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# Surface morphology evolution of m-plane (1 $\bar{1}$ 00) GaN during molecular beam epitaxy growth: Impact of Ga/N ratio, miscut direction, and growth temperature

Jiayi Shao

*Birck Nanotechnology Center, Purdue University, shao20@purdue.edu*

Liang Tang

*Birck Nanotechnology Center, Purdue University*

Colin Edmunds

*Purdue University*

Geoff C. Gardner

*Birck Nanotechnology Center, Purdue University, geoff@purdue.edu*

Oana Malis

*Birck Nanotechnology Center, Purdue University, omalis@purdue.edu*

*See next page for additional authors*

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**Authors**

Jiayi Shao, Liang Tang, Colin Edmunds, Geoff C. Gardner, Oana Malis, and Michael J. Manfra



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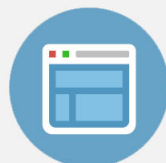
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# Surface morphology evolution of m-plane ( $\bar{1}\bar{1}00$ ) GaN during molecular beam epitaxy growth: Impact of Ga/N ratio, miscut direction, and growth temperature

Jiayi Shao,<sup>1,2</sup> Liang Tang,<sup>1,2</sup> Colin Edmunds,<sup>2</sup> Geoff Gardner,<sup>1,3</sup> Oana Malis,<sup>1,2,a)</sup> and Michael Manfra<sup>1,2,3,4,a)</sup>

<sup>1</sup>Birck Nanotechnology Center, Purdue University, West Lafayette, Indiana 47907, USA

<sup>2</sup>Department of Physics, Purdue University, West Lafayette, Indiana 47907, USA

<sup>3</sup>School of Materials Engineering, Purdue University, West Lafayette, Indiana 47907, USA

<sup>4</sup>School of Electrical and Computer Engineering, Purdue University, West Lafayette, Indiana 47907, USA

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We present a systematic study of morphology evolution of  $[\bar{1}\bar{1}00]$  m-plane GaN grown by plasma-assisted molecular beam epitaxy on free-standing m-plane substrates with small miscut angles towards the  $-c$   $[000\bar{1}]$  and  $+c$   $[0001]$  directions under various gallium to nitrogen (Ga/N) ratios at substrate temperatures  $T = 720^\circ\text{C}$  and  $T = 740^\circ\text{C}$ . The miscut direction, Ga/N ratio, and growth temperature are all shown to have a dramatic impact on morphology. The observed dependence on miscut direction supports the notion of strong anisotropy in the gallium adatom diffusion barrier and growth kinetics. We demonstrate that precise control of Ga/N ratio and substrate temperature yields atomically smooth morphology on substrates oriented towards  $+c$   $[0001]$  as well as the more commonly studied  $-c$   $[000\bar{1}]$  miscut substrates. © 2013 AIP Publishing LLC.

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## I. INTRODUCTION

The nonpolar surfaces (m-plane and a-plane) of GaN have attracted significant interest as there are neither spontaneous polarization nor piezoelectric polarization fields along these growth directions.<sup>1–3</sup> This simplification is beneficial for the design of many nitride-based optoelectronic and electronic devices.<sup>4–11</sup> In particular, the lack of internal polarization fields also simplifies the design of complex near-infrared (IR) optoelectronic devices such as the quantum cascade laser.<sup>12–15</sup> While considerable effort has been devoted to the study of nonpolar GaN growth by metalorganic vapour phase epitaxy (MOCVD),<sup>20–25</sup> much remains to be understood concerning the impact of molecular-beam epitaxy growth conditions and substrate orientation on m-plane films grown by plasma-assisted molecular beam epitaxy (PAMBE).<sup>26–29</sup> For PAMBE homoepitaxy along the  $c$ -axis, it is well known that the gallium to nitrogen flux ratio (Ga/N), substrate temperature, and substrate preparation have an enormous impact on the resulting surface morphology, as well as the structural, optical, and electronic properties.<sup>16,30–32</sup>

In Waltereit's original report of plasma-assisted MBE growth of  $[\bar{1}\bar{1}00]$  m-plane GaN on (100)-oriented  $\gamma$ -LiAlO<sub>2</sub> substrates, elongated stripes were observed along the  $[\bar{1}\bar{1}\bar{2}0]$  direction with atomic force microscopy (AFM) imaging.<sup>1</sup> More recently, Sawicka *et al.*<sup>17</sup> studied the impact of substrate miscut direction on growth morphology for m-plane GaN grown by PAMBE under nitrogen-rich conditions at  $T = 730^\circ\text{C}$  and determined that, for their growth conditions, the best miscut angle direction for achieving atomically flat m-plane GaN layers is  $0.2^\circ$ – $0.5^\circ$  towards  $-c$  with a preferable

step-flow direction of  $17^\circ \pm 2^\circ$  off the  $[000\bar{1}]$  axis. These experimental results were interpreted as confirmation of theoretical analysis of Lymperakis *et al.*<sup>18</sup> Lymperakis *et al.* analyzed adatom kinetics on m-plane GaN and concluded that growth anisotropy arises from the anisotropy of Ga adatom diffusion barriers along and perpendicular to the  $c$ -axis.<sup>18,19</sup>

Despite these important experimental and theoretical works, a systematic study of m-plane GaN surface morphology evolution under variable Ga/N ratio and miscut direction is still lacking. In this paper, we study surface morphology evolution of m-plane GaN during PAMBE growth with a wide range of Ga/N ratio on free-standing m-plane substrates with small miscut towards the  $-c$   $[000\bar{1}]$  and  $+c$   $[0001]$  axes. We explore both nitrogen-rich and gallium-rich conditions at substrate temperatures  $T = 720^\circ\text{C}$  and  $T = 740^\circ\text{C}$ . Different surface morphologies with varying step widths were observed as a function of Ga/N ratio. Importantly, the miscut direction and angle had a dramatic impact on morphology, supporting the notion of strong anisotropy in the gallium adatom diffusion barrier and growth kinetics. We find that surface morphology is also extremely sensitive to substrate temperature in the range  $T = 720^\circ\text{C}$  to  $740^\circ\text{C}$ . Precise control of substrate temperature and Ga/N ratio can yield extremely smooth surface morphology on substrates miscut both towards  $[0001]$  and  $[000\bar{1}]$ .

## II. EXPERIMENT

Our m-plane substrates are provided by Kyma Technologies Inc. They have rectangular shape (approximately 10 mm by 5 mm) and are cut from few-millimeter thick free-standing crystals grown in the  $c$ -direction. The nominal threading dislocation density is  $\sim 5 \times 10^6 \text{ cm}^{-2}$ . The quoted miscut angles for each substrate are determined by

<sup>a)</sup>Authors to whom correspondence should be addressed. Electronic addresses: omalis@purdue.edu and mmanfra@purdue.edu

x-ray diffraction (XRD) by Kyma Technologies. Prior to loading into the MBE, the wafers are cleaned with acetone, methanol, isopropanol, deionized water, and dried with  $N_2$  gas. The substrates are mounted onto a 2 in. GaN/sapphire template for mechanical stability and to aid substrate thermometry. The template we employ consists of an  $80\text{ }\mu\text{m}$  thick layer of GaN grown by hydride vapor phase epitaxy (HVPE) grown on a 2 in. sapphire substrate. The backside of the  $80\text{ }\mu\text{m}$  GaN/sapphire template is coated with tungsten to facilitate uniform heating in the MBE system. The small m-plane substrate is mounted with gallium metal to this 2 in. GaN/sapphire template in order to fit into standard 2 in. MBE hardware. Two m-plane substrates were grown on simultaneously in order to explore the impact of substrate miscut direction (towards both  $-c$  and  $+c$ ) on surface morphology. The miscut angles of our  $-c$  miscut substrates ranges from  $0.2^\circ$  to  $1.45^\circ$  while the  $+c$  miscut substrates range from  $0.4^\circ$  to  $0.6^\circ$ . Nominally, all substrates have no miscut towards the  $a$ -axis  $[11\bar{2}0]$  within an accuracy of  $\pm 0.1^\circ$ . As received, the substrates are smooth with typical root-mean-square (rms) roughness of  $0.2\text{--}0.3\text{ nm}$  over  $16\text{ }\mu\text{m}^2$ . We note that polishing scratches remaining from substrate preparation are sometimes evident.

$50\text{ nm}$  thick layers of GaN were grown by PAMBE at a constant substrate temperature of  $740^\circ\text{C}$  or  $720^\circ\text{C}$  on both  $-c$  and  $+c$  miscut m-plane GaN substrates. Gallium flux was supplied by a conventional effusion cell and nitrogen flux was provided by a Veeco Unibulb radio-frequency plasma source operating at  $300\text{ W}$  forward power with  $0.5\text{ sccm}$  of nitrogen ( $N_2$ ) flow. The substrate temperature is monitored with a pyrometer. The growth rate of m-plane GaN under varying Ga beam flux is initially calibrated by transmission electron microscopy (TEM) using a structure that consists of  $20\text{ min}$  of GaN growth under several different Ga beam fluxes with AlGaIn marker layers to separate each GaN layer. These TEM images are not shown here. For comparison, the maximum, nitrogen-limited, growth rate for c-plane GaN at  $740^\circ\text{C}$  with the stated plasma conditions is  $7\text{ nm/min}$ . The growth rates of m-plane GaN as a function of Ga beam flux at substrate temperature of  $740^\circ\text{C}$  and  $720^\circ\text{C}$  on m-plane are shown in Figure 1(a). Operationally, we define the Ga

beam flux corresponding to a saturated growth rate of  $7\text{ nm/min}$  at  $720^\circ\text{C}$  as a ratio  $\text{Ga/N} = 1$  for m-plane growth. Based on this definition, the Ga/N ratio for m-plane growth is plotted in Figure 1(b). Compared to the growth of c-plane GaN in our MBE, a higher Ga beam flux is required to reach the saturation growth rate on m-plane at a given substrate temperature. This fact indicates that the Ga sticking coefficient (thermal decomposition rate) on m-plane is lower (higher) than that on c-plane at a given substrate temperature. Before growth and after each subsequent  $50\text{ nm}$  GaN growth, the surface morphology was measured by AFM in tapping mode and the rms roughness was measured over areas of  $1\text{ }\mu\text{m}^2$ ,  $16\text{ }\mu\text{m}^2$ , and  $100\text{ }\mu\text{m}^2$ . We examined morphology at three different locations along the long axis of the sample: the center of the substrate,  $2.5\text{ mm}$  left of the center, and  $2.5\text{ mm}$  right of the center. However, our results show that, for each substrate, the surface morphology is similar at these three locations and the deviation in the RMS roughness is less than  $10\%$ . The images discussed in the remainder of the paper are taken from the center of each substrate.

### III. RESULTS AND DISCUSSION

#### A. Impact of Ga/N ratio on surface morphology on $-c$ miscut m-plane substrates at $740^\circ\text{C}$

The surface morphology generated over a  $16\text{ }\mu\text{m}^2$  area during PAMBE growth on  $-c$  miscut m-plane GaN substrates at  $T = 740^\circ\text{C}$  is summarized in Figure 2. The first 3 specimens (a)–(c) with Ga/N ratio  $0.30$ ,  $0.40$ , and  $0.55$  were grown sequentially on a single substrate with miscut toward  $-c = 0.46^\circ$ . Under plasma rich-growth conditions, the rms roughness over  $16\text{ }\mu\text{m}^2$  decreased from  $0.46\text{ nm}$  for the starting substrate to  $0.2\text{ nm}$  after  $50\text{ nm}$  of growth under Ga/N ratio of  $0.3$ , and stayed at  $0.2\text{ nm}$  under Ga/N ratio of  $0.4$ , although clear atomic steps were less readily discerned with AFM after growth with  $\text{Ga/N} = 0.4$ . Thus, we confirm the results of Ref. 17 that smooth films can be grown under nitrogen-rich conditions on m-plane GaN substrates with miscut angles towards  $-c$   $[000\bar{1}]$ . However, further increase of the Ga/N ratio produced dramatic and unanticipated morphology evolution. With  $\text{Ga/N} = 0.55$ , there is a clear transition to a terraced, or

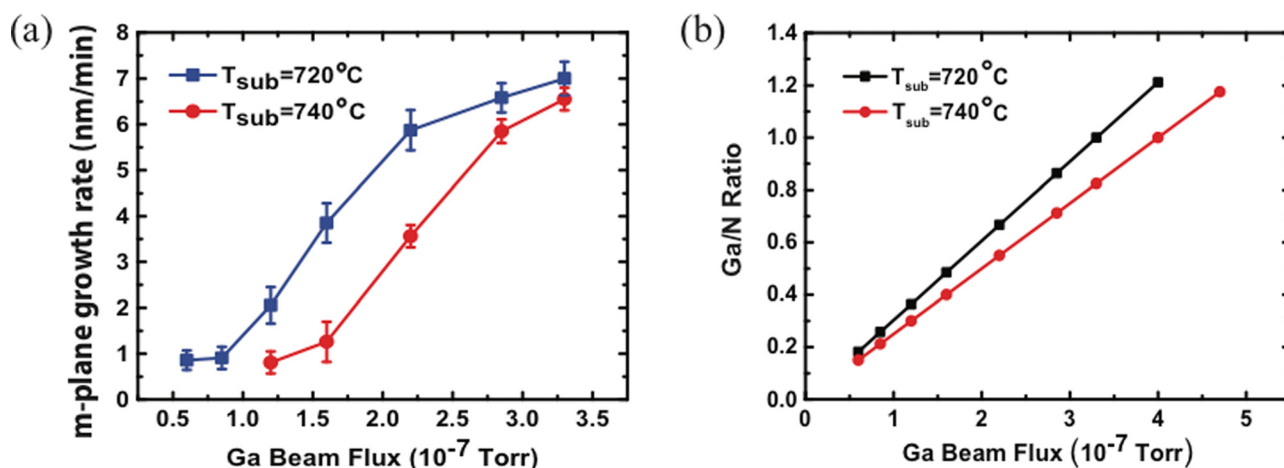


FIG. 1. (a) Growth rates of m-plane GaN as a function of Ga beam flux at substrate temperature of  $740^\circ\text{C}$  and  $720^\circ\text{C}$  (b) Ga/N ratio as a function of Ga beam flux at substrate temperature of  $740^\circ\text{C}$  and  $720^\circ\text{C}$ .



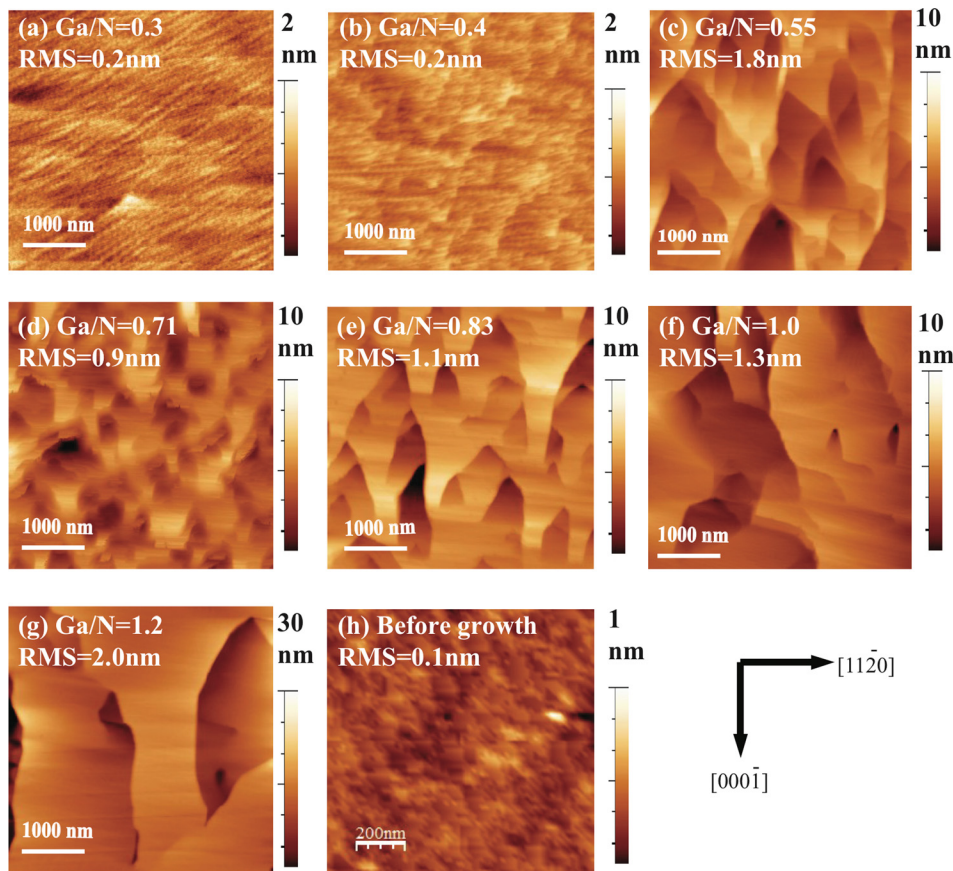


FIG. 2. Surface morphology evolution of m-plane GaN as a function of Ga/N ratio on  $-c$  miscut substrates at  $740^{\circ}\text{C}$  over an area of  $16\mu\text{m}^2$ . The first 3 specimens (a)–(c) were grown sequentially on a single substrate with miscut toward  $-c = 0.46^{\circ}$ . The specimen (d) was grown on a substrate with miscut toward  $-c = 0.67^{\circ}$ . The last 3 specimens (e)–(g) were grown sequentially on another single substrate with miscut toward  $-c = 0.61^{\circ}$ . Typical morphology of a starting Kyma substrate with miscut toward  $-c = 0.14^{\circ}$  prior to MBE growth is shown in (h). Note this last image is over an area of  $1\mu\text{m}^2$ .

slate-like, morphology. The top of the slates appear to be quite smooth possessing visible atomic steps. Nevertheless, these slates are separated by deep trenches, resulting in a dramatically increased rms roughness of  $1.8\text{ nm}$  over  $16\mu\text{m}^2$ . We also note that these terraces often possess a roughly triangular shape and are oriented along the  $[0001]$  direction, not along the  $[11\bar{2}0]$  where the Ga-adatom diffusion barrier is theoretically expected to be lower.<sup>18,19</sup> We have carefully and repeatedly confirmed that the orientation of this anisotropic morphology is along the  $[0001]$  direction. This result suggests that while the anisotropy of Ga-adatom diffusion barrier on the m-plane influences surface morphology, other factors like the relative growth rates of exposed  $c$ - and  $a$ -plane surfaces on miscut substrates may play an equally important role. In order to test whether this transformation in morphology continued at higher Ga/N ratio, we then loaded an additional substrate. This procedure was necessary to guarantee that subsequent growths were not unduly biased by growing on an already roughened morphology. The sample in Fig. 2(d) with Ga/N ratio 0.71 was grown on a substrate with miscut toward  $-c = 0.67^{\circ}$ . The last 3 specimens (e)–(g) with Ga/N ratio 0.83, 1.0, and 1.2 were grown sequentially on another single substrate with miscut toward  $-c = 0.61^{\circ}$ . It is apparent that the general morphological features are maintained at higher Ga/N ratios of 0.71, 0.83, 1.0, and 1.2. The RMS roughness was approximately  $1\text{ nm}$  over  $16\mu\text{m}^2$  and the terraced morphology was still evident, indicating that the terraced morphology is representative of high Ga/N ratios at  $T = 740^{\circ}\text{C}$  for miscut towards the  $[000\bar{1}]$  direction. A representative image of m-plane substrate morphology prior to initiating MBE growth is shown in (h).

At this juncture, a few comments are warranted. Unlike for PAMBE growth at  $740^{\circ}\text{C}$  on  $c$ -plane substrates, smooth film morphologies ( $\text{RMS} < 1\text{ nm}$ ) are obtained in nitrogen-rich growth regime. Thus, we confirm the results of Ref. 17 and extend these findings to a lower range of nitrogen-rich growth conditions. Interestingly, at  $\text{Ga/N} = 0.4$  and  $T = 740^{\circ}\text{C}$ , the surface morphology is accompanied by non-parallel step flow, reminiscent of the behavior observed under similar conditions in Ref. 17 with  $\text{Ga/N} = 0.6$ , and  $T = 730^{\circ}\text{C}$ . In our study, the large scale terraced morphology appears stable at Ga/N ratio above 0.55 and the features are elongated along the  $[0001]$  direction, not  $[11\bar{2}0]$ . We suggest that a small additional miscut towards  $[11\bar{2}0]$  can result in slate-like morphology along  $[0001]$  and may be responsible for the morphology observed here.<sup>17</sup> It is important to note that on the  $1\mu\text{m}$  length scale, these films are still extremely smooth ( $\text{rms} < 0.5\text{ nm}$ ) and display visible atomic steps. In total, these results indicate that growth dynamics and resulting surface morphology are extremely sensitive to Ga/N ratio for growth of GaN on m-plane substrates miscut approximately  $\sim 0.5^{\circ}$  towards  $-c$  at  $T = 740^{\circ}\text{C}$ . Furthermore, slight variations in miscut towards  $[11\bar{2}0]$  can influence the direction of anisotropic growth.

## B. Impact of Ga/N ratio on surface morphology on $+c$ miscut m-plane substrates at $740^{\circ}\text{C}$

Substrate miscut clearly plays a crucial role for determining surface morphology for m-plane growth. Yet little information is currently available concerning PAMBE growth

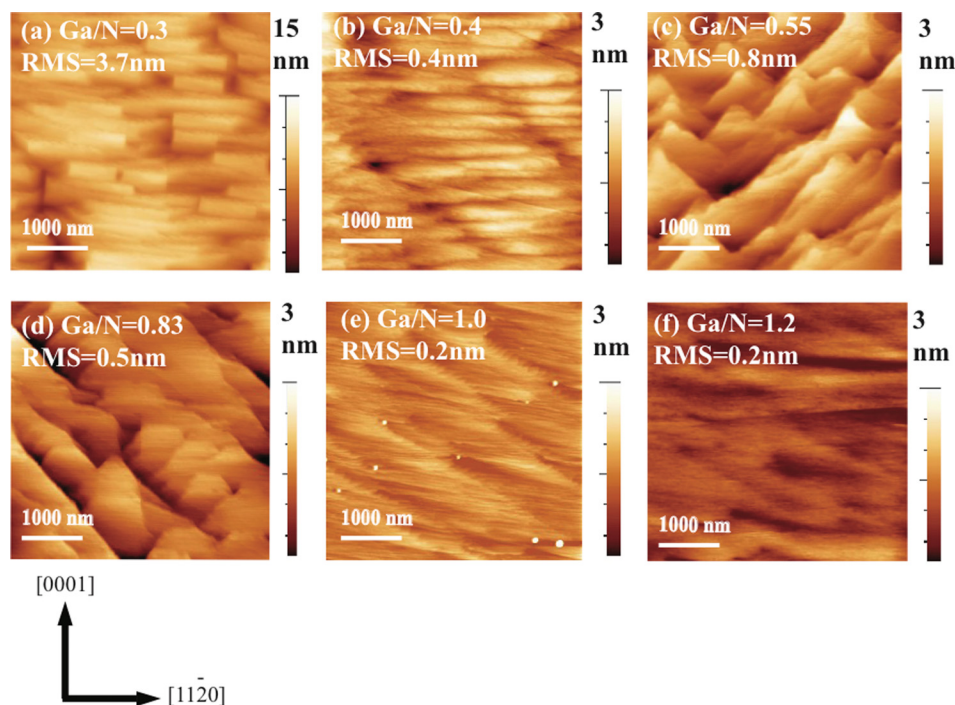


FIG. 3. The surface morphology evolution on +c miscut m-plane GaN substrates at  $T = 740^\circ\text{C}$ . The first 3 specimens (a)–(c) were grown on a single substrate with miscut toward  $+c = 0.38^\circ$ . Specimen (c) was grown first while specimen (a) was grown last. The last 3 specimens (d)–(f) were grown sequentially on another single substrate with miscut toward  $+c = 0.52^\circ$ .

using substrate miscut towards  $+c[0001]$ . Sawicka *et al.* noted that the surface has a strong tendency to roughen in the shape of rectangular terraces.<sup>17</sup> Image analysis was limited to one specific Ga/N = 0.6 and  $T = 730^\circ\text{C}$ . Data for our growth study on +c miscut m-plane GaN substrates at  $T = 740^\circ\text{C}$  over a broad range of Ga/N ratios are summarized in Figure 3. The first 3 specimens (a)–(c) with Ga/N ratios 0.30, 0.40, and 0.55 were grown on a single substrate with miscut toward  $+c = 0.38^\circ$ . Specimen (c) was grown first while specimen (a) was grown last. The lowest Ga/N ratio of 0.30 resulted in a slate-like morphology with large RMS roughness. The last 3 specimens (d)–(f) with Ga/N ratio 0.83, 1.0, and 1.2 were grown sequentially on another single substrate with miscut toward  $+c = 0.52^\circ$ . In Ref. 17, slate-like morphology was observed for growth under nitrogen rich conditions with a miscut toward +c. The direction of the slates were along the  $[11\bar{2}0]$  direction. Indeed, with Ga/N ratios of 0.3 and 0.4, we observe similar structure including a high rms roughness of 3.7 nm over  $16\mu\text{m}^2$  for Ga/N = 0.3. However, at increased Ga/N ratio, a dramatically improved morphology is realized. Using Ga/N ratio  $\geq 1$  yields much flatter surfaces. At Ga/N = 1, a rms roughness of 0.2 nm over  $16\mu\text{m}^2$  is measured, indicating that atomically flat surfaces can be obtained via PAMBE growth on m-plane substrates miscut towards +c under the appropriate growth stoichiometry at  $T = 740^\circ\text{C}$ . This result greatly expands the parameter space of substrate

preparations available for PAMBE growth. The impact of using each direction (+c and -c) on AlGaIn/GaN superlattice formation for intersubband devices will be described in an upcoming publication.

### C. Impact of growth temperature: $740^\circ\text{C}$ vs. $720^\circ\text{C}$ for -c and +c miscut substrates

Given the dependence of growth rate on substrate temperature displayed in Figure 1, it is natural to ask how substrate temperature influences the surface morphology for m-plane GaN growth. For m-plane samples miscut towards -c, it was shown in Figure 2 that while large scale smooth surface morphology could be obtained at Ga/N = 0.4, a terraced surface morphology persisted at the highest Ga/N ratios explored (Ga/N ratios 1 and 1.2) for growth at  $T = 740^\circ\text{C}$ . We also explored growth at reduced substrate temperature of  $720^\circ\text{C}$  under high Ga/N ratio conditions. In general, a significant widening of the terraces was evident with a concomitant reduction in the size and depth of the intervening trenches. Figure 4 reveals the surface morphology generated on three different length scales with Ga/N = 1.2,  $T = 720^\circ\text{C}$ , and the substrate is miscut towards -c at an angle of  $0.2^\circ$ . Here, very smooth morphology is obtained at all length scales with atomic steps clearly visible in the  $1\mu\text{m}^2$  image. Note that damage associated with substrate

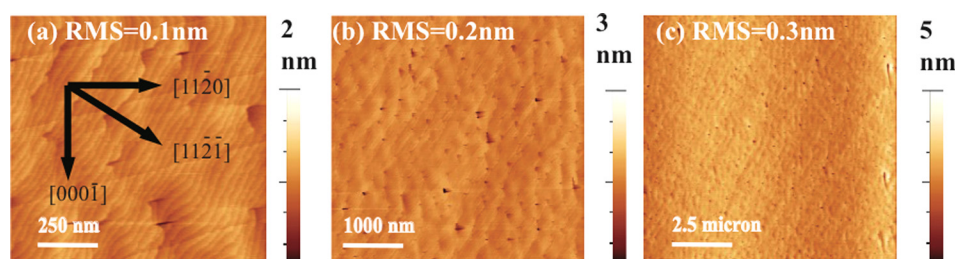


FIG. 4. Surface morphology of 50 nm GaN growth on -c miscut m-plane GaN substrate with  $T = 720^\circ\text{C}$  and Ga/N = 1.2. The substrate is miscut towards  $-c = 0.2^\circ$ . (a) over  $1\mu\text{m}^2$  area, (b) over  $16\mu\text{m}^2$  area, and (c) over  $100\mu\text{m}^2$  area.



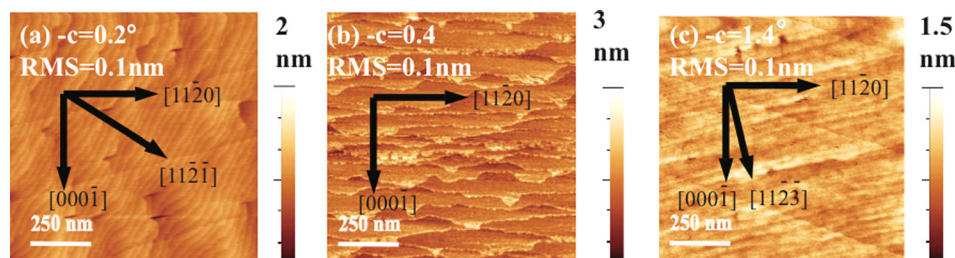


FIG. 5. Surface morphology for 100 nm of GaN grown at  $T = 720^\circ\text{C}$  and  $\text{Ga}/\text{N} = 1.2$  for three substrates with miscut angle of (a)  $0.2^\circ$ , (b)  $0.4^\circ$ , and (c)  $1.4^\circ$  towards  $-c$ .

polishing may also be visible. The utilization of reduced substrate temperature evidently suppresses the formation of the deep trenches seen at  $T = 740^\circ\text{C}$ , resulting in surface morphology suitable for heterostructure growth. This indicates that the growth kinetics dramatically changed when substrate temperature decreased from  $740^\circ\text{C}$  to  $720^\circ\text{C}$ .

We speculate that the improvement in surface morphology at  $720^\circ\text{C}$  arises as the lateral growth rates in the  $c$ - and  $a$ -directions become closer in value. As seen in Figure 1, the growth rate of along the  $m$ -direction GaN at a given gallium beam flux increases substantially as the substrate temperature is reduced from  $740^\circ\text{C}$  to  $720^\circ\text{C}$ , coming closer the nitrogen-limited growth rate of 7 nm/min observed for both the  $c$ - and  $m$ -directions. Although not proven here, it is reasonable to think that growth rate for  $a$ -direction would evolve in a similar fashion as that observed for  $m$ -direction. At a decreased substrate temperature of  $720^\circ\text{C}$  and high gallium flux, the growth rate along the  $a$ -direction may be closer to the nitrogen-limited growth rate as well. In this case, the growth rates in the  $a$ - and  $c$ -directions would be nearly equal, producing more isotropic growth and smoother surface morphology.

For substrates miscut towards  $+c$ , as shown in Figure 3, high  $\text{Ga}/\text{N}$  ratios already yielded smooth epilayers at  $T = 740^\circ\text{C}$ . Decreasing the substrate temperature to  $T = 720^\circ\text{C}$  did not substantially alter this behavior and smooth surface morphologies were once again obtained.

For completeness, we also investigated whether the surface morphology on  $-c$  miscut substrates at growth temperature  $720^\circ\text{C}$  is impacted significantly for changing the miscut angle towards  $-c$   $[000\bar{1}]$ . Figure 5 displays representative data over  $1\ \mu\text{m}^2$  for 100 nm films grown with  $\text{Ga}/\text{N} = 1.2$  on substrates with miscut of  $0.2^\circ$ ,  $0.4^\circ$ , and  $1.4^\circ$  towards  $-c$ . Two features are noteworthy. First, as we move towards larger miscut angle, the steps become less pronounced and no longer strictly parallel with the  $[0001]$  direction. We observe non-parallel step formation. Nevertheless, the overall rms roughness remains extremely low, less than 0.3 nm over  $100\ \mu\text{m}^2$  (not shown), for all three miscut angles and provides a suitable platform for further growth of devices.

## D. CONCLUSIONS

Surface morphology evolution of  $m$ -plane  $[1\bar{1}00]$  GaN homoepitaxially grown by plasma-assisted molecular beam epitaxy is studied on free-standing substrates with small miscut angles towards the  $-c$   $[000\bar{1}]$  and  $+c$   $[0001]$  directions under various gallium to nitrogen ratios at substrate temperatures  $T = 720^\circ\text{C}$ . The resulting surface morphology is shown to depend critically and in a complex fashion on  $\text{Ga}/\text{N}$  ratio,

substrate miscut, and growth temperature. Relatively smooth morphology of  $m$ -plane GaN was obtained in distinct growth regimes including both  $\text{Ga}/\text{N} \leq 0.4$  with  $T = 740^\circ\text{C}$  and  $\text{Ga}/\text{N} \geq 1.2$  with  $T = 720^\circ\text{C}$  on  $-c$  miscut substrates, and  $\text{Ga}/\text{N} \geq 1.0$  with  $T = 740^\circ\text{C}$  on  $+c$  miscut substrates. Importantly, the miscut direction and angle had a dramatic impact on morphology, supporting the notion of strong anisotropy of the gallium adatom diffusion barrier and growth kinetics. These results also highlight that surface morphologies suitable for device fabrication can be obtained for  $m$ -plane substrates miscut toward  $-c$  and  $+c$  once proper growth stoichiometry is determined.

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