Mechanisms of dislocation reduction in GaN using an intermediate temperature interlayer

E.D. Bourret-Courchesne, K.M. Yu, M. Benamara, Z. Liliental-Weber and J. Washburn

E.O. Lawrence Berkeley National Laboratory, 1 Cyclotron Road (M.S. 2-200), Berkeley CA 94720.

Abstract:

A dramatic reduction of the dislocation density in GaN was obtained by insertion of a single thin interlayer grown at an intermediate temperature (IT-IL) after the initial growth at high temperature. A description of the growth process is presented with characterization results aimed at understanding the mechanisms of reduction in dislocation density. A large percentage of the threading dislocations present in the first GaN epilayer are found to bend near the interlayer and do not propagate into the top layer which grows at higher temperature in a lateral growth mode. TEM studies show that the mechanisms of dislocation reduction are identical to those described for the epitaxial lateral overgrowth process, however a notable difference is the absence of coalescence boundaries.

Keywords: GaN, OMVPE, dislocation, TEM, interlayer.

Corresponding author: Edith Bourret-Courchesne, Tel.: (510) 486-5553
Fax: (510) 486-5530, e-mail: EDBourret@lbl.gov

INTRODUCTION

It is well known that GaN grown on sapphire contains a high density of threading dislocations in the range of 10^9 to $10^{10}/cm^2$ due to the large difference of the lattice constants and the thermal expansion coefficients between GaN and sapphire. Threading dislocations have been found to act as non-radiative centers and scattering centers in electron transport that is detrimental to the performance of light emitting diodes and field effect transistors [1]. Efforts around the world are aimed at reducing the density of structural defects in GaN. The most successful techniques to date are the epitaxial lateral overgrowth techniques (ELO). Many studies have focused on understanding the mechanisms of dislocation reduction using this technique[2-6]. In the ELO process, a dielectric mask is deposited on a first GaN layer. Lithographic techniques are used to open patterns in the mask. Then, growth of GaN is resumed, nucleation of growth occurs in the openings and, with the proper growth conditions, lateral growth above the mask allows the selected epitaxial areas to fully coalesce. Dislocations do propagate through the openings but bend over the mask where the growth is predominantly lateral. A number of devices with improved performance have been produced using the ELO technique.

The use of pendeo-epitaxy has also resulted in production of more performant devices [7-9]. In pendeo-epitaxy, lateral overgrowth is initiated on etched GaN mesas. Similarly to the standard ELO technique, dislocations propagate above the seed areas but bend over due to lateral growth in the pendeo area.

Another technique, the cantilever method, makes use of the a patterned substrate[10]. GaN does not nucleate easily in the etched stripes of the pattern and lateral overgrowth occurs above these grooves.

ELO and the related techniques briefly described above are multi-step processes and simpler methods to reduce dislocation density would be preferable in production. Iwaya and Amano [11,12] have shown that a series of low temperature interlayers (LT-IL) grown at 500˚C improves the crystalline quality of GaN, AlN and AlGaN by reducing the density of threading dislocations. One such interlayer is shown to reduce the density of threading dislocations by about one order of magnitude. Two orders of magnitude reduction is reported by using 5 interlayers. However a high number of such interlayers increase the level of stress in the material which shows a tendency to crack. Similar results have been obtained by Yang et al. [13]. Here, we report on a process that reduces the density of threading dislocations by about three orders of magnitude from well above $10^{10}/cm^2$ down to below $4x10^7/cm^2$. This process makes use of a single intermediate temperature interlayer (IT-IL).

EXPERIMENT

GaN was deposited on sapphire (c-plane) substrates by organometallic vapor phase epitaxy. Pd-purified hydrogen was used as the carrier gas. Triethylgallium (TEGa) and water-free dimethylhydrazine (DMHz) [14] supplied by Epichem Inc., were used as gallium and nitrogen precursors. The sapphire substrates were first cleaned in situ with pure hydrogen at 1000˚C. Dimethylhydrazine was introduced at 1000˚C for nitridation of the surface. Deposition of GaN was initiated with TEGa and DMHz introduced simultaneously in the deposition chamber at 580˚C. The flow of TEGa was

7.8 μmole/min and the molar ratio of V/III precursors was 50. The differences between growth of GaN using dimethylhydrazine and growth of GaN using ammonia have been previously reported [15]. About 600 nm to 1000 nm of GaN was deposited at 1000˚C (first high temperature layer). The growth rate was about 1μm/hr. The temperature was then rapidly ramped down to an intermediate temperature (700 to 900˚C) for growth of the intermediate temperature-interlayer (IT-IL) and growth was allowed to proceed at this temperature for 15 minutes. The temperature was then rapidly ramped back up to 1000˚C to grow the top high temperature GaN layer.

RESULTS AND DISCUSSION

Using laser reflectance we determined the transition temperature at which the growth mode changes from a three-dimensional mode (3D) to a two dimensional mode (2D) [11]. A 3D growth mode results in the roughening of the surface which results in a decrease of the reflectance of the layer. A 2D growth mode smooths the surface and the reflectance increases to its maximum value that can be calculated theoretically [16]. With our growth conditions 2D growth occurs above about 995˚C. When the temperature is suddenly decreased to yield a 3D growth mode, with a low flow of reactants, a low density of small islands is nucleated on the growing surface. We found that at lower temperatures (below 900˚C) the islands are Ga-rich. At 800˚C, these islands are randomly distributed, not uniform in sizes and have a metallic, rounded appearance when observed by optical microscopy. These islands then convert to GaN islands with a distinctive pyramidal shape during the temperature increase back to 1000˚C at which temperature a 2D growth mode is reestablished and growth of the islands progresses mostly laterally. The dislocations propagate in the growth direction and therefore bend over near the interface where the lateral growth takes place as seen in the cross-section TEM image shown in fig.1. The mechanism of dislocations is then identical to that described for the epitaxial lateral overgrowth process.

Figure 1(a and b) are dark field images taken under two beam conditions with $(1\bar{1}02)$ reflecting planes of a thin and a thick GaN/(IT-IL)GaN/GaN structure. The intermediate-temperature GaN (IT-IL) layer appears as a separation line between the two high-temperature layers in the image. Under these diffraction conditions, most of the dislocations are visible in the image. The most striking feature of these images is that it shows clearly that most of the threading dislocations in the first high temperature GaN layer do not propagate into the upper layer. Lateral growth from islands formed in a 3D growth mode at an intermediate temperature is a highly efficient process to avoid propagation of threading dislocations of any Burgers vector. In the thin sample, the dislocation density is reduced by three orders of magnitude, from well above 10^{10} cm^{-2} in the first HT-GaN to an average of 8.10^{7} cm^{-2} in the second one. For the thicker sample an average of only 4×10^{7} dislocations/cm^{-2} was found.

It is of particular interest to note that the residual threading dislocations are grouped at specific sites which correspond to the positions of the islands formed at the intermediate temperature. The material in between two successive dislocation groups is defect free and this over large areas (over 1.5 µm in fig.1). These areas are of comparable size to those obtained by conventional lateral epitaxial overgrowth. Detailed TEM study shows that the mechanisms of dislocation reduction are identical to those described for the ELO process [17,18]. However a notable difference is the absence of any coalescence boundaries indicative of the coherent nature of the 3D islands. It also appears that the contact between the flat areas of the lower layer and the laterally grown areas of the top layer is minimal: in figure 1b, it can be seen that the top layer separated from the lower layer during preparation of the TEM sample. This lack of contact may favor lateral growth with minimum stress and no driving force for a tilt of the layer as observed for lateral growth over a mask.

The surface morphology of the first high temperature GaN layer appears to determine the nucleation sites for the 3D islands nucleated at the intermediate temperature. The optical micrograph shown in figure 2a was obtained using Normarski interference contrast. The image shows that the surface has large flat areas and areas of non-planarity due to bunching of growth steps. Nucleation of the 3D islands is most likely to occur on these steps as can be seen on the TEM image, figure 2b.

CONCLUSIONS

The use of an intermediate temperature layer instead of a low temperature layer as studied by others [11-13] , allows formation of isolated islands at the IL interface instead of forming a pseudo-amorphous continuous buffer layer. The islands appear to nucleate coherently as there are no coalescence front boundaries. Multiple IT-ILs will most likely further reduce the defect density and the absence of a continuous low temperature layer should reduce the overall strain and tendency for cracking. Immediate establishment of lateral overgrowth between the islands makes the interlayer efficient at bending over most of the threading dislocations. At 800˚C, the precursors used in this study are fully decomposed[19] however, the islands nucleated at this temperature are Ga-rich with a metallic appearance. The process presented here is independent of the substrate used.

We expect that the IT-IL process will be useful as a nucleation scheme for homoepitaxy on GaN substrates grown by hydride vapor phase epitaxy that may still have a large number of dislocations. In terms of defect reduction, the results presented here are similar to those obtained with ELO however the IT-IL process does not require ex-situ processing steps and there is no defect accumulation at the coalescence front.

Acknowledgments:

This work was supported by the Director, Office of Science, Office of Basic Energy Sciences, Materials Sciences Division, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098. The use of the facility at the National Center for Electron Microscopy at E.O. Lawrence Berkeley National Laboratory is greatly appreciated.

Figure captions:

Figure 1: Transmission electron microscopy image of a thin (a) and a thick (b) IT-IL structure. Dark-field images taken under two beam conditions with $(1\bar{1}02)$ reflecting planes of the GaN/(IT-IL) GaN/ GaN structure.

Figure 2: Optical micrograph (left) with outlined bunched steps and TEM images (right) showing propagation of dislocations due to nucleation of 3D islands at intermediate temperature on these steps.

References

[1] T.-B. Ng, J. Han, R. M. Biefeld, and M. V. Weckwerth, J. Eletron. Mater. **27,** 190 (1998).

[2] D. Kalponek, S. Keller, R. Vetury, R. D. Underwood, P.Kozodoy, S. P. DenBaars, and U. K. Mishra, Appl. Phys. Lett. **71,** 1204 (1997).

[3] O.-H. Nam, M. D. Bremser, T. S. Zheleva, and R. F. Davis, Appl. Phys. Lett. **71,** 2836 (1997).

[4] H. Marchand, X. H. Wu, J. P. Ibbetson, P. T. Fini, P. Kozodoy, S. Keller, J. S. Speck, S. P. DenBaars, and U. K. Mishra, Appl. Phys. Lett. **73,** 747 (1998).

[5] A. Usui, H. Sunakawa, A. Sakai, and A. A. Yamaguchi, Jpn. J. Appl. Phys. Part 2 **36,** L899 (1997).

[6] A. Sakai, H. Sunakawa, and A. Usui, Appl. Phys. Lett. **71,** 2259 (1997).

[7] T. Zheleva, S. Smith, D. Thomson, K. Linthicum, T. Gerhke, P. Rajagopal, and R. Davis, J. Electron. Materials **28,** L5 (1999).

[8] K. J. Linthicum, T. Gehrke, D. Thomson, E. Carlson, P. Rajagopal, T. Smith, and R. Davis, Appl. Phys. Lett. **75,** 196 (1999).

[9] R. Davis, T. Gehrke, K. J. Linthicum, S. Zheleva, P. Rajagopal, C. A. Zorman, and M. Mehregany, MRS Internet J. Nitride Semicond. Res. **5S1,** W2.1 (2000).

[10] C. I. H. Ashby, C. C. Mitchell, J. Han, N. A. Missert, P. P. Provencio, D. M. Follstaedt, G. M. Peake, and L. Griego, Applied Physics Letters **77,** 3233 (2000).

[11] M. Iwaya, T. Takeuchi, S. Yamaguchi, C. Wetzel, H. Amano, and I. Akasaki, Jpn. J. Appl. Phys. Part 2 **37,** L316 (1998).

[12] H. Amano, M. Iwaya, N. Hayashi, T. Kashima, M. Katsuragawa, T. Takeuchi, C. wetzel, and I. Akasaki, MRS Internet J. of Nitride Semicond. **Res. 4S1,** G10.1 (1999).

[13] C.-C. Yang, M.-C. Wu, C.-H. Lee, and G.-C. Chi, J. of Appl. Phys. **87,** 4240 (2000).

[14] R. Odedra, L. M. Smith, S. A. Rushworth, M. S. Ravetz, J. Clegg, R. Kanjolia, S. J. C. Irvine, M. U. Ahmed, E. D. Bourret-Courchesne, N. Y. Li, and J. Cheng, J. Electron. Mater. (1999).

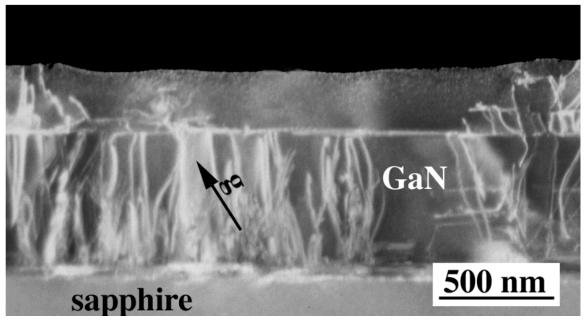
[15] E. D. Bourret-Courchesne, K. M. Yu, S. J. C. Irvine, A. Stafford, S. A. Rushworth, L. M. Smith, and R. Kanjolia, Journal of Crystal Growth **221,** 246 (2000).

[16] A. Stafford, S. J. C. Irvine, K. L. Hess, and J. Bajaj, Semicond. Sci. Technol. **13,** 1407 (1998).

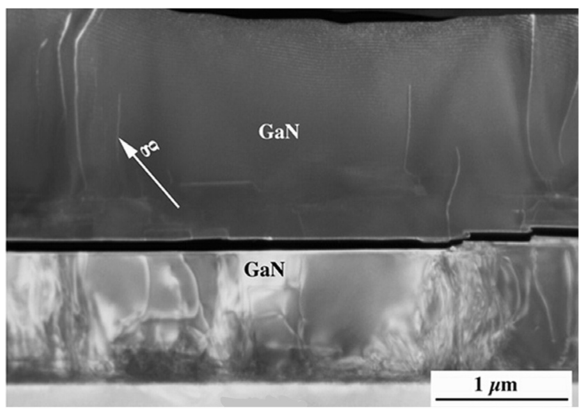
[17] M. Benamara, Z. Liliental-Weber, S. Kellermann, W. Swider, J. Washburn, and E. D. Bourret-Courchesne, in MRS Book 622, (North-Holland, Amsterdam), T6.14 (2000).

[18] M. Benamara, Z. Lilental-Weber, S. Kellermann, W. Swider, J. Washburn, and E. D. Bourret-Courchesne, J. of Crys. Growth **218,** 447 (2000).

[19] E. Bourret-Courchesne, Q. Ye, D. W. Peters, J. Arnold, M. Ahmed, S. J. C. Irvine, R. Kanjolia, L. M. Smith, and S. A. Rushworth, J. of Crystal Growth **217,** 47 (2000).



(a)



(b)

Fig. 1.

E.D. Bourret-Courchesne et al.

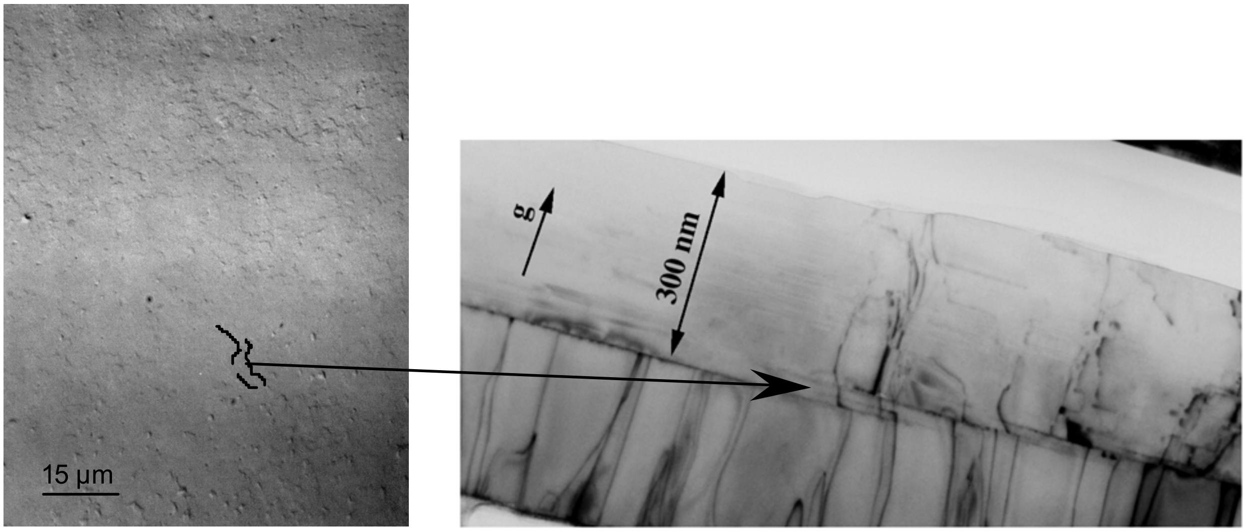


Fig. 2.

E.D. Bourret-Courchesne et al.