Dislocation density reduction in GaN using porous SiN interlayers

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Received zzz, revised zzz, accepted zzz

Published online zzz

PACS 81.05.Ea, 81.15.Hi, 68.37.Lp

The influence of a thin porous SiN_x interlayer on the growth of GaN by metalorganic chemical vapor deposition (MOCVD) has been studied. The interlayer is deposited on a GaN template by introducing silane in the presence of ammonia into the MOCVD chamber, and a GaN overlayer is deposited on the interlayer. The SiN_x interlayer produces inhomogeneous nucleation and lateral growth of the overlayer, causing bending of dislocations towards facet walls, and it also blocks some dislocations from entering the overlayer. The dislocation density for a GaN overlayer grown on a SiN_x interlayer was reduced to $7 \times 10^8 \text{ cm}^{-2}$, which is an order of magnitude less than that for a control sample grown without an interlayer.

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1 Introduction The large band gap, high breakdown field, and high electron saturation velocity of GaN make it ideal for use in visible-to-UV optoelectronic devices and in high speed, high power electronic applications. However, the large lattice mismatch between GaN and conventionally used substrates such as SiC or sapphire results in a high threading dislocation density in the GaN films [1]. One recent approach to reduce this dislocation density has been lateral epitaxial overgrowth (LEO) [2,3]. In this technique, a patterned dielectric mask (SiO₂ or SiN_x) is deposited on a GaN template layer, which stops the propagation of threading dislocations into the overlayer. The standard LEO process, however, requires several processing steps, which makes it a labor-intensive and expensive technique. Recently, there has been considerable interest in depositing a SiN_x mask *in situ* and thereby reducing the dislocation density by a spontaneously occurring LEO-like mechanism [4–11]. Such porous SiN_x nanomasks have been successfully employed as interlayers between GaN layers grown on sapphire [4–7], for GaN growth directly on sapphire [8], and for growth of GaN on AlN seed layers on Si [9]. The lateral growth of the overlayers in all cases appears to follow from the anti-surfactant effects of SiN_x on the GaN or AlN surfaces [10,11].

In this work, we follow these prior authors in employing porous SiN_x interlayer (i.e. a thin SiN_x layer which only incompletely covers the surface) on a GaN template, using both sapphire and SiC substrates. The thin porous SiN_x layer is deposited by flowing diluted silane in the MOCVD chamber in the presence of ammonia for a short duration. It is found that the GaN growth starts from inside the pores in the SiN_x film, resulting in the formation of GaN islands during the initial stages of growth. These GaN islands subsequently grow laterally over the SiN_x covered area and, finally, a fully coalesced film is achieved. Transmission electron microscope images show that the porous SiN_x layer effectively blocks a majority of threading dislocations from propagating vertically through the overlayer leading to an order of magnitude reduction in the dislocation density in the GaN overlayer.

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2 Experimental All samples grown for this study were grown by metalorganic chemical vapor deposition (MOCVD), using *c*-plane sapphire and 6H-SiC (Si-face) substrates. Trimethylgallium (TMG) and ammonia gases were used as precursors for GaN growth, and silane was used as source of Si for the deposition of SiN_x. The sapphire substrate was pre-heated in a stream of hydrogen at 1030°C for 3 min, after which a 30 nm thick GaN buffer layer was deposited at 560°C. That buffer layer was then annealed at 1030°C for 5 min before the growth of a ~3 μm thick GaN template layer at the same temperature. During the growth, the V/III ratio was kept at 4000 and the chamber pressure was maintained at 200 Torr. The 6H-SiC (3.5° miscut) substrates were H-etched at 1700°C to eliminate polishing damage [12], and then a ~100 nm thick GaN buffer layer was deposited directly on it at 565°C (the buffer layer was found to be continuous for this relatively low deposition temperature). No further annealing of this layer was performed, and the SiN_x interlayer was deposited on it as described below.

A thin layer of porous SiN_x was deposited on the GaN template or buffer layers prepared as described above. The porous SiN_x layer was deposited by flowing 50 sccm of 100 ppm silane (i.e. formed from 500 ppm silane diluted by hydrogen). The chamber pressure was kept at 200 and 30 Torr for growth experiments on sapphire and SiC, respectively, with ammonia flow maintained (TMG off), and the deposition temperature was 1030°C. SiN_x deposition times of 2–10 min (using 100 ppm silane) resulted in porous SiN_x films, as evidenced by the formation of well oriented epitaxial GaN islands during the subsequent overgrowth. However, longer deposition times or use of more concentrated silane (e.g. 2% silane for 5 min) resulted in a polycrystalline GaN overlayer indicating complete coverage of the template surface with SiN_x (these overlayer crystallites perhaps nucleate at defects in the SiN_x overlayer, but in any case such misoriented GaN crystallites were not observed for the lower silane exposures). Growth of the GaN on top of the SiN_x layers was performed with the same conditions as for the GaN template layers, and for various growth times in order to observe the GaN overlayer at various stage of coalescence. The dislocation density and defect structure in the films were evaluated by plan-view and cross-sectional TEM, and by triple-axis x-ray diffraction (XRD).

3 Results and Discussion Figure 1(a) shows a plan view SEM image of a GaN film overgrown to an average thickness of ~1.5 μm on the GaN template layer, using a SiN_x interlayer, and with a sapphire substrate. The film has not grown uniformly over the template surface, but rather, hexagonal islands are observed. In contrast a film grown on the template layer without a SiN_x interlayer displays a featureless

Fig. 1 (a) and (b) Plan-view SEM images of a ~1.5 μm thick GaN film overgrown on a GaN template layer, with a SiN_x interlayer deposited on the template using 5 min silane exposure. (c) SAM map of the Si content of the same surface area as in (b).

Fig. 2 Cross-sectional TEM images of a $\sim 3 \mu\text{m}$ thick GaN film overgrown on a $\sim 3 \mu\text{m}$ thick GaN template, with a SiN_x interlayer (marked by white arrow) deposited on the template using 2 min silane exposure. (a) A pit ("p") arising from incomplete coalescence of the growth facets is seen at the corner of the image, and dislocations bend ("b") towards the walls (growth facets) of the pit. The SiN_x interlayer acts as a mask ("m") blocking the propagation of dislocations. (b) Some dislocations pass through the interlayer and extend straight into the overgrown film ("s"), and further bending of dislocations is seen ("b").

SEM image (not shown). From this result, together with the TEM images below, we attribute the islanding seen in Fig. 1(a) to nucleation on specific areas (i.e. inside the SiN_x pores on the exposed GaN template surface) and subsequent lateral growth along specific crystallographic directions, as observed by previous workers [3-6]. Figure 1(b) shows another SEM image of the same sample, and Fig. 1(c) displays scanning auger microscopy (SAM) data for that surface region. The white dots there represent the presence of Si on the surface and the black area represents the absence of Si. By comparing Figs. 1(b) and (c), it is clear that the area in between the hexagonal GaN islands is indeed covered by SiN_x .

Figure 2 shows cross-sectional TEM images of a nearly fully coalesced GaN film grown on a template layer, again using a sapphire substrate (for this film, the V/III ratio was reduced to 2000 in the latter 80% of the overgrowth in order to achieve nearly complete coalescence). It is clear from these images that the SiN_x interlayer significantly affects the structure of the dislocations in the overgrown film. Some surface pits were observed due to incomplete coalescence, one of which is marked by "p" in Fig. 2(a), and bending of dislocations (marked "b") towards the growth facets of these pits is observed. In addition, the SiN_x layer acts as a mask to block some dislocations (marked "m") from propagating into the overlayer. As seen in Fig. 2(b), although some dislocations pass through the SiN_x mask and extend straight into the overgrown film (marked "s"), most dislocations bend during growth of the islands. This bending of the dislocations increases their chances of combining and annihilating with each other, and therefore reduces the dislocation density near the top of the film. Thus, as in a conventional LEO process [1,2], in addition to bending the dislocations laterally the SiN_x film also blocks them from propagating from the template into the overlayer. XRD results for this sample for the (0002) and (10 $\bar{1}$ 2) rocking curve FWHM values are 5.3 and 5.8 arcmin, respectively, compared to 5.0 and 6.7 arcmin for control sample without a SiN_x interlayer. (It should be noted that these values integrate over the entire film thickness so they do not reflect the same degree of improvement as can be inferred from the TEM images).

Greater improvement in structural properties is found for growth on SiC substrates, as shown in Fig. 3. As described in section 2, a 100 nm thick GaN buffer layer is used and the SiN_x interlayer is deposited directly on that. A control sample, with no SiN_x interlayer, is shown in Fig. 3(a); a relatively high dislocation density in the buffer layer is seen, with some dislocation reduction occurring in the overlying GaN

film. Plan-view TEM images (not shown) reveal a dislocation density at the top of this GaN film of $6 \times 10^9 \text{ cm}^{-2}$. A sample containing a SiN_x interlayer is shown in Fig. 3(b). The overlayer is again nearly fully coalesced, with a few remaining surface pits seen in the image. A significantly lower dislocation density in the film is apparent compared to (a), with plan-view TEM images revealing a density of $7 \times 10^8 \text{ cm}^{-2}$. An improvement in structural quality is also evident in XRD measurements, with the (0002) and (10 $\bar{1}$ 2) rocking curve FWHM values being, respectively, 9.6 and 23 arcmin for the control sample and 8.4 and 13 arcmin for the sample with the SiN_x interlayer. Electron diffraction further reveals that the SiN_x interlayer induces no c-axis tilt in the GaN overlayers, a problem commonly observed in conventional LEO [13].

Fig. 3 Cross-sectional TEM images of $\sim 1.5 \mu\text{m}$ thick GaN films overgrown on a 100 nm thick GaN buffer layer, on SiC. The film shown in (a) was grown without an interlayer, and that in (b) is grown with a SiN_x interlayer (marked by white arrow) deposited using 10 min silane exposure. Surface pits ("p") observed in (b) indicate incomplete coalescence of the film

4 Conclusions We have studied the effect of using an in-situ deposited porous SiN_x on the dislocation density of the GaN overlayer. It was found that the GaN overgrowth starts inside the pores of the SiN_x film resulting in the formation of hexagonal islands with facets in specific crystallographic directions. Lateral growth of these islands causes the dislocations to bend normal to the facet walls, thus reducing the dislocation density near the top of the film. Many of the remaining dislocations near the top surface are clustered near the growth facet walls (surface pits), so that thicker overgrowth may produce further dislocation reduction through combination and annihilation. Like in a conventional LEO process, the porous SiN_x layer is also found to be effective in blocking (or masking) dislocations from propagating from the template up into the overlayer. The dislocation density in GaN films grown on porous SiN_x interlayers is found to be reduced by an order of magnitude compared to those for films grown without an interlayer.

Acknowledgements We thank Dr. Wayne Jennings of Case Western Reserve University for his assistance in acquiring SAM data. This work was supported by a Defense University Research Initiative on Nanotechnology (DURINT) program administered by the Office of Naval Research under grant N00014-01-1-0715 (program monitor Colin Wood).

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] T. S. Zheleva, O.-H. Nam, M. D. Bremser, and R. F. Davis, Appl. Phys. Lett. **71**, 2472 (1997).
- [3] A. Sakai, H. Sunakawa, and A. Usui, Appl. Phys. Lett. **71**, 2259 (1997).
- [4] H. Lahrèche, P. Vennéguès, B. Beaumont, and P. Gibart, J. Cryst. Growth **205**, 245 (1999).
- [5] E. Frayssinet, B. Beaumont, J. P. Faurie, P. Gibart, Zs. Makkai, B. Pécz, P. Lefebvre, and P. Valvin, MRS Internet J. Nitride Semicond. Res. **7**, 8 (2002).
- [6] X. L. Fang, Y. Q. Wang, H. Meidia, and S. Mahajan, Appl. Phys. Lett. **84**, 484 (2004).
- [7] K. Pakula, R. Bozek, J. M. Baranowski, J. Jasinski, and Z. Liliental-Weber, J. Cryst. Growth **267**, 1 (2004).
- [8] T. Wang, Y. Morishima, N. Naoi, and S. Sakai, J. Cryst. Growth **213**, 188 (2000).
- [9] A. Dadgar, M. Poschenriede, A. Reiher, J. Bläsing, J. Christen, A. Krtschil, T. Finger, T. Hempel, A. Diez, and A. Krost, Appl. Phys. Lett. **82**, 28 (2003).
- [10] S. Tanaka, S. Iwai, and Y. Aoyagi, Appl. Phys. Lett. **69**, 4096 (1996).
- [11] S. Tanaka, M. Takeuchi, and Y. Aoyagi, Jpn. J. Appl. Phys. Part 2, **39**, L831 (2000).
- [12] V. Ramachandran, M. F. Brady, A. R. Smith, R. M. Feenstra, and D. W. Greve, J. Electron. Mater. **27**, 308 (1998).
- [13] R. F. Davis, A. M. Roskowski, E. A. Preble, J. S. Speck, B. Heying, J. A. Freitas, E. R. Glaser, and W. E. Carlos, Proc. IEEE **90**, 993 (2002), and references therein.