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High-quality GaN on intentionally roughened c-sapphire

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Abstract. GaN films were grown using metal-organic chemical vapor deposition (MOCVD) on intentionally roughened c-sapphire in order to study the effect of the substrate roughness. Controlled substrate damage was achieved by exposing the substrate surfaces to controlled amounts of trimethylgallium followed by exposure to hydrogen at elevated temperature. The bulk film quality was studied by high-resolution X-ray diffraction and the surface topography was examined by atomic force microscopy. It was observed that mild substrate roughness (surface pit depth up to 30 nm) does not have a significant effect on the bulk film quality nor the GaN surface topography, and that recovery of the surface roughness is observed in very early stages of film growth. Cross sectional transmission electron microscopy confirmed that MOCVD GaN grown on inclined sidewalls etched in c-sapphire maintained the $\{0001\}_{\text{sapphire}} \parallel \{0001\}_{\text{GaN}}$ epitaxial relationship despite the substrate miscut of *ca.* 15° with respect to the *c*-axis.

PACS. 68.35.Ct Interface structure and roughness – 68.55.Ac Nucleation and growth: microscopic aspects

Although many efforts are spent on the preparation of high-quality sapphire substrates for GaN epitaxy, the correlation between the substrate surface roughness and the film quality of GaN grown by metalorganic chemical vapor deposition (MOCVD) has not been directly established. Cui and coworkers evaluated the optimal thermal treatment parameters for c-sapphire substrates for GaN growth by molecular beam epitaxy (MBE) using triple axis X-ray diffraction (XRD) and atomic force microscopy (AFM) [1]. When an optimized thermal treatment of the sapphire substrates was not carried out, these authors reported a dramatic increase by sevenfold and fivefold in the full width at half maximum (FWHM) of X-ray rocking curves obtained for symmetric and asymmetric Bragg reflections, respectively. Parillaud *et al.* studied the effect of sapphire substrate misalignment on GaN epilayers grown by MOCVD and hydride vapor phase epitaxy using AFM, XRD and scanning electron microscopy (SEM). The FWHM broadening of the (0002) X-ray rocking curves in GaN films grown on c-sapphire that was misaligned by 2° towards the *a*-direction was found to be smaller compared to the FWHM of GaN films grown on exact *c*-substrates and on substrates misaligned by 2° towards the *m*-direction [2].

The partial dissociation of sapphire substrates under the physical and chemical conditions present in MBE

growth of GaN has been previously demonstrated using secondary ion mass spectrometry [3–5]. In previous experiments, we have exposed c-sapphire substrates to controlled amounts of substances that are normally present in the MOCVD/GaN environment [*e.g.*, trimethylgallium (TMGa), that resulted in deposition of submicron droplets of metallic Ga on the sapphire surface] in realistic experimental conditions. AFM and lateral force microscopy showed that as in MBE, in the MOCVD/GaN environment c-sapphire substrates tend to undergo partial dissociation which appears as etch pits on the sapphire surface [6]. Hence, this approach, termed “controlled contamination” (CC), can be used for preparing c-sapphire substrates with etch pits of controlled depths, *i.e.*, for controlling the surface roughness of these substrates. Preliminary AFM experiments carried out with an excess of metallic Ga present on the surface suggested that the surface morphology of GaN epilayers grown on CC-roughened sapphire is insensitive to surface roughness [7]. In the present work, we have used XRD, AFM and transmission electron microscopy (TEM) for evaluating the effect of substrate roughness on the quality of MOCVD-grown GaN epilayers.

AFM images of CC-treated sapphire substrates that were exposed to a TMGa flow for 10 s and 100 s are shown in Figures 1a, b, respectively (full details on the CC experimental conditions are given in Ref. 6). These images show that sapphire surfaces are indeed reactive

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Fig. 1. AFM images of controlled contamination (cc)-treated sapphire substrates, and the corresponding GaN epilayer surface topographies. (a) Sapphire substrate after a 10 s TMGa exposure. Height (Z) range: 2.4 nm. (b) Sapphire substrate after a 100 s TMGa exposure. Z-range: 12.0 nm. (c) 1 μm thick GaN epilayer deposited on (a). Z-range: 3.0 nm. (d) 1 μm thick GaN epilayer deposited on (b). Z-range: 3.0 nm. All magnification bars correspond to 500 nm.

under these experimental conditions and demonstrate the potential use of CC for intentionally etching pit defects in the surfaces that result in substrate surfaces with controlled roughness. The average pit depth seen in Figure 1a is ~ 1 nm, while the average pit depth in Figure 1b is ~ 5 nm. The white features seen in Figure 1b were identified by Auger electron spectroscopy (AES) as particles of unreacted metallic Ga that remained on the surface following the CC process. The corresponding AFM images of 1 μm thick GaN films grown by MOCVD on the CC-treated substrates are shown in Figures 1c, d. The images show crystallographic steps with heights corresponding to one half of the wurtzite GaN *c*-lattice parameter. The dark point depressions seen in the image correspond to threading dislocations (TDs) intersecting the GaN surface. The screw character of many of the TDs is evident as points in which at least one crystallographic step is terminated. Also seen are arrays of depressions which most probably consist of edge TDs (Fig. 1d), in agreement with TEM studies reporting TDs with predominantly mixed character with $b = 1/3\langle 1\bar{1}23 \rangle$, pure edge character with $b = 1/3\langle 1\bar{1}20 \rangle$, and very few of pure screw character with $b = \langle 0001 \rangle$ in as-grown device quality MOCVD GaN [8–11]. It is important to note that although grown on substrate surfaces of increasing roughness, the images in Figures 1c and 1d are very similar, both indicative of high-quality, fully coalesced GaN sur-

Fig. 2. High-resolution XRD rocking curve peak broadening at FWHM obtained for sapphire (006), GaN (002) and GaN (101) reflections, plotted *vs.* etch pit depths as observed with AFM.

faces that are not different from surfaces observed on GaN grown on undamaged sapphire [12]. The bulk quality of GaN films grown by MOCVD on CC-treated *c*-sapphire was studied using triple-axis XRD. The FWHM broadening of the wurtzite (0002)_{GaN} and (10 $\bar{1}$ 1)_{GaN} rocking curves together with that of (0006)_{sapphire}, as a function of the etch pit depths obtained as a result of CC exposure to TMGa, is shown in Figure 2. Interestingly, for the substrate surface roughness range examined, ~ 0 to 30 nm, no notable differences were observed in the FWHM broadening of the (10 $\bar{1}$ 1)_{GaN} (off-axis) reflections, while a small increase of up to 20% was obtained for the broadening of the (0002)_{GaN} (on-axis) reflections. Experiments were repeated also under CC conditions that ensured that all the Ga metal deposited on the surfaces had reacted in full. This was achieved by shorter TMGa exposures and longer exposures to H₂ at 1050 $^{\circ}\text{C}$, until no Ga particles were seen by AFM. In the absence of excess Ga, the (0002)_{GaN} broadening remained essentially unaffected by the surface roughness, while a slight increase of up to 13% was obtained for the (10 $\bar{1}$ 1)_{GaN} reflections.

To further investigate the surprisingly subtle effect of the substrate surface roughness on the bulk and surface quality of MOCVD-grown GaN films, an interrupted growth series was carried out for monitoring the film morphology in the initial stages of growth on damaged *c*-sapphire. A series of AFM images representing these stages is shown in Figure 3. Figure 3a shows sapphire treated with a 1000 s exposure to TMGa, corresponding to pit depths of several tens of nm. Also here, the white features in the image were correlated with metallic Ga using AES. Figure 3b shows CC-treated sapphire as in Figure 3a, following the deposition of a 20 nm thick GaN nucleation layer deposited at 525 $^{\circ}\text{C}$. The next stage in the MOCVD growth of GaN included growth of the main GaN

Fig. 3. AFM images showing the morphology evolution sequence of GaN grown on cc-treated sapphire substrates. (a) Sapphire substrate after a 1000 s TMGa exposure. Bar = $1.0\ \mu\text{m}$; Z-range: 60 nm. (b) Same as (a), followed by a 20 nm thick GaN buffer layer deposited at $525\ ^\circ\text{C}$. Bar = $1.0\ \mu\text{m}$; Z-range: 60 nm. (c) Same as (b), followed by a controlled temperature ramp to $1050\ ^\circ\text{C}$. Bar = $1.0\ \mu\text{m}$; Z-range: 36 nm. (d) same as (c), with 10 nm nominal thickness of GaN deposited at $1050\ ^\circ\text{C}$. Bar = $0.5\ \mu\text{m}$; Z-range: 60 nm.

layer on the nucleation layer at $1050\ ^\circ\text{C}$. Figure 3c shows a nucleation layer sample as in Figure 3b, after a temperature ramp to $1050\ ^\circ\text{C}$ followed by immediate cooldown. As a result of the higher temperature, significant coarsening of the nucleation layer is seen, together with planarization of the sample surface and disappearance of the CC etch pits. Hence, at this point, full recovery of the surface roughness is observed, which is followed by normal film growth *via* growth of truncated islands [also called high-temperature (HT) islands] on top of the nucleation layer, as seen in Figure 3d. Note that despite the rapid recovery of interfacial roughness observed, it is nonetheless necessary to grow GaN templates to a thickness of several microns to allow for TD density reduction by means of dislocation reactions.

Cross-sectional TEM (XTEM) was used to study GaN films which were grown by MOCVD on patterned surfaces [13,14], and specifically relevant to this work, to monitor the film morphology in regions grown on inclined (miscut) sapphire surfaces. Samples were prepared by patterning parallel linear trenches $0.5\text{--}0.6\ \mu\text{m}$ deep and $2\text{--}4\ \mu\text{m}$ wide in the *c*-sapphire substrates using reactive ion etching (RIE) along the $\langle 1\bar{1}00 \rangle$ and $\langle 11\bar{2}0 \rangle$ directions. Subsequently, a $2\ \mu\text{m}$ thick GaN layer was grown on the patterned sapphire using a thin GaN nucleation layer [13,14]. An SEM image of a $0.7\ \mu\text{m}$ thick GaN film

Fig. 4. Plan view SEM image showing GaN on patterned sapphire. Pattern mesa width is $50\ \mu\text{m}$.

grown on patterned sapphire is shown in Figure 4. Due to the great difficulty of etching vertical walls in sapphire using RIE, the trenches exhibited curved rather than vertical walls. The cross-sections shown in Figure 5 were prepared along the $[1\bar{1}00]$ direction, with images taken close to the trench edges, where the angle between the actual sapphire surface and the $(0001)_{\text{sapphire}}$ planes was ~ 15 degrees. Figure 5a shows an XTEM image taken with $\mathbf{g} = 01\bar{1}0$. Under these particular diffraction conditions, stacking faults parallel to the $(0001)_{\text{sapphire}}$ are observed, indicating stacking of the $(0001)_{\text{GaN}}$ planes along the sapphire *c*-axis despite the large miscut of the local sapphire substrate surface. Further demonstration of the robust growth of GaN on sapphire is shown in Figure 5b, which was taken from the opposite side of the trench¹ from Figure 5a with $(\mathbf{g} = 11\bar{2}0)$. The contrast of the truncated features seen on top of the substrate surface under these diffraction conditions appears to be due to either stacking faults or threading dislocations along the $\{1\bar{1}01\}_{\text{GaN}}$ planes. We speculate that these features are associated with the HT islands which are formed during the early stages of MOCVD GaN growth [8]. In order to accommodate the inclined sapphire substrate surface and yet grow in a nearly perfect $\{0001\}_{\text{GaN}} \parallel \{0001\}_{\text{sapphire}}$ orientation relationship, these HT islands apparently adapt an asymmetrical shape, with longer facets towards the lower (left) side and shorter facets towards the upper (right) side of the substrate surface. Interestingly, the GaN grown in the trench appeared to be rotated by about one degree about the $[11\bar{2}0]$ axis with respect to the GaN grown on the mesa [13]. Furthermore, it is interesting to note that this crystallographic tilt of the sidewalls is comparable in magnitude to the wing tilt observed in GaN grown by lateral epitaxial overgrowth [15,16].

In conclusion, we have used the CC approach for introducing controlled roughness in *c*-sapphire substrates in order to study its effect on the morphology evolution of MOCVD GaN epilayers. The effect of substrate roughness up to several tens of nm was found to be very

¹ The asymmetry of the microstructures observed on opposite sides of the same trench can be explained by the threefold symmetry of the corundum structure.

Fig. 5. Two-beam TEM images of MOCVD GaN on inclined sapphire surfaces. Cross-sections were prepared along the $\langle 1\bar{1}00 \rangle$ direction. (a) $g = 01\bar{1}0$ diffraction conditions. (b) $g = 11\bar{2}0$ diffraction conditions.

small also without excess of highly mobile Ga atoms on the surface [17]. Hence, MOCVD GaN growth appears to be extremely robust with respect to substrate surface roughness. The recovery of the surface roughness apparently occurs immediately after the nucleation layer is deposited, upon ramping the temperature to 1050 °C for growth of the primary (HT) layer. XTEM of GaN on patterned sapphire confirmed that MOCVD GaN can remarkably recover from substrate roughness, and that the $\{0001\}_{\text{sapphire}} \parallel \{0001\}_{\text{GaN}}$ epitaxial relationship is still observed in material grown on local substrate miscut of *ca.* 15° with respect to the *c*-axis.

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References

1. J. Cui, A. Sun, M. Reshchikov, F. Yun, A. Baski, H. Morkoc, MRS Internet J. Nitride Semicond. Res. **5**, 7 (2000)
2. O. Parillaud, V. Wagner, H.-J. Bühlmann, F. Lelarge, M. Illegems, MRS Internet J. Nitride Semicond. Res. **5S1**, W3.13 (2000)
3. S. Mitha, R. Clark-Phelps, J.W. Erickson, Y. Gao, W. Kim, H. Morkoc, Mat. Res. Soc. Symp. Proc. **468**, 183 (1997)
4. G. Popovici, W. Kim, A. Botchkarev, H. Tang, J. Solomon, H. Morkoc, Mat. Res. Soc. Symp. Proc. **468**, 3 (1997)
5. J.E. Van Nostrand, J. Solomon, A. Saxler, Q.-H. Xie, D.C. Reynolds, D.C. Look, J. Appl. Phys. **87**, 8766 (2000)
6. Y. Golan, P. Fini, S.P. DenBaars, J.S. Speck, Jpn. J. Appl. Phys. Part 1 **37**, 4695 (1998)
7. Y. Golan, P. Fini, S.P. DenBaars, J.S. Speck, Mat. Res. Soc. Symp. Proc. **482**, 57 (1998)
8. X.H. Wu, P. Fini, E.J. Tarsa, B. Heying, S. Keller, U.K. Mishra, S.P. DenBaars, J.S. Speck, J. Cryst. Growth **189**, 231 (1998)
9. S.D. Lester, F.A. Ponce, M.G. Craford, D.A. Steigerwald, Appl. Phys. Lett. **66**, 1249 (1995)
10. W. Qian, M. Skowronski, M. DeGraef, K. Doverspike, L.B. Rowland, D.K. Gaskill, Appl. Phys. Lett. **66**, 1252 (1995)
11. N.-E. Lee, R.C. Powell, Y.-W. Kim, J.E. Greene, J. Vac. Sci. Technol. A **13**, 2293 (1995)
12. X.W. Wu, L.M. Brown, D. Kapolnek, S. Keller, B.P. Keller, S.P. DenBaars, J.S. Speck, J. Appl. Phys. **80**, 3228 (1996)
13. F. Wu, S. Zamir, B. Meyler, J. Salzman, Y. Golan, J. Electron. Mater. **32**, 23 (2003)
14. S. Zamir, B. Meyler, J. Salzman, F. Wu, Y. Golan, J. Appl. Phys. **91**, 1191 (2002)
15. P. Fini, H. Marchand, J.P. Ibbetson, S. Keller, S.P. DenBaars, U.K. Mishra, J.S. Speck, J. Cryst. Growth **209**, 581 (2000)
16. P. Fini, A. Munkholm, C. Thompson, G.B. Stephenson, J.A. Easterman, M.V. Ramana Murty, O. Auciello, L. Zhao, S.P. DenBaars, J.S. Speck, Appl. Phys. Lett. **76**, 3898 (2000)
17. J. Neugebauer, Phys. Stat. Sol. B **227**, 93 (2001)