Optimized structural properties of wurtzite GaN on SiC(0001) grown by molecular beam epitaxy

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Abstract

We have investigated optimal conditions for molecular beam epitaxial growth of high quality GaN on 6H-SiC(0001) substrates. The quality of these films is reflected in both the narrow x-ray peakwidths as well as the excellent surface morphology. In this work, it is shown that increasing growth temperature leads to an improvement in bulk quality and lower x-ray peakwidth for both symmetric and asymmetric reflections. We also note a marked improvement in surface morphology, from a columnar appearance to a 2-D surface, under extremely Ga-rich growth conditions.

I INTRODUCTION

Gallium nitride (GaN) is a wide bandgap semiconductor with direct bandgap which makes it attractive both as a short wavelength light emitter and a high temperature electronic material. There are no easily available, stable, lattice-matched substrates for this material for device development and so heteroepitaxy has been performed on sapphire (mismatch of about 14 %) and silicon carbide (SiC, mismatch of 3.4 %) [1]. It is expected that SiC with a smaller mismatch would lead to heteroepitaxial GaN of higher quality than sapphire, but several factors other than mismatch are believed to be reasons for GaN films on SiC not being significantly better [2]. Nevertheless, heteroepitaxy of GaN on SiC has continued to be studied and continual improvements are being made [3].

In this work, we have grown GaN on Si-polar SiC(0001) using molecular beam epitaxy (MBE). Films have been characterized using high resolution x-ray diffraction (HRXRD) and atomic force microscopy (AFM) for different growth temperatures and surface Ga fluxes. Based on our measurements on these films we observe that the bulk defect density of our films decreases with increasing growth temperature. Also, the surface morphology is seen to improve markedly with increasing surface Ga flux. From these studies, we conclude that high quality GaN heteroepitaxy on 6H-SiC by MBE is possible for high growth temperatures and highly Ga-rich conditions.

II EXPERIMENT

Si-face 6H-SiC(0001) surfaces for our experiments were prepared by removal of polish damage from the wafers using hydrogen etching [4]. This technique is known to produce large, flat areas

on the SiC wafer minimizing the probability of defects originating in the scratches arising from the polishing process. After the *ex-situ* etch, the substrates are introduced into ultra high vacuum (UHV, pressure less than 10^{-10} Torr), the samples are outgassed at about 800° C for 30 minutes. Prior to desorption of the native oxide layer, Si is deposited on the substrate using an electron beam source. This replenishes any Si that may be lost by the surface during the oxide removal step. Oxide desorption is done by annealing the substrate at about 1000° C till a 3×1 reflection high energy electron diffraction (RHEED) pattern (indicative of a $\sqrt{3}\times\sqrt{3}$-R30° surface reconstruction) is obtained.

GaN films of upto 600 nm thickness were grown by MBE on this substrate using a Ga effusion cell and an RF-plasma nitrogen source. The growth was a single-step process with no nucleation layer growth and the films were monitored *in situ* by reflection high energy electron diffraction (RHEED). The substrate temperature was varied between 550° C and 750° C for different growth runs. Experiments were also performed at a constant substrate temperature of 725° C with varying Ga flux to note the effect of Ga concentration during growth. The growth regime was always kept Ga-rich relative to the N concentration as indicated by the streaky RHEED pattern during growth [5]. Characterization *ex situ* was done by AFM, HRXRD, and cross-sectional transmission electron microscopy (XTEM) measurements on the samples.

III RESULTS AND DISCUSSION

In Fig. 1, we show a HRXRD radial (ω-2θ) scan for a film grown at 700° C for one hour under extremely Ga-rich conditions. In Ref. [6] we have reported morphology and x-ray studies for a similar film, where the surface morphology is seen to be clearly 2-D. This x-ray diffractogram clearly shows several thickness fringes corresponding to a thickness of 2000 Å, which matches very well with our expectations of the film thickness based on our knowledge of the growth rate in our chamber, 2000 Å per hour. Not only do these fringes allow us to confirm our growth rate measurements, but also they indicate that these films are of very high quality. The presence of thickness fringes shows that the coherence length of the crystal is equal to its thickness, suggesting that the average spacing between defects is at least this distance, and it also indicates that the film surface is fairly flat.

Figure 2 shows HRXRD full width at half maximum (FWHM) data for a sequence of films, each grown at a different temperature, in the range from 550° C to 750° C for 3 hours. In Fig. 2(a), we show FWHM of symmetric triple crystal radial (ω-2θ) scans obtained from these films. We see that the FWHM decreases with increasing growth temperature, a fact that was also noted in Ref. [6], although there the FWHM of the x-ray data was close to that expected from the thickness of the films. We find that the FWHM of the symmetric peak is as low as 30 arcsecs for films grown at 700° C. This value is considerably smaller than that reported in some early GaN/SiC MBE growth studies [7,8], and is quite close to that reported in two recent studies [3,9]. Figure 2(b) shows FWHM data for the $(11\bar{2}4)$ and $(\bar{2}024)$ asymmetric peaks from the same films obtained in double crystal rocking curves(ω scans). (The low intensity of the reflections made it impossible to introduce the analyzer crystal to perform triple crystal measurements). We see that the trend of FWHM with increasing growth temperature is similar to that of the symmetric peak. However, the peak width is much larger for the asymmetric reflections. This may arise from a large *total* dislocation density. The symmetric peak measures the density of dislocations which distort the (0001) planes while the asymmetric peaks are a better measure of total dislocation densities. Our observa-

tions imply that dislocations seen in the symmetric reflection make up a small fraction of the total dislocation density. However, another possible explanation for the large asymmetric peakwidth could be the absence of the analyzer crystal. This leads to more diffusely reflected x-rays being collected at the detector, which, as a rule, increases the FWHM.

In both Figs. 2(a) and (b), we observe that the sample grown at 750° C does not follow the trend displayed by the samples grown at lower temperatures. From the x-ray data, this film appears to be of poorer structural quality than the film grown at 650° C. However, XTEM measurements of this film, shown below, reveal that it is about four times thinner than the films grown at lower temperatures for the same length of time (3 hours). This reduced thickness is probably because the growth temperature is very close to the temperature at which decomposition of GaN becomes significant (approximately 800° C). At this temperature and above, significant decomposition of GaN takes place during growth and consequently there is a reduction in the growth rate. A thinner film leads to a wider FWHM, since the measurements are sampling the film very close to the heterointerface, and causes the FWHM of the 750° C film to deviate from the expected trend.

We have also performed reciprocal space mapping around the symmetric (0002) reflections for films grown at 600° C and 750° C, as shown in Figs. 3(a) and (b) respectively. We note that the map for the film grown at 600° C shows a significantly greater elongation along the $k_{||}$ axis compared to the film grown at 750° C, implying a larger degree of tilt, or mosaicity, in the lower temperature films. Tilt in a film would be caused by dislocations with a screw Burger's vector component in the (0001) direction, implying a decrease in the number of screw-type threading dislocations with increasing growth temperature. Again, this is seen to agree with our observations in Ref. [6], where the AFM images were reported to show a decreasing number of spiral growth fronts with increasing growth temperature.

XTEM studies of these films also show a decreasing defect density with increasing growth temperature as well. Figure 4 shows low resolution TEM images of films grown at 600° C and 750° C for 3 hours each. It is clear that the film grown at 750° C is about four times thinner than the one grown at 600° C, indicating the reduced growth rate at elevated temperatures mentioned above. Also, the defect density in the high temperature film is observed to be much lower and its surface seen to be much smoother. From the TEM images the high temperature film shows a dislocation density of about 4×10^9 cm^{-2}, which is approximately half that of the lower temperature film. In the high temperature film the density of threading dislocations which intersect the surface is about 2×10^8 cm^{-2}. The low temperature film has several planar defects lying in the (0001) plane which are most probably stacking faults. High resolution images and diffraction patterns, not shown here, of the film grown at 600° C reveal several regions of cubic stacking. In general we find from both conventional and high resolution TEM the presence of a large variety of defects in films grown at low temperatures – stacking faults, cubic inclusions, prismatic dislocations as well as regular dislocations. All of these TEM results are discussed in greater detail elsewhere [10].

The observations above strongly suggest the use of high growth temperatures for GaN MBE heteroepitaxy on SiC. However, high temperature epitaxy is limited by two factors: (1) the decreasing growth rate caused by the decomposition of GaN at temperatures above about 800° C, and (2) the decreasing surface Ga concentration due to the higher Ga desorption rate. The second factor does not affect the quality of the GaN film as long as the growth is Ga-rich. However, a dra-

matic effect of surface Ga concentration on film surface morphology is seen. In Fig. 5, we show AFM images of films grown at 725° C with different Ga concentrations. Both of these films are grown in the Ga-rich growth regime, in which the RHEED pattern during growth is streaky implying relatively smooth morphology [5,11]. The image shown in Fig. 5(a) is of a film grown with Ga/N flux ratio close to the boundary between Ga-rich and N-rich growth, while that in (b) has been grown at almost a two times higher Ga flux. We observe the striking improvement in surface morphology in (b), where the growth is seen to be smooth and 2-D while the film in (a) is seen to have deep trenches separating relatively smooth areas. These two films have similar bulk quality as indicated by x-ray measurements which show a 1.5 arcmin FWHM for both films and thickness fringes for both at a thickness of 2000 Å. We note that the bulk quality of these films is not as good as the films mentioned in our earlier x-ray studies, although this may be due to a significant depletion of our Ga source which occurred during these varying Ga flux experiments. Additional experiments are planned to further study the optimization of the x-ray FWHM values.

IV CONCLUSIONS

We have investigated the optimization of GaN growth on hydrogen-etched 6H-SiC (0001) substrates by MBE. We find that increasing the growth temperature improves the bulk quality of the films, as suggested in Ref. [6]. In particular, the mosaicity of the film is seen to decrease sharply, as indicated by x-ray k-space maps. XTEM studies also reveal a large number of stacking faults and cubic GaN inclusions in the low temperature films. Further, we find that films of excellent surface morphology, that compare well with films grown by metal organic chemical vapor deposition [12], are grown under highly Ga rich conditions. From our studies, we conclude that high quality heteroepitaxy of wurtzite GaN on 6H-SiC (0001) is possible using Ga rich conditions with a growth temperature very close to the decomposition temperature of GaN.

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[1] S. Strite, M. E. Lin and H. Morkoç, Thin Solid Films, **231**, 197 (1993).

[2] R. F. Davis, T. W. Weeks, M. D. Bremser, S. Tanaka, R. S. Kern, Z. Sitar, K. S. Ailey, W. G. Perry and C. Wang, Mater. Res. Soc. Symp. Proc. Vol. **395**, 3 (1996).

[3] P. Waltereit, O. Brandt, A. Trampert, M. Ramsteiner, M. Reiche, M. Qi, and K. H. Ploog, Appl. Phys. Lett. **74**, 3660 (1999); A. Thann, O. Brandt, Y. Takemura, A. Trampert, and K. H. Ploog, Appl. Phys. Lett. **75**, 944 (1999).

[4] V. Ramachandran, M. F. Brady, A. R. Smith, R. M. Feenstra, D. W. Greve, J. Electron. Mater. **27**, 308 (1998).

[5] A. R. Smith. V. Ramachandran, R. M. Feenstra, and D. W. Greve, A. Ptak, T. H. Myers, W. L. Sarney, L. Salamanca-Riba, M.-S. Shin and M. Skowronski, MRS Internet J. Nitride Semicond. Res. **3**, 12 (1998).

[6] V. Ramachandran, A. R. Smith, R. M. Feenstra and D. W. Greve, J. Vac. Sci. Technol. A **17**, 1289 (1999). X-ray double crystal (0002) line widths in this work are in the range 1–2 arcmin. That is correctly shown in Fig. 5 of the work, although at one place in the text of the paper the widths are incorrectly stated as “1–2 arcsec” rather than 1–2 arcmin.

[7] M. E. Lin, S. Strite, A. Agarwal, A. Salvador, G. L. Zhou, N. Teraguchi, A. Rockett, and H.

Morkoç, Appl. Phys. Lett. **62**, 702 (1993).

[8] M. A. L. Johnson, Shizuo Fujita, W. H. Rowland, Jr., K. A. Bowers, W. C. Hughes, Y. W. He, A. El-Masry, J. W. Cook, Jr., J. F. Schetzina, J. Ren, and J. A. Edmond, J. Vac. Sci. Technol. B **14**, 2349 (1996).

[9] R. Lantier, A. Rizzi, D. Guggi, H. Lüth, B. Neubauer, D. Gerthsen, S. Frabboni, G. Colì, and R. Cingolani, MRS Internet J. Nitride Semicond. Res. **4S1**, G3.50 (1999).

[10] W. L. Sarney, L. Salamanca-Riba, V. Ramachandran, R. M. Feenstra, D. W. Greve, submitted to MRS Internet J. Nitride Semicond. Res.

[11] E. J. Tarsa, B. Heying, X. H. Wu, P. Fini, S. P. DenBaars, and J. S. Speck, J. Appl. Phys. **82**, 5472 (1997).

[12] D. Kapolnek, X. H. Wu, B. Heying, S. Keller, B. P. Keller, U. K. Mishra, S. P. DenBaars, and J. S. Speck, Appl. Phys. Lett. **67**, 1541 (1995).

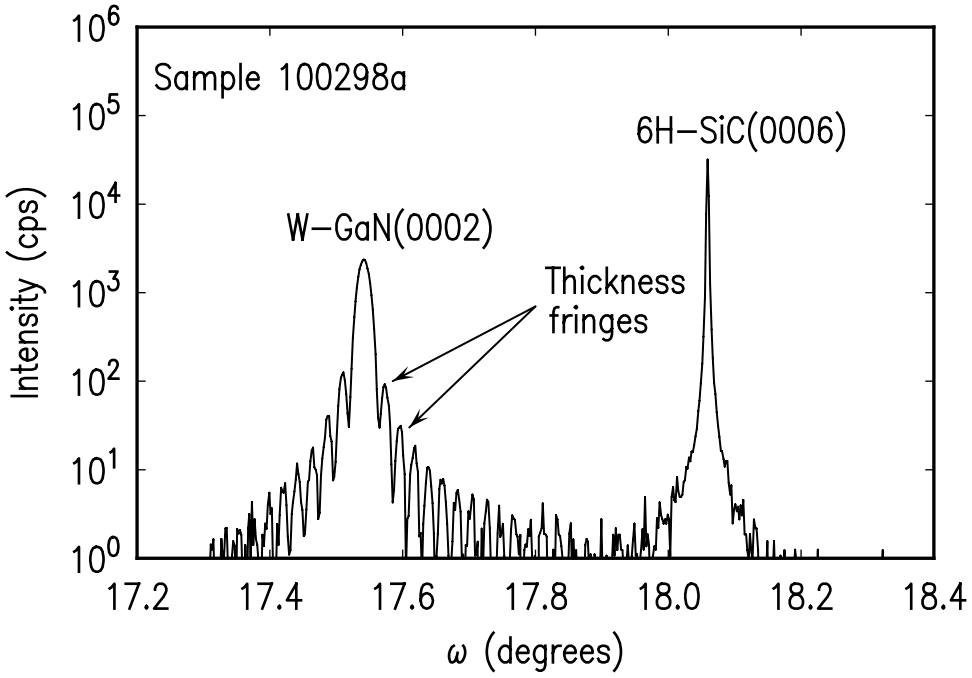

Figure 1 Triple crystal HRXRD radial scan for a GaN film of thickness 2000 Å grown on 6H-SiC (0001) at 700 ° C.

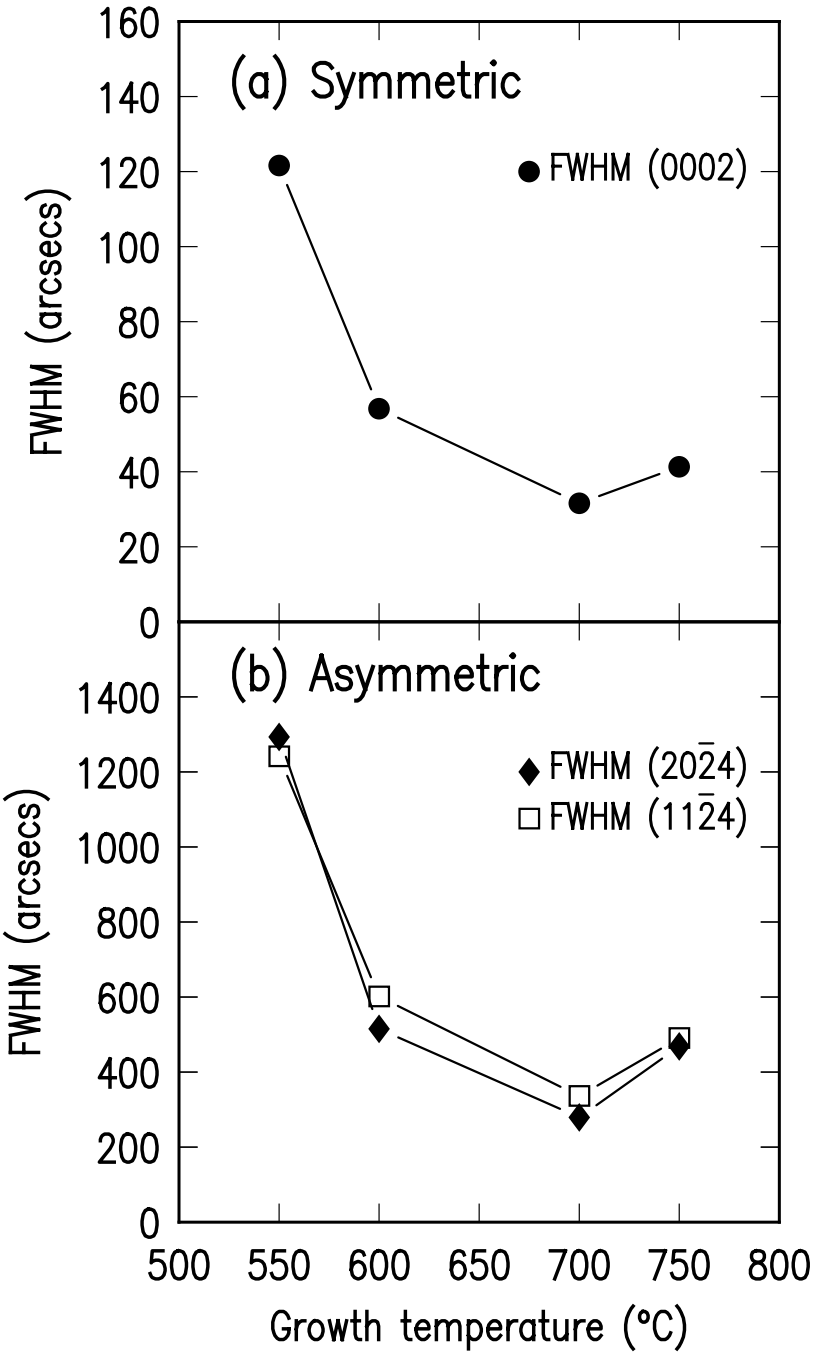

Figure 2 HRXRD FWHM data for GaN films grown on 6H-SiC (0001) as a function of temperature: (a) Symmetric (0002) reflection (triple crystal ω-2θ scans); (b) asymmetric (11$\bar{2}$4) and ($\bar{2}$024) reflections (double crystal ω scans).

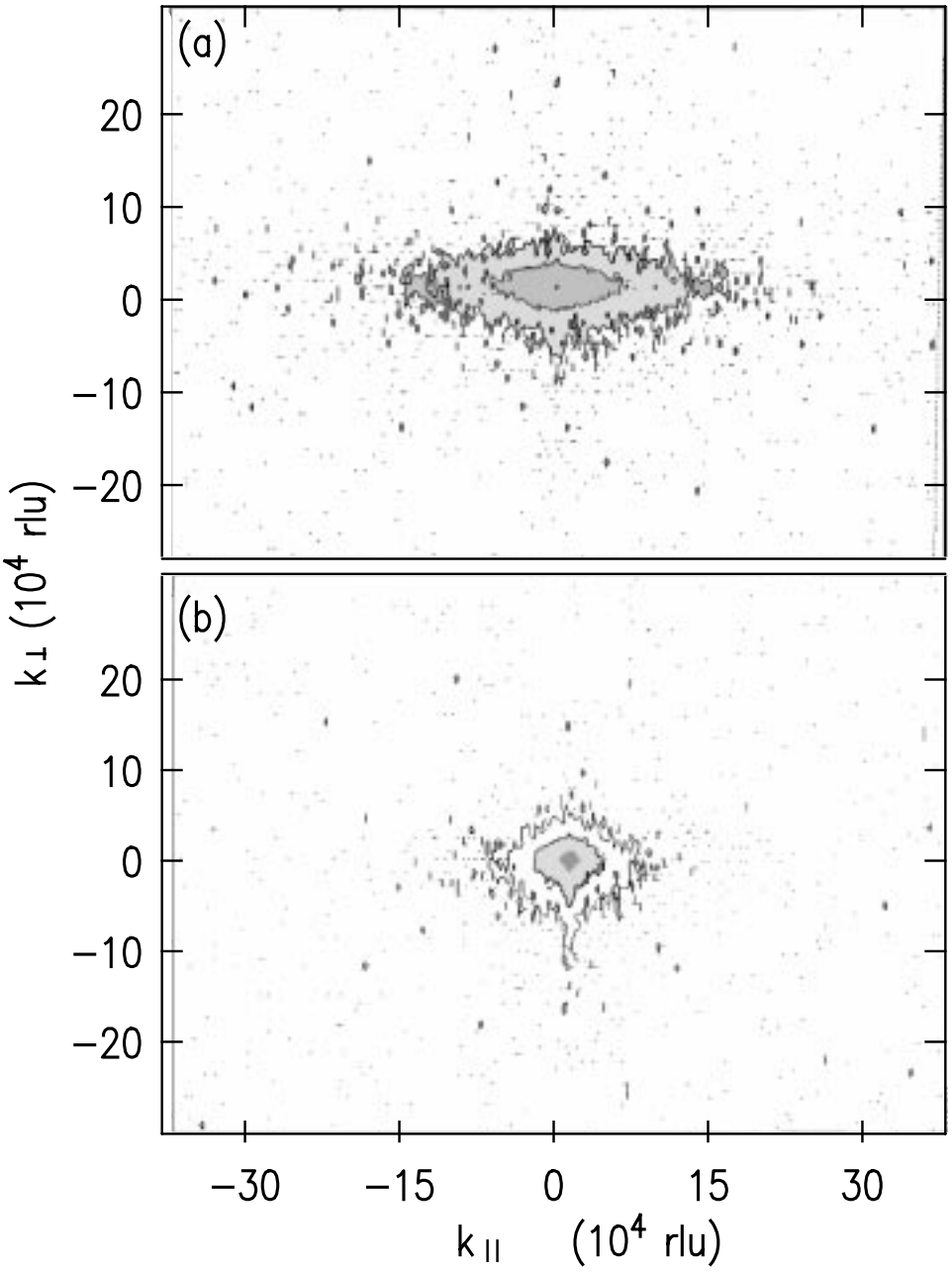

Figure 3 X-ray k-space maps of the (0002) reflection for films grown at (a) 600 ° C and (b) 750 ° C.

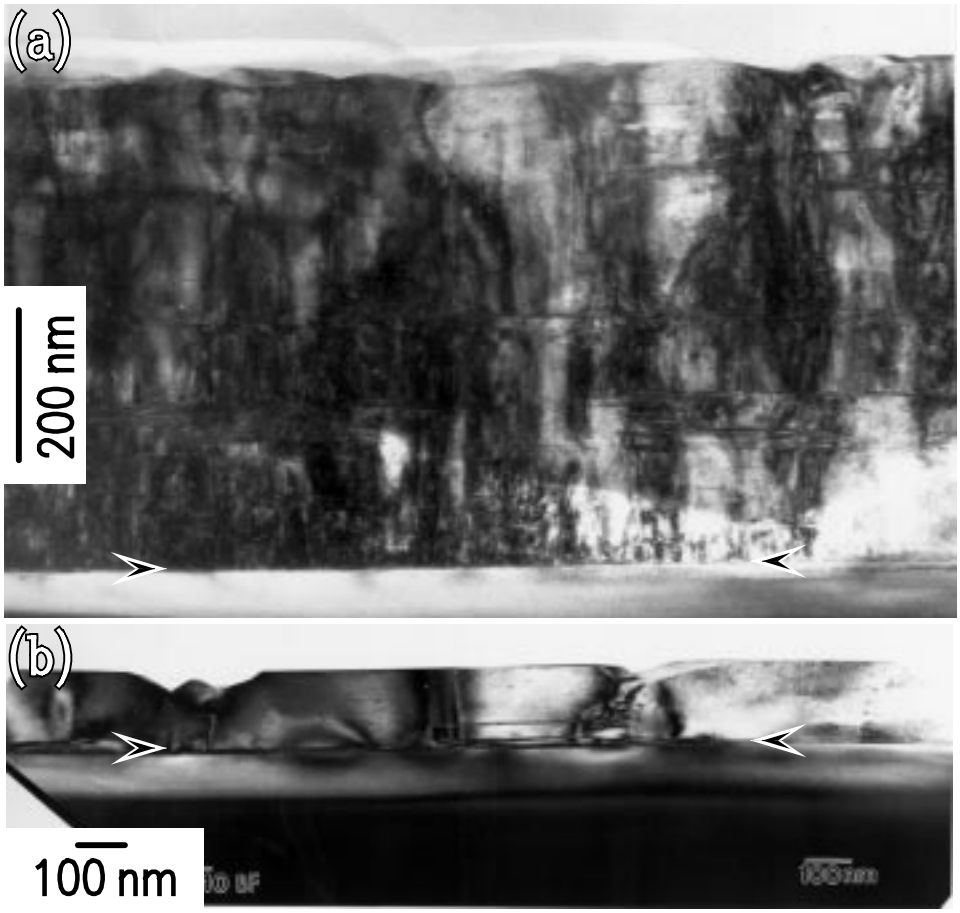

Figure 4 Cross-sectional TEM images of GaN films grown at (a) 600°C and (b) 750°C. Arrowheads indicate the GaN–SiC heterointerface.

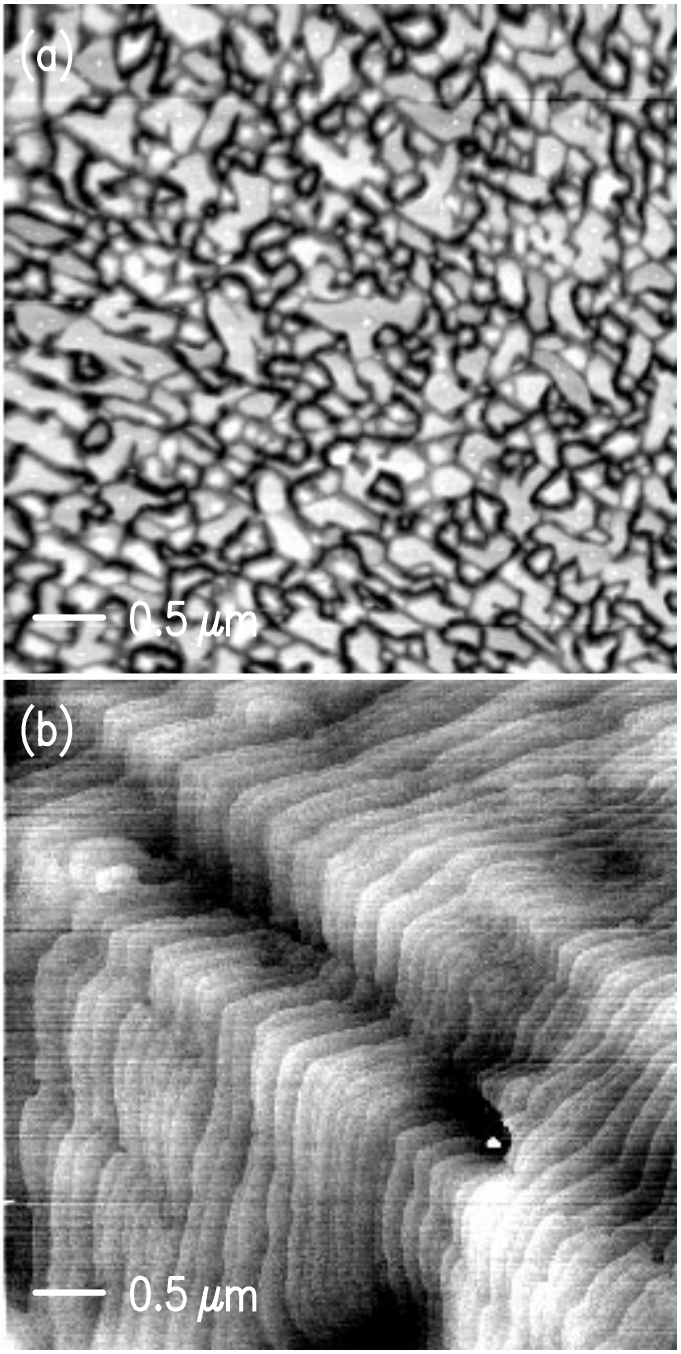

Figure 5 GaN film morphology in the Ga-rich growth regime with Ga flux during growth increasing from (a) to (b). Grey scale ranges are 2.8 and 1.8 nm for (a) and (b), respectively.