersoff, *Physical Review B* **58**, 10871 (1998).
10. K. Mukherjee, D. Beaton, T. Christian, E. Jones, K. Alberi, A. Mascarenhas, M. Bulsara and E. Fitzgerald, *Journal of Applied Physics* **113**, 183518 (2013).
11. I. Bryan, Z. Bryan, S. Mita, A. Rice, J. Tweedie, R. Collazo and Z. Sitar, *Journal of Crystal Growth* **438**, 81 (2016).
12. Z. Herro, D. Zhuang, R. Schlessner, R. Collazo and Z. Sitar, *Journal of Crystal Growth* **286**, 205 (2006).
13. D. Zhuang, Z. Herro, R. Schlessner and Z. Sitar, *Journal of Crystal Growth* **287**, 372 (2006).
14. P. Lu, R. Collazo, R. Dalmau, G. Durkaya, N. Dietz, B. Raghothamachar, M. Dudley and Z. Sitar, *Journal of Crystal Growth* **312**, 58 (2009).
15. I. Bryan, A. Rice, L. Hussey, Z. Bryan, M. Bobea, S. Mita, J. Xie, R. Kirste, R. Collazo and Z. Sitar, *Applied Physics Letters* **102**, 061602 (2013).
16. S. Mita, R. Collazo, A. Rice, R. Dalmau and Z. Sitar, *Journal of Applied Physics* **104**, 013521 (2008).
17. J. Tweedie, R. Collazo, A. Rice, J. Xie, S. Mita, R. Dalmau and Z. Sitar, *Journal of Applied Physics* **108**, 043526 (2010).
18. W.-K. Burton, N. Cabrera and F. Frank, *Philosophical Transactions of the Royal Society of London. Series A: Mathematical, Physical and Engineering Sciences* **243**, 299 (1951).
19. B. Heying, E. Tarsa, C. Elsass, P. Fini, S. DenBaars and J. Speck, *Journal of Applied Physics* **85**, 6470 (1999).
20. J. Tersoff, Y. Phang, Z. Zhang and M. Lagally, *Physical Review Letters* **75**, 2730 (1995).
21. W. Guo, Z. Bryan, J. Xie, R. Kirste, S. Mita, I. Bryan, L. Hussey, M. Bobea, B. Haidet and M. Gerhold, *Journal of Applied Physics* **115**, 103108 (2014).
22. T. Narita, Y. Honda, M. Yamaguchi and N. Sawaki, *Physica Status Solidi B* **243**, 1665 (2006).

Figure 1: AFM images representing AlN (left) and Al_{0.60}Ga_{0.40}N (right) heteroepitaxial films grown on AlN templates on sapphire.

Figure 2: AFM image of double growth spirals on AlGa_N films grown on AlN templates on sapphire.

Figure 3: AFM images representing $\text{Al}_{0.80}\text{Ga}_{0.20}\text{N}$ on AlN homoepitaxial films grown at two different supersaturations.

Figure 4: Step height and width as a function of misorientation angle for nominal $\text{Al}_{0.70}\text{Ga}_{0.30}\text{N}$ on AlN homoepitaxial films. The $\text{Al}_{0.70}\text{Ga}_{0.30}\text{N}$ was grown with a growth rate and temperature of 600 nm/hr and 1100 °C, respectively.

Figure 5: AFM images representing AlGaN grown on a low (upper left) and high (upper right) misoriented AlN native substrate resulting in bilayer and step-bunched morphologies, respectively. Corresponding low temperature PL spectra along with specified Al contents (lower left and right).

Figure 6: HAADF STEM images along the $\langle 1-100 \rangle$ zone axis of bilayer stepped (left) and step-bunched (right) AlGa_N grown on homoepitaxial AlN.

Figure 7: Schematic representing the locations of the Ga-rich and Ga-poor regions in relation to the step density at the surface.

Figure 8: The dependence of misorientation angle on the Ga content of AlGa_N films with bilayer step morphology.