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Role of Ga-flux in dislocation reduction in GaN films grown on SiC(0001)

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GaN films are grown by plasma-assisted molecular beam epitaxy on SiC substrates. The width of the x-ray rocking curve for the $(10\bar{1}2)$ reflection exhibits a distinct minimum for Ga/N flux ratios which are only slightly greater than unity. Correlated with this minimum the surface morphology is somewhat rough, with a hill and valley topography. Based on transmission electron micrographs, the reduction in rocking curve width is attributed to enhanced annihilation of edge dislocations due to their tendency to cluster at topographic valleys.

The large band gap, high breakdown field, and high electron saturation velocity of GaN makes it ideal for use in visible-to-UV optoelectronic devices and in high speed, high power electronic applications. Most GaN epitaxial films used for devices have been deposited on sapphire, which despite its large lattice mismatch to GaN (≈16%) has nevertheless produced epitaxial GaN of relatively high quality [1]. Silicon carbide has a much better lattice match to GaN (3.4%), and has gained in popularity in recent years as a substrate for both molecular beam epitaxy (MBE) and metal-organic vapor phase epitaxy of GaN. Although some early studies of GaN deposition on SiC employed low-temperature buffer layers for nucleation of the GaN [2] (in analogy with nucleation on sapphire), more recent workers have deposited the GaN directly on a suitably prepared SiC surface [3-8].

The preparation method for our 6H-SiC substrates employs a two-step procedure [3-8]. First, hydrogen-etching of the SiC at 1600–1700°C and 1 atm pressure is performed to eliminate polish damage (and produce only half or full unit-cell high steps) [9]. Second, in ultra-high-vacuum, Si deposition followed by annealing is 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 is then taken to the growth temperature and deposition of the GaN is performed. Films of thickness around 1 μm are grown at temperatures of 730–800°C. Films are characterized using transmission electron microscopy (TEM), atomic force microscopy (AFM), and rocking curves of high-resolution x-ray diffraction reflections, the latter performed in a triple-axis configuration.

Previous studies have revealed the dependence of the GaN surface morphology with the ratio of active Ga flux to active N flux [7,8,10]. By "active flux" we mean the flux of atoms staying on the surface (i.e. incident minus desorbed fluxes). Following prior work we take this ratio to be unity at the point where a rough to smooth transition is seen by reflection high-energy electron-diffraction (RHEED) [10,11]. Flux ratios are scaled from this point in accordance with the varying Ga flux. For samples grown under very Ga-rich conditions, Ga/N ratio above about 1.5, the morphology is flat with only monoatomic steps revealed in AFM. In contrast, for Ga/N ratios near 1.3, pits with typical separation of about 1 μm are seen in the morphology. These pits are believed to arise from some sort of preferential film decomposition during growth at threading dislocations. As the Ga/N ratio is reduced to about 1.1 these pits proliferate, and merge into trenches on the surface which separate plateaus of atomically flat morphology.

Associated with the change in morphology of the GaN films as a function of Ga/N flux ratio, we also find significantly different values for the rocking curve width of $(10\bar{1}2)$ reflections, as shown in Fig. 1(a). We plot there the rocking curve FWHM as a function of Ga flux, for fixed N flux, and for films grown at various temperatures. For each temperature, the point at which a rough to smooth transition is seen in RHEED (corresponding to a Ga/N ratio of unity) is marked by a dashed line. This transition point is very well defined at 730°C, but less so at higher temperatures. We see from Fig. 1(a) that the $(10\bar{1}2)$ width shows a minimum at Ga/N ratios slightly greater than unity, and increases for larger Ga fluxes. This minimum in the rocking curve width is quite sharp and deep for the case of 730°C growth and is less so for the higher temperature growth, in agreement with the general variation in the surface morphology vs. Ga flux as seen by both RHEED and AFM.

Results for the (0002) rocking curve width are shown in Fig. 1(b). In this case we do not observe any dramatic variation with Ga flux or temperature. As we have previously established, there are 1–2 orders of magnitude fewer threading screw dislocations in our GaN films than threading edge dislocations [8] (the width of the $(10\bar{1}2)$ rocking curve is thus a measure of the density of threading edge dislocations). From Fig. 1(b), there is a slight tendency for the (0002) rocking curve width to increase at high Ga fluxes, although within our sample to sample uncertainty this is not so significant.

From Fig. 1, it appears that the density of edge dislocations is significantly reduced in the films grown with Ga/N ratio only slightly greater than unity, i.e. for films whose morphology is somewhat rough. This reduction could of course result from the initial nucleation of the film, or it could arise from enhanced dislocation annihilation in those rough films. We believe that the latter mechanism is dominant in our growths, as evident in the TEM images presented below.

Figure 2 shows TEM results from GaN films grown under conditions of high Ga/N ratio, near 1.5 and at growth temperature of 750°C. The cross-sectional result, Fig. 2(a), reveals a very high density of dislocations near the SiC/GaN interface, as expected for heteroepitaxy of such large mismatched materials. Significant annihilation of the dislocations occurs in the initial nanometers of the film (where GaN nuclei formed in the initial stages of deposition coalesce [8]). Some annihilation or combination of dislocation is also seen as the threading dislocations extend toward the film surface, as indicated by the arrows in Fig. 2(a), but most of the threading dislocations are quite straight and parallel as they extend up to the surface. The surface is flat, due to the high Ga/N ratio used. Note that the TEM specimen used for Fig. 2(a) was somewhat thinner near the film surface as compared to at the GaN/SiC interface. Thus, some of the threading dislocations seen there appear to "terminate" before they reach the film surface, but actually they are just exiting the side of the TEM specimen. Figure 2(b) shows a plan-view image of a film grown under similar conditions. The edge dislocations at the surface are clearly seen, and they appear to be more or less randomly distributed. Their density is about 2×10^{10} cm^{-2}, which is typical of these films grown with high Ga/N ratio.

Figure 3 shows TEM results from GaN films grown under conditions of relatively low Ga/N flux ratio, near 1.1, and at a temperature of 800°C. The film surface seen in Fig. 3(a) is rough, with facetted trenches, as also seen in AFM. The dislocations evident in Fig. 3(a) again form with high density near the GaN/SiC interface, and threading dislocations extend up toward the film surface. Numerous loops or inverted "Y" shape branches are seen where dislocations meet each other and annihilate (when they have opposite Burgers vectors) or combine into a single dislocation (when they have different but suitable Burgers vectors). Most revealing in terms of the difference between growth with high or low Ga/N flux ratio are the plan-view images, Figs. 3(b) and (c). Since the film morphology is undulated, those images display brighter and darker regions (corresponding to variations in specimen thickness) with the former being the topographic valleys. Almost all the dislocations in the film are seen to be located at the valleys, indicating that they have moved there during the film growth. Dislocation densities for such films grown with Ga/N ratio near unity are reproducibly found to be $\leq$ 1×10^{9} cm^{-2}, consistent with the low $(10\bar{1}2)$ rocking curves widths observed for these films.

As demonstrated above, we observe significant dislocation reduction for films grown with low Ga/N flux ratio. It appears that these rough films lead to a clustering of the dislocations near topographic valleys. The mechanism for this clustering may be a modulated stress field induced by the morphology. Alternatively, we also note that a dislocation intersecting a surface obliquely feels a force which pulls the dislocation towards a normal orientation [12]; this mechanism, for a hill and valley morphology, will lead to a gradual clustering of the dislocations in the valleys. In any case, for nearby dislocations their own strain fields will ultimately determine attractive or repulsive interactions between them [13]. We believe that the probability of annihilation or combination is greatly enhanced when dislocations are clustered [Figs. 3(b) and (c)] rather than randomly spaced [Fig. 2(b)] since the interactive force is, in general, inversely proportional to the distance between dislocations. We note finally that the mobility of dislocations may be affected by the Ga/N ratio, but we know that during growth all of our films are terminated by a bilayer of mobile Ga atoms [14], thus implying no change in dislocation mobility.

In summary, we find a significant reduction in the width of the $(10\bar{1}2)$ rocking curve as the Ga/N flux ratio is reduced from values near 1.5 down to values near 1.1. Associated with this reduction in the rocking curve width we observe a change in morphology, from flat to rough. We attribute the reduction in the rocking curve width to annihilation of edge dislocations due to their tendency to cluster at topographic valleys in the rougher films. We note finally that the morphology of our films that have minimum dislocation density end up to be somewhat rough, and in this respect may not be optimal for device applications. We have also investigated two-step growth procedures in which the Ga/N flux ratio is initially low in order to promote annihilation and then is increased to produce flat morphology. Some limited success with this procedure was obtained, although further research is needed to simultaneously optimize the dislocation density and the surface morphology.

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References

[1] B. Heying, X. H. Wu, S. Keller, Y. Li, D. Kapolnek, B. P. Keller, S. P. DenBaars, and J. S. Speck, Appl. Phys. Lett. **68**, 643 (1996).

[2] F. R. Chien, X. J. Ning, S. Stemmer, P. Pirouz, M. D. Bremser and R. F. Davis, Appl. Phys. Lett. **68**, 2678 (1996).

[3] V. Ramachandran, A. R. Smith, R. M. Feenstra, and D. W. Greve, J. Vac. Sci. Technol. A **17**, 1289 (1999).

[4] V. M. Torres, J. L. Edwards, B. J. Wilkens, D. J. Smith, R. B. Doak, and I. S. T. Tsong, Appl. Phys. Lett. **74**, 985 (1999).

[5] Q. Z. Xue, Q. K. Xue, Y. Hasegawa, I. S. T. Tsong, and T. Sakurai, Appl. Phys. Lett. **74**, 2468 (1999).

[6] 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).

[7] O. Brandt, R. Muralidharan, P. Waltereit, A. Thamm, A. Trampert, H. von Kiedrowski, and K. H. Ploog, Appl. Phys. Lett. **75**, 4019 (1999).

[8] 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, J. Electron. Mat. **30**, 162 (2001).

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

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

[11] A. R. Smith, V. Ramachandran, R. M. Feenstra, 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).

[12] F. R. N. Nabarro, *Theory of Crystal Dislocations* (Oxford, London, 1967), p. 288.

[13] Almost all of the threading dislocations in our films have edge character, with any one of the six $1/3\langle 11\bar{2}0\rangle$ type Burgers vectors. The sign of force between a dislocation and its neighbor lying on its glide plane depends mainly on the angle θ between their Burgers vectors [see e.g. A. H. Cottrell, *Dislocations and Plastic Flow in Crystals* (Oxford, London, 1953)]: the force is attractive when $\theta = 120°$ and $180°$ and is repulsive when $\theta = 0°$ and $60°$. For $\theta = 120°$ the pair will combine into one with a Burgers vector of the same magnitude and for $\theta = 180°$ they will annihilate.

[14] A. R. Smith, R. M. Feenstra, D. W. Greve, M.-S. Shin, M. Skowronski, J. Neugebauer, and J. Northrup, J. Vac. Sci. Technol B **16**, 2242 (1998).

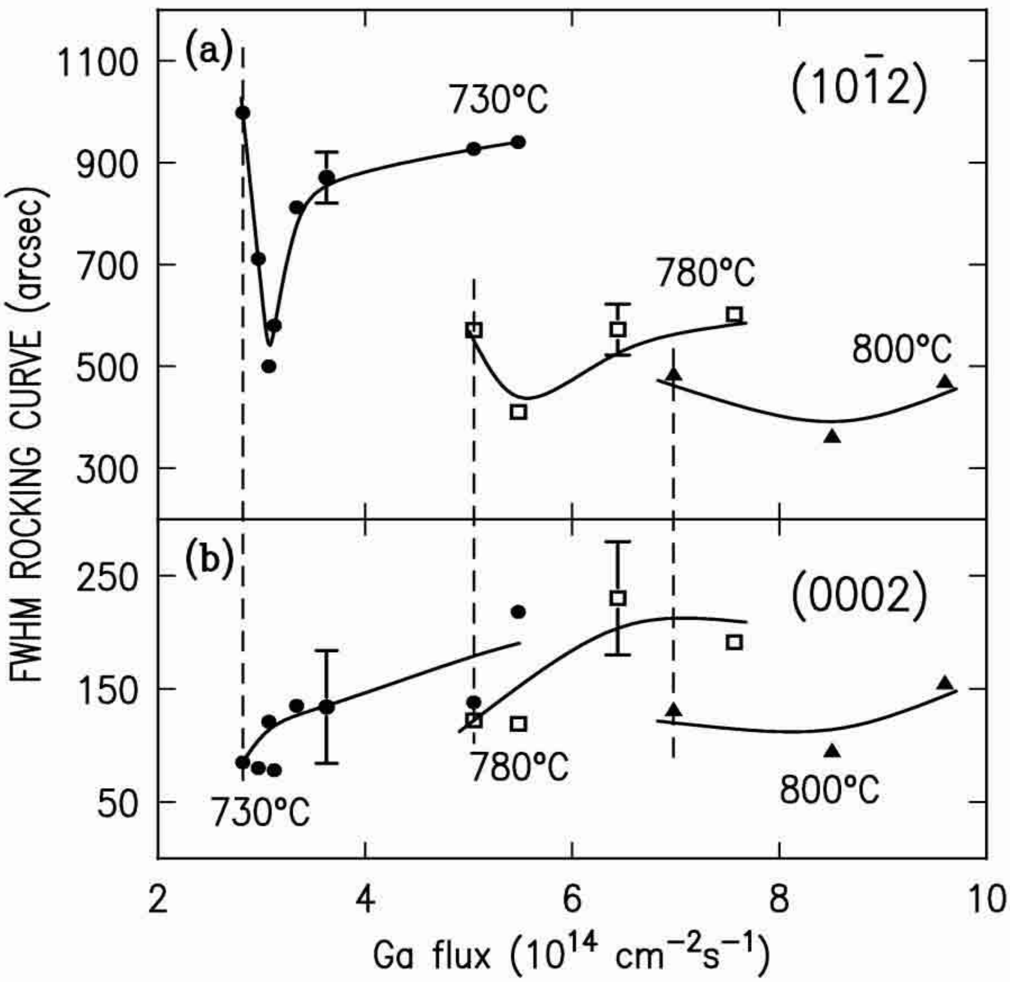

Fig. 1. FWHM of the x-ray rocking curves for (a) (10$\bar{1}$2) and (b) (0002) reflections. Different symbols refer to films grown at temperatures of 730, 780, and 800°C. At each temperature, the dashed lines correspond to the observed Ga flux at which the RHEED patterns indicated a rough to smooth transition. Solid lines are drawn as a guide to the eye. Typical uncertainties due to sample to sample variation are indicated.

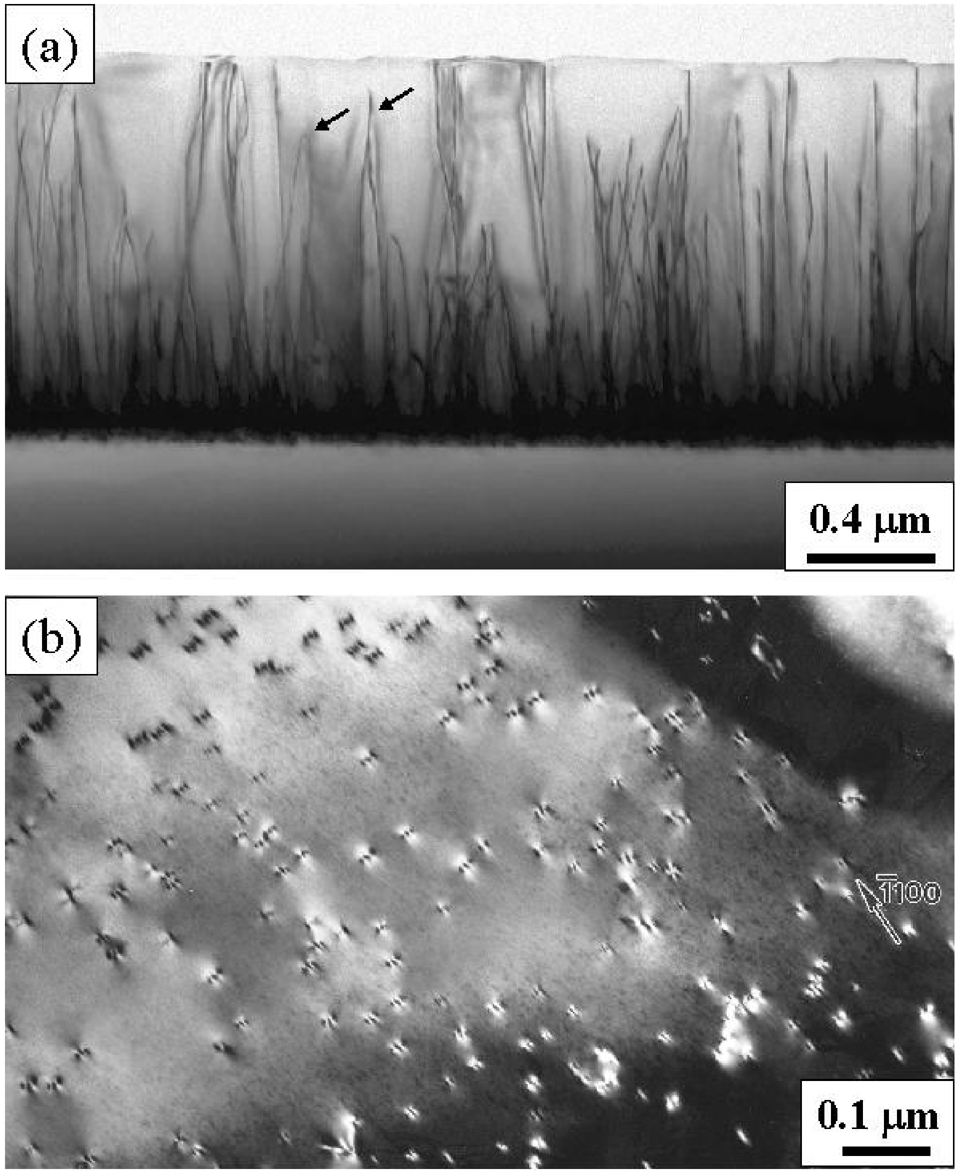

Fig. 2 TEM bright-field images obtained from films grown with a relatively high value of Ga/N ratio (R~1.5): (a) Cross-sectional image of a 1.2-μm-thick film. Pairs of threading dislocations were observed to combine into a half loop (left arrow) or into a single dislocation (right arrow). (b) Plan-view image indicating a dislocation density of about 2×10^{10} cm^{-2} at the top part of the film.

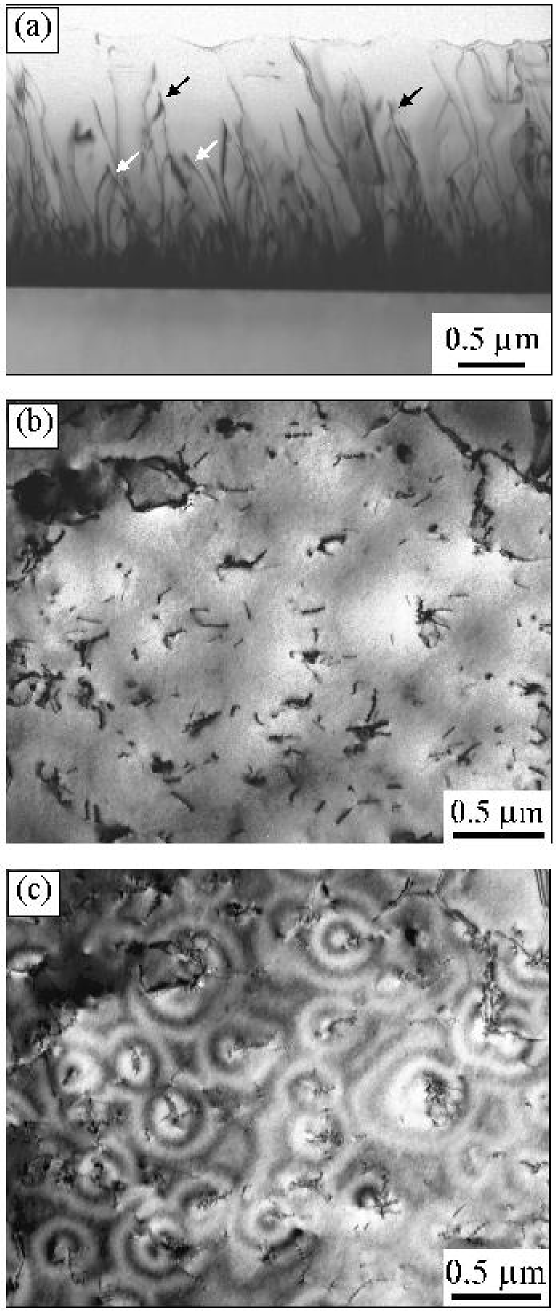

Fig. 3 TEM bright-field images of a 1.9-μm-thick film grown with a relatively low Ga/N ratio (R~1.1). (a) Cross-sectional image revealing surface undulation with amplitude ~50 nm. Pairs of threading dislocations were observed to combine into one dislocation (black arrows). Dislocations also annihilated each other by joining into a half loop (white arrows). (b) and (c) Plan-view images revealing dislocations remaining mostly at the valleys of the morphology (bright areas). Image (c) was acquired from the same sample area as in (b) but with a dynamical, two-beam condition that results in thickness contours that reflect the film morphology. Dislocation density is about 1×10^{9} cm^{-2} at the top part of the film.