Carnegie Mellon University
Research Showcase @ CMU

Department of Physics | Mellon College of Science

2001

Structural Properties of GaN Films Grown by Molecular Beam Epitaxy on Singular and Vicinal 6H-SiC(0001)

C. D. Lee
Carnegie Mellon University

Randall M. Feenstra
Carnegie Mellon University, feenstra@andrew.cmu.edu

O. Shigiltchoff
University of Pittsburgh

R. P. Devaty
University of Pittsburgh

W. J. Choyke
University of Pittsburgh

Follow this and additional works at: http://repository.cmu.edu/physics

Published In
MRS Internet J. Nitride Semi cond. Res., 7, 2, .

This Article is brought to you for free and open access by the Mellon College of Science at Research Showcase @ CMU. It has been accepted for inclusion in Department of Physics by an authorized administrator of Research Showcase @ CMU. For more information, please contact research-showcase@andrew.cmu.edu.

Published in MRS Internet J. Nitride Semi cond. Res. 7, 2 (2001).

Structural Properties of GaN Films Grown by Molecular Beam Epitaxy on Singular and Vicinal 6H-SiC(0001)

C. D. Lee,[1] R. M. Feenstra,[1] O. Shigiltchoff,[2] R. P. Devaty[2] and W. J. Choyke[2]
[1]Department of Physics, Carnegie Mellon University, Pittsburgh, PA 15213
[2]Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, PA 15260

ABSTRACT

Gallium nitride films are grown by plasma-assisted molecular beam epitaxy (MBE) on 6H-SiC(0001) substrates with no miscut and with 3.5° miscuts in both the $[1\bar{1}00]$ and $[11\bar{2}0]$ directions. The hydrogen-etched substrates display straight or chevron shaped steps, respectively, and the same morphology is observed on the GaN films. X-ray rocking curves display substantially reduced width for films on the vicinal substrates compared to singular substrates, for the same Ga/N flux ratio used during growth.

INTRODUCTION

A number of research groups have in recent years studied the growth using molecular beam epitaxy (MBE) of GaN on SiC [1-7]. Hydrogen etching of the substrates is becoming a standard preparation step for removing polishing damage [8]. Most growth has been performed on singular (no intentional miscut, or on-axis) (0001) Si-face substrates, although a few studies have employed vicinal (miscut, or off-axis) substrates. One report in particular, by Xie et al. [6], has reported significantly improved film quality on the vicinal substrates, as discussed in more detail below.

In this work we report on results for GaN film growth by plasma-assisted MBE on singular 6H-SiC(0001) substrates and on vicinal substrates with $[1\bar{1}00]$ and $[11\bar{2}0]$ miscut directions. After H-etching, $\langle 1\bar{1}00\rangle$ oriented steps are observed on all types of substrates, independent of miscut direction or magnitude. Subsequent GaN film growth is found to preserve the general morphology of the substrate. For high Ga/N flux ratios (where smooth films are achieved), the structural properties of the films grown on the vicinal substrates are found to be substantially better than for those films grown on the singular substrates.

EXPERIMENTAL

The 6H polytype of SiC was used for all the studies reported here. The on-axis SiC and the substrates miscut 3.5° towards $[11\bar{2}0]$ were n-type material, purchased commercially from Cree, Inc. The substrates misoriented towards $[1\bar{1}00]$ were cut from a boule of n-type material. Samples were cut at an angle of 3.5±0.5° from the [0001] direction towards $[1\bar{1}00]$ using a goniometer accurate to ±0.5°. X-ray Laue patterns were taken to verify the angle. These samples were polished with 90, 30, 15, 6, 3, 1, 0.5 and finally 0.25 μm diamond paste. The last four stages took about 20-25 hours each. No visible scratches on the resulting surfaces were detected using the naked eye. Hydrogen etching of the SiC was performed at 1600–1700°C and 1 atm pressure to remove polishing damage [8]. Following transfer of the substrates to the MBE

system, Si deposition followed by annealing was done to remove surface oxide and obtain a surface with well-defined structure and stoichiometry, namely, the $\sqrt{3}\times\sqrt{3}$-R30° reconstruction containing 1/3 monolayer (ML) of Si adatoms on the Si-face SiC(0001) surface. This substrate was then taken to the growth temperature and deposition of the GaN was performed. Our GaN films have Ga polarity [9].

GaN films of thickness near 1 μm were grown at temperatures of 730–800°C using various Ga/N flux ratios [10]. As discussed previously, as the Ga flux exceeds some critical value the growth morphology as viewed by reflection high-energy electron diffraction (RHEED) goes through a rough to smooth transition [11]. We define an effective N flux such that the Ga/N flux ratio is unity at this transition point. (In terms of actual fluxes, this effective flux is given by N_i-N_d*+Ga_d* where N_i is the total incident N flux, N_d* is the desorbed N flux at the transition point, and Ga_d* is the desorbed Ga flux at the transition point). Films were characterized using atomic force microscopy (AFM) and x-ray rocking curves, the latter performed in a triple-axis configuration. In most cases the AFM was performed in contact mode, although for the SiC miscut towards $[11\bar{2}0]$ significant surface charging problems were observed and noncontact (tapping) mode was needed to overcome them.

RESULTS & DISCUSSION

Morphology of H-etched SiC substrates

Our 6H-SiC(0001) substrates are prepared by H-etching, as illustrated in Fig. 1. Figures 1(a) and (b) review results for singular substrates [8]: The morphology of as-received substrates [Fig. 1(a)] displays many polishing scratches. Following etching, full-unit-cell height (15 Å) steps, arising from an unintentional miscut of the substrate, are seen on the surface [Fig. 1(b)]. On a larger scale the surface is found to break into low-angle facets, with neighboring facets having a different $\langle 1\bar{1}00\rangle$ step direction and the angle of each facet being less than the overall miscut of the wafer.

Results for a H-etched substrate miscut by 3.5° towards $[1\bar{1}00]$ are shown in Fig. 1(c). In this case the surface is seen to consist of a series of $\approx$ 300 nm wide strips, with normal vector oriented along the miscut direction. These strips are separated by step bunches, typically 3 nm high. In contrast, results for a H-etched substrates miscut by 3.5° towards $[11\bar{2}0]$ are shown in Fig. 1(d). We now observe half-unit-cell height (7.5 Å) steps separated by $\approx$ 12 nm wide terraces. The steps, on average, have normal vector oriented in the miscut direction. However, it is clearly seen in Fig. 1(d) that on a small scale the steps have a different orientation, i.e. the step edges have a zig-zag or chevron morphology. The precise orientation of this small-scale structure of the step edges is difficult to discern from Fig. 1(d), but we believe the normal vectors to be $[01\bar{1}0]$ and $[10\bar{1}0]$ (±30° from $[11\bar{2}0]$) since (i) this is a low energy step orientation for SiC steps [8], and (ii) the same step orientation is seen on GaN films grown on these substrates as shown below.

GaN films on H-etched SiC

Before discussing results for GaN films grown on vicinal substrates, we first briefly review results for growth on singular material as shown in Fig. 2 [7]. For samples grown with Ga/N ratio only slightly greater than unity, the surface is covered with pits [Fig. 2(a)]. The pits are formed with facetted sidewalls and flat-topped (0001) oriented ridges, as illustrated by the linecut in Fig. 2(a). From AFM linescans the sidewalls are found to have an angle of 30° ± 5° relative to the (0001) surface. From facet-derived features in RHEED patterns this angle is found more precisely to be 33° ± 2°. The facets thus correspond to $\{10\bar{1}3\}$ planes, which have an angle of 32.0° relative to the (0001) surface. This result is similar to that of Heying et al. who report $\{10\bar{1}3\}$ or $\{10\bar{1}4\}$ facets [12]. An analysis of facet angles in the AFM image (of the type done e.g. in Ref. [13]) reveals that all six equivalent $\{10\bar{1}3\}$ facets are present in approximately equal amounts on the surface. As the Ga/N ratio is increased to about 1.1 fewer pits appear on the surface, and those which are present tend to merge into trenches which separate plateaus of atomically flat morphology [Fig. 2(b)]. For Ga/N ratios near 1.3, isolated pits with typical separation of about 1 μm are seen in the morphology [Fig. 2(c)]. TEM studies reveal that these pits are associated with dislocations intersecting the surface [7,14]; some type of preferential film decomposition or reduced growth rate apparently occurs at these points. Finally, for growth under very Ga-rich conditions, Ga/N ratio above about 1.5, the morphology is flat with only monatomic steps revealed in AFM [Fig. 2(d)]. On a large scale, the GaN film morphology follows that of the SiC, with low-angle facets on the surface [10]. Gallium droplets are observed on these surfaces grown with high Ga/N ratios, in agreement with the observations of Heying et al. [12].

As shown in Fig. 2, the roughness of films grown with Ga/N flux ratios near unity is significantly greater than for those with higher Ga/N ratios. Rms roughness values are 35, 20, 4, and 3 nm for the images shown in Figs. 2(a)–(d) respectively. However, for these singular substrates, we find that even though the morphology at low Ga/N flux ratio is worse (i.e. rougher) than that at high Ga/N flux ratio, the structural properties of the low-flux-ratio films is much improved. In particular, the FWHM of asymmetric $(11\bar{2}0)$ x-ray rocking curves are about 2× less for films with flux ratio near unity compared to that of films with flux ratio of 1.3 – 1.5 [14]. TEM reveals an order-of-magnitude reduction in dislocation density for the low-flux-ratio films. The mechanism for this improvement in film quality is found to be the tendency of dislocations to cluster near the topographic minima, where their probability of annihilation is increased due to their closer proximity [14].

The surface morphology of our GaN films grown on vicinal SiC is pictured in Fig. 3. These results can be seen to reflect the morphology of the underlying SiC substrates. The films grown on a substrate miscut towards $[1\bar{1}00]$ display similar size terraces (with $[1\bar{1}00]$ normal vectors) as the substrate, although these GaN strips are not completely continuous along their length [Fig. 3(a)]. The films grown on the substrate miscut towards $[11\bar{2}0]$ display steps with average orientation having $[11\bar{2}0]$ normal, but with $[01\bar{1}0]$ and $[10\bar{1}0]$ oriented step edges seen on a smaller scale [Fig. 3(b)]. Note however that these features are much larger than the corresponding ones seen in Fig. 1(d), indicating significant step bunching and terrace growth during the GaN film growth. Rms roughness is 3 nm for both Figs. 3(a) and (b).

X-ray results for our films are listed in Table I. We report FWHM of rocking curves acquired in both symmetric (0002) and asymmetric $(10\bar{1}2)$ configurations. The former reflects the screw dislocation density in the material and the latter reflects a combination of screw and edge dislocation densities. Actually, since in our material the edge dislocation density is 1–2 orders of magnitude larger than the screw dislocation density (and there are very few dislocations of mixed character), the $(10\bar{1}2)$ width reflects primarily the edge dislocation density. It is this $(10\bar{1}2)$ width which is the important parameter in Table I for distinguishing the various films.

The first two entries in Table I list films grown on on-axis SiC with Ga/N ratio near unity. These films have $(10\bar{1}2)$ widths of 400–500 arcsec. The surface morphology for those films is somewhat rough, as described above. The next three entries in Table I list films grown with higher flux ratios. The $(10\bar{1}2)$ widths are correspondingly larger, for the reason discussed in the previous section. The final four entries in Table I give results for growth on the vicinal substrates, for Ga/N ratios of 1.5. We find that the $(10\bar{1}2)$ widths for these films are significantly less than those for the films grown on singular substrates using the same flux ratios. In fact, the results for the films grown on vicinal substrates are comparable to those obtained for our best (i.e. low-flux-ratio) growth on the singular substrates, whereas the morphology of the growth on the vicinal substrates as seen in Fig. 3 is improved compared to that on those low-flux-ratio films on the singular substrates.

For several of the films grown on miscut substrates listed in Table I we measured the x-ray rocking curves for two inequivalent orientations of the vicinal substrates relative to the incident beam. Not surprisingly some variation in the width is seen, implying that the mechanism for the dislocation reduction in these films does indeed involve some specific aspect of their vicinality. TEM measurements have not been performed to date, so no further information in available concerning the mechanism for dislocation reduction in these films. Recently Xie et al. [6] have reported results for dislocation reduction in GaN films grown on vicinal SiC. In their case the initial SiC substrates were *not* H-etched, so that their screw dislocation density for growth on singular substrates was very high. This value was greatly reduced for the growth on vicinal substrates, which they interpret as being due to nucleation, growth and coalescence of GaN islands at step edges rather than on terraces [6]. They also observed a reduction in edge dislocation density, similar to that reported here, although no specific mechanism was suggested for that reduction. Their results are thus consistent with those reported here, although their model for the reduction in screw dislocation density is not relevant for our situation of H-etched substrates in which our screw dislocation density is low for both singular and vicinal substrates.

SUMMARY

In summary, we have grown GaN films by plasma-assisted MBE on singular 6H-SiC(0001) substrates and on vicinal substrates with $[1\bar{1}00]$ and $[11\bar{2}0]$ miscut directions. X-ray rocking curves display substantially reduced width for films on the vicinal substrates compared to singular substrates, for the same Ga/N flux ratio used during growth. Films grown on vicinal substrates at high Ga/N flux ratio have rocking curve widths comparable to those obtained for our best growth on singular substrates (i.e. using low Ga/N ratio, near unity), but the roughness

for those films on vicinal substrates is about an order-of-magnitude lower than that of the low-flux-ratio films on singular substrates.

ACKNOWLEDGMENTS

This work was supported the Office of Naval Research (grant N00014-96-1-0214, monitored by C. Wood).

REFERENCES

[1] V. Ramachandran, A. R. Smith, R. M. Feenstra, and D. W. Greve, *Temperature Dependence of Molecular Beam Epitaxy of GaN on SiC(0001)*, J. Vac. Sci. Technol. A **17**, 1289 (1999).

[2] V. M. Torres, J. L. Edwards, B. J. Wilkens, D. J. Smith, R. B. Doak, and I. S. T. Tsong, *Influence of 6H–SiC(0001) substrate surface morphology on the growth of AlN epitaxial layers*, Appl. Phys. Lett. **74**, 985 (1999).

[3] Q. Z. Xue, Q. K. Xue, Y. Hasegawa, I. S. T. Tsong, and T. Sakurai, *Two-step preparation of 6H–SiC(0001) surface for epitaxial growth of GaN thin film*, Appl. Phys. Lett. **74**, 2468 (1999).

[4] R. Lantier, A. Rizzi, D. Guggi, H. Lüth, B. Neubauer, D. Gerthsen, S. Frabboni, G. Colì, and R. Cingolani, *Influence of the First Preparation Steps on the Properties of GaN Layers Grown on 6H-SiC by MBE*, MRS Internet J. Nitride Semicond. Res. **4S1**, G3.50 (1999).

[5] O. Brandt, R. Muralidharan, P. Waltereit, A. Thamm, A. Trampert, H. von Kiedrowski, and K. H. Ploog, *Critical issues for the growth of high-quality (Al,Ga)N/GaN and GaN/(In,Ga)N heterostructures on SiC(0001) by molecular-beam epitaxy*, Appl. Phys. Lett. **75**, 4019 (1999).

[6] M. H. Xie, L. X. Zheng, S. H. Cheung, Y. F. Ng, H. Wu, S. Y. Tong, and N. Ohtani, *Reduction of threading defects in GaN grown on vicinal SiC(0001) by molecular-beam epitaxy*, Appl. Phys. Lett. **77**, 1105 (2000).

[7] C. D. Lee, V. Ramachandran, A. Sagar, R. M. Feenstra, D. W. Greve, W. L. Sarney and L. Salamanca-Riba, D. C. Look, S. Bai, W. J. Choyke and R. P. Devaty, *Properties of GaN epitaxial layers grown on 6H-SiC(0001) by plasma-assisted molecular beam epitaxy*, J. Electron. Mat. **30**, 162 (2001).

[8] V. Ramachandran, M. F. Brady, A. R. Smith, R. M. Feenstra, and D. W. Greve, *Preparation of Atomically Flat Surfaces on Silicon Carbide using Hydrogen Etching*, J. Electron. Mater. **27**, 308 (1998).

[9] A. R. Smith, R. M. Feenstra, D. W. Greve, M.-S. Shin, M. Skowronski, J. Neugebauer, and J. Northrup, *Determination of Wurtzite GaN Lattice Polarity Based on Surface Reconstruction*, Appl. Phys. Lett. **72**, 2114 (1998).

[10] C. D. Lee, A. Sagar, R. M. Feenstra, W. L. Sarney, L. Salamanca-Riba, and J. W. P. Hsu, *Growth of GaN on SiC(0001) by Molecular Beam Epitaxy*, Phys. Stat. Sol. (a) **188**, 595 (2001).

[11] A. R. Smith, V. Ramachandran, R. M. Feenstra, D. W. Greve, A. Ptak, T. H. Myers, W. Sarney, L. Salamanca-Riba, M.-S. Shin, and M. Skowronski, *Surface Reconstructions during Molecular Beam Epitaxy of GaN(0001)*, MRS Internet J. Nitride Semicond. Res. **3**, 12 (1998).

[12] B. Heying, R. Averbeck, L. F. Chen, E. Haus, H. Riechert, and J. S. Speck, *Control of GaN surface morphologies using plasma-assisted molecular beam epitaxy*, J. Appl. Phys. **88**, 1855 (2000).

[13] M. A. Lutz, R. M. Feenstra, P. M. Mooney, J. Tersoff, and J. O. Chu, *Facet Formation in Strained SiGe Films*, Surf. Sci. Lett. **316**, L1075 (1994).

[14] C. D. Lee, A. Sagar, R. M. Feenstra, C. K. Inoki, T. S. Kuan, W. L. Sarney and L. Salamanca-Riba, *Role of Ga-flux in dislocation reduction in GaN films grown on SiC(0001)*, Appl. Phys. Lett. **79**, 3248 (2001).

Table I. X-ray rocking curve FWHM values for GaN films of specified miscut, growth temperature, Ga/N ratio, and thickness. Two values for the FWHM of films on miscut substrates refer to different sample orientations.

Miscut	T (°C)	Ga/N ratio	thickness (μm)	FWHM X-ray rocking curves (arcsec)	
				(0 0 0 2)	(1 0 –1 2)
On-axis	730	1.1	1.0	121	500
On-axis	780	1.1	2.0	119	410
On-axis	750	1.4	1.3	128	927
On-axis	750	1.7	1.0	57	960
On-axis	730	1.9	1.0	218	960
3.5º towards <1 1 –2 0>	750	1.5	1.0	68	520
3.5º towards <1 1 –2 0>	750	1.5	1.0	135	442/575
3.5º towards <1 1 –2 0>	750	1.5	1.2	115	490/634
3.5º towards <1 –1 0 0>	750	1.5	1.0	93	612/418

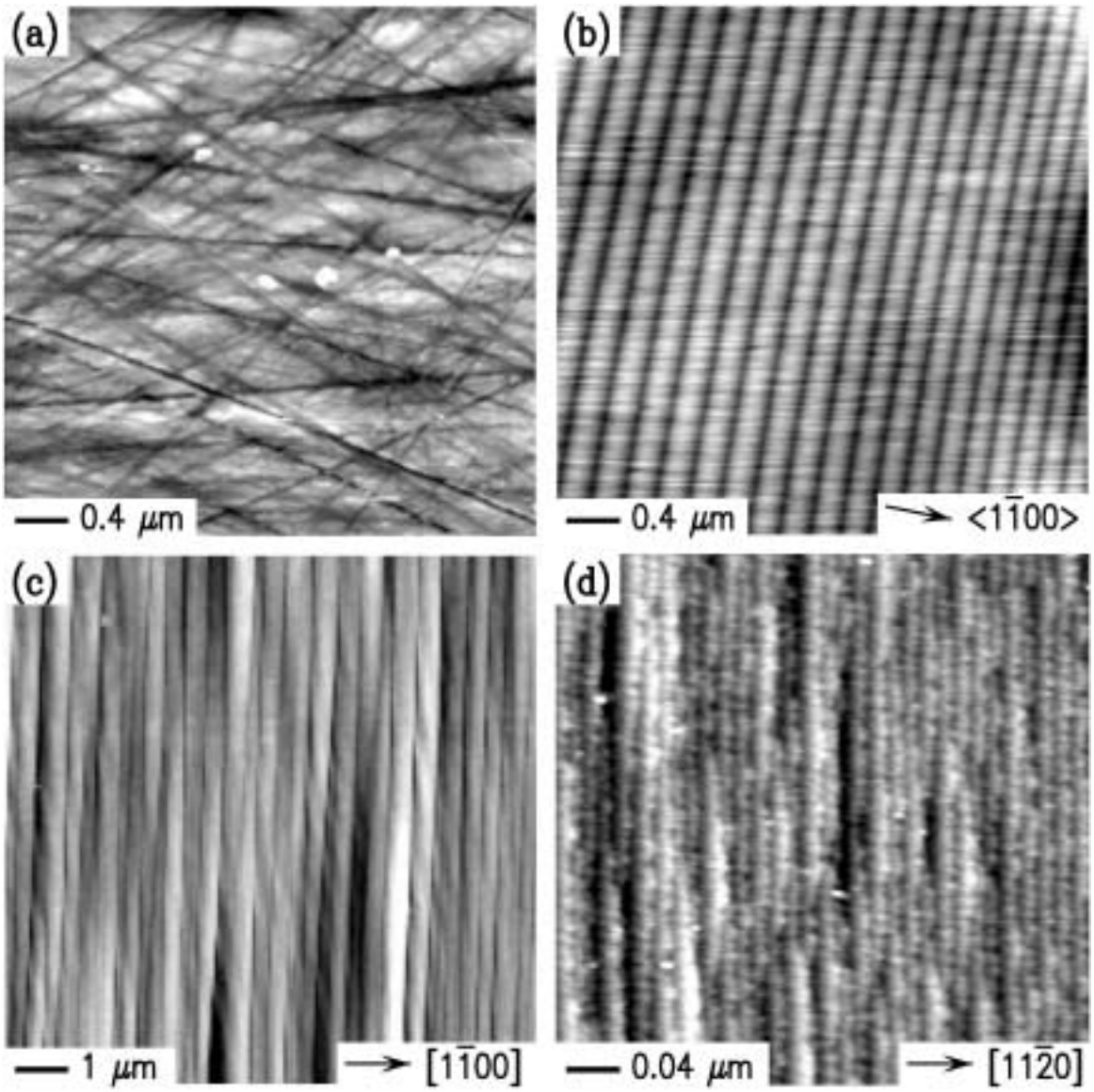

Figure 1. AFM images of 6H-SiC(0001) surfaces: (a) on-axis, as-received; (b) on-axis, H-etched; (c) off-axis, 3.5º towards [1$\bar{1}$00], H-etched; (d) off-axis, 3.5º towards [11$\bar{2}$0], H-etched. Gray scale ranges are (a) 10 nm, (b) 0.7 nm, (c) 7 nm, and (d) 1 nm. Note the varying lateral scales.

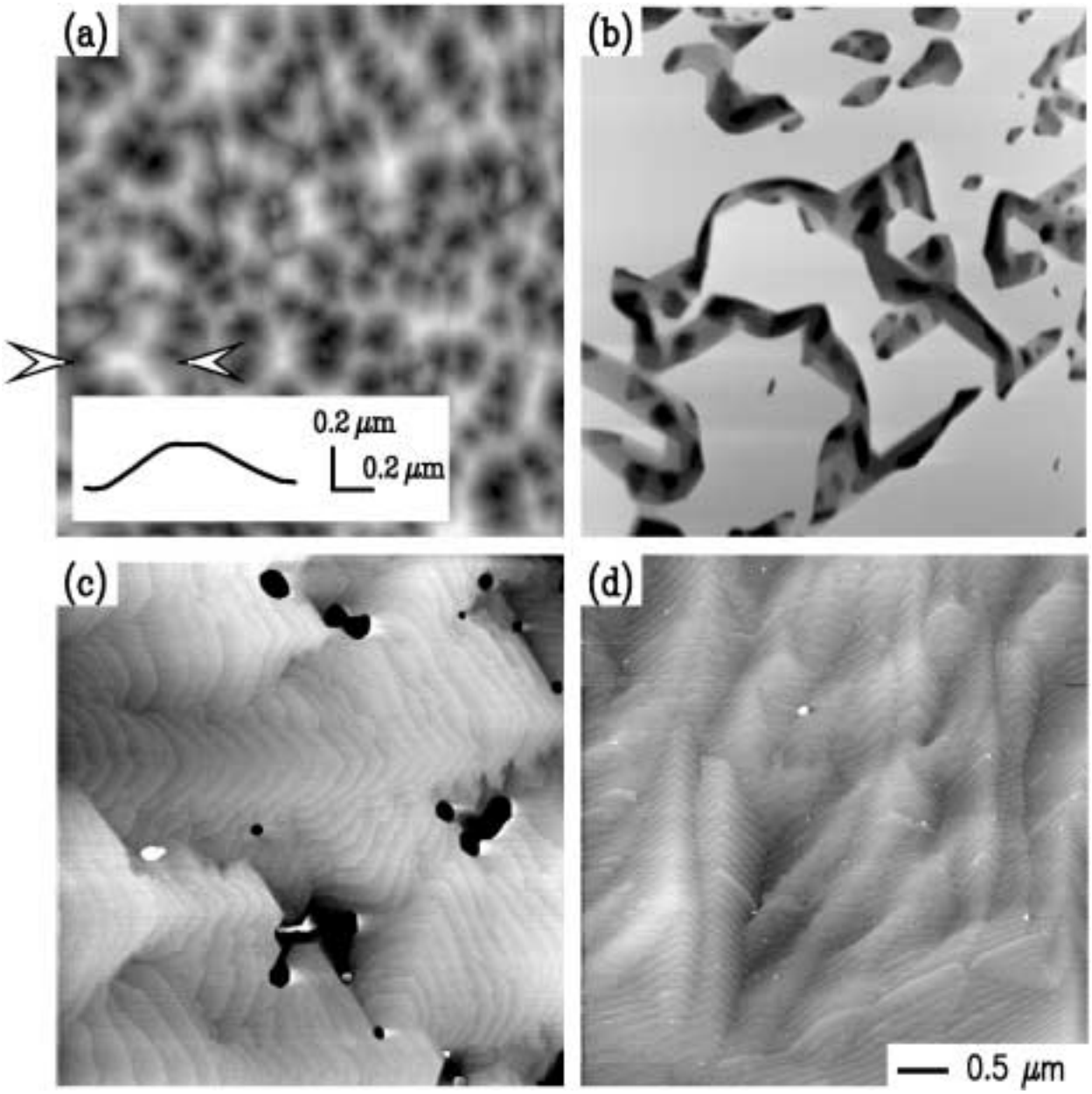

Figure 2. AFM images of GaN(0001) films grown on H-etched 6H-SiC, with Ga/N flux ratios of (a) 1.05, (b) 1.1, (c) 1.3, and (c) 1.6. The gray scale ranges are 210 nm, 86 nm, 5 nm, and 5 nm for (a)–(d) respectively. The film pictured in (a) was grown at 800ºC and the other films were grown at 750ºC. In (a), a line cut taken between the arrows is shown in the inset.

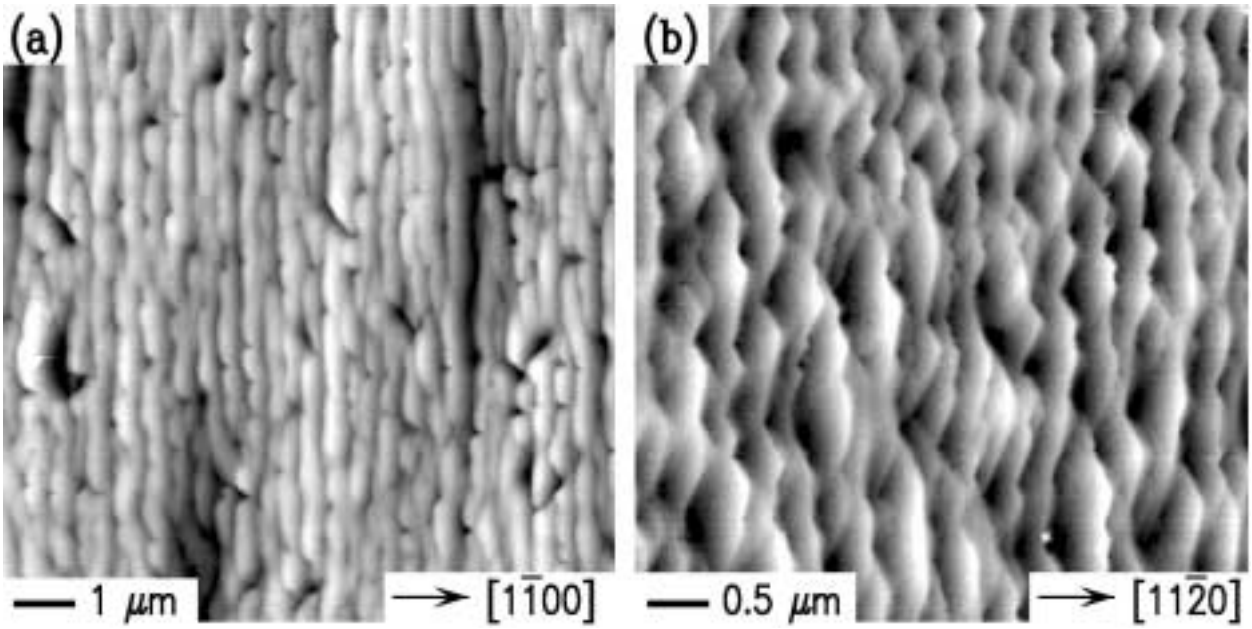

Figure 3. AFM images of GaN(0001) films grown on H-etched miscut SiC, with Ga/N ratio of 1.5 for both films. Substrates used are: (a) off-axis, 3.5º towards [1$\bar{1}$00], and (b) off-axis, 3.5º towards [11$\bar{2}$0]. Gray scale ranges are (a) 15 nm and (b) 12 nm.