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# A transmission electron microscopy investigation of buried defect sources within epitaxial GaN

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**Abstract.** Site-specific focused ion beam milling procedures are used to prepare cross-sectional TEM sample foils through a range of epitaxial GaN thin films. The high threading defect densities associated with heteroepitaxial GaN/sapphire prohibit the unambiguous identification of hillock nucleation events. Inversion domains are, however, identified at the core of hillocks formed on CVD grown GaN on N-polar bulk GaN substrates. It is considered that remnant contamination from the KOH chemomechanical polishing procedure is responsible for the inversion domain nucleation.

## 1. Introduction

There is continued need to gain improved understanding of the nature of crystallographic defects within semiconductors, e.g. to ascertain the fine scale association of structural defects with material (opto-) electronic recombination activity, or to identify and thereby control (or eliminate) the reasons for defect nucleation. Indeed, complete understanding of the nature of structural defects within GaN can only come about from a combined structural, optical and electronic study of defects combined with theoretical calculation. Generally one approaches the nucleation question through the interpretation of static projected images of discrete defects whilst seeking to gain insight into the dynamics of layer evolution during growth. Accordingly, models for defect propagation and interaction, e.g. multiplication or annihilation mechanisms, can be constructed from consideration of the crystallographic nature of the defect, the crystal slip system and the likely defect interactions that may occur. However, the process of defect nucleation is often inferred, simply because direct, unambiguous evidence for some localized perturbation responsible for the nucleation of an extended defect is very difficult to obtain. Nevertheless, localized strain fields arising from compositional fluctuations have been unambiguously identified as being responsible for misfit dislocation introduction within relatively low lattice mismatched systems such as SiGe/Si [1] and InGaAs/GaAs [2]. Further, x-ray topography has been used to identify the location of specific sources, thereby facilitating the controlled plan view TEM sample preparation through such nucleation events [3, 4]. Debate still continues about the precise nature of nucleation events within GaN-based heteroepitaxial systems, but such consideration becomes difficult in view of the extremely high density of threading defects. The advent of focused ion beam (FIB) technology, however, now makes 'needle in haystack' type problems more accessible provided sufficient reference information is

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available to identify the presence of a sub-surface defect and its associated nucleation event from the sample surface topography. In particular, homoepitaxial GaN constitutes an interesting model system to examine threading defects in isolation. Here, we comment on how FIB milling procedures can provide useful site-specific information on the nature of defects and defect sources within epitaxial GaN.

## 2. Experiment

Details of the molecular beam epitaxy (MBE) and chemical vapour deposition (CVD) procedures used to deposit the epitaxial GaN films on sapphire or bulk GaN substrate surfaces examined here have been described previously [5, 6]. Bulk GaN substrates were grown under high hydrostatic pressure of nitrogen (15–20 kbar) from liquid Ga at 1600 °C [7]. Prior to homoepitaxial growth, the bulk GaN{0001} platelets were mechanically polished using 0.1 $\mu$m diamond paste. GaN layers grown on N-polar surfaces that had been chemomechanically polished using aqueous KOH were examined and compared with those that subsequently received a short deoxidizing procedure using aqueous NaCl prior to growth. Similarly, GaN layers grown on mechanically polished Ga-polar substrates were compared with samples grown on substrates exposed to a subsequent Ar–$SiCl_4$–$SF_6$ reactive ion etch [8]. CVD growth of GaN/sapphire and homoepitaxial GaN was performed at 1050 °C under an $N_2/NH_3$ atmosphere using tri-methylgallium and $NH_3$, with $H_2$ as the carrier gas. Some of the CVD layers examined were subsequently treated with a defect revealing etchant following growth, based on $H_3PO_4$:$H_2SO_4$ at 60 °C for 2 minutes, for the purpose of correlating etch profile with sub-surface defect microstructure, as detailed elsewhere [9].

Cross-sectional TEM samples were prepared using focused ion beam milling procedures. Samples were initially coated with gold prior to insertion into the FIB chamber, then regions chosen for sectioning coated with thick platinum strips by the decomposition of a platinum alkyl under the rastered Ga beam. This provided protection for the samples during subsequent exposure to the high Ga flux used for milling. For the case of site-specific sample preparation, e.g. through the emergent core of a chosen growth hillock, vertical sections were prepared through the hillock apices with a view to isolating the associated nucleation event, presumed to be located at the epilayer/substrate interface a few micrometres below the surface.

## 3. Results and discussion

Figure 1 shows a section through an MBE GaN/sapphire sample grown under non-optimized conditions. The image is selected simply to illustrate the high density and diverse range of defects within heteroepitaxial GaN/sapphire. Growth columns containing a high density of basal plane stacking faults delineated by an equally high density of threading defects predominate, with the addition of surface faceting. Consequently, the isolation and unambiguous identification of discrete nucleation events within such a complex microstructure is not an option.

Both CVD and MBE grown homoepitaxial and heteroepitaxial GaN layers can in fact show very high degrees of surface planarization, mediated by a step-flow growth. Thus, surface features providing clue to some sub-surface perturbation responsible for defect formation are generally limited to growth hillocks, surface pits or maybe gross embedded inclusions. By way of example, homoepitaxial GaN grown by CVD on N-polar substrates that had been exposed to a KOH etch prior to growth exhibited a non-uniform density of 0.5–2 $\times$ $10^5$ $cm^{-2}$ of hexagonal pointed pyramids and flat topped hillocks [5]. Plan view TEM imaging, following


**Figure 1.** TEM image illustrating the high density and diverse range of dislocations, domain boundaries, basal plane stacking faults and surface facets that can form within MBE grown GaN/sapphire.

conventional sequential mechanical polishing and ion beam thinning, indicated the presence of columns of material approximately 100 nm across at each these hillock cores [10], with the apparent absence of any other defect. Thus, it becomes of interest to establish the nature and origin of such defect cores.

Figures 2(a)–(d) show a sequence of secondary electron images illustrating the procedure used to section through the emergent core of a growth hillock. A CVD grown GaN/sapphire sample is shown in this instance, and this particular sample has been etched using $H_3PO_4$:$H_2SO_4$ prior to examination, hence the presence of surface irregularities. Sectioning through to the location of the nucleation site of the defect, presumed to be located at the line of the original epilayer/substrate vertically below the sample surface, places a very strict set of limitations on the sample orientation during FIB milling. By way of example, establishing a $<$100 nm electron transparent membrane through a buried interface $\sim$3 $\mu$m below the sample surface necessitates milling at an angle $<1^\circ$ off the normal to the growth plane, using the emergent defect as reference, otherwise one would completely miss the buried nucleation event. Even though FIB procedures are very accurate at locating and rotating about chosen surface features, there is no optimum procedure for setting up the beam normal to the growth surface other than orienting visually using facets at the sample edge as reference. Consequently, the likelihood of routinely intersecting a sub-surface nucleation site becomes significantly diminished, particularly if one wishes to section a foil thin enough for high-resolution chemical microanalysis. Accordingly, platinum stripes were layered over the hillock facet edges, in addition to a column of platinum deposited at the stripe intersection (figure 2(b)) in order to retain sight of the precise position of the emergent hillock core during FIB sectioning. Rapid milling using high Ga flux was then used to define a broad access trench (figure 2(c)), with successively decreasing conditions of flux being used to delineate the final electron transparent membrane (figure 2(d)) in the approximate location of the defect core. Figure 3 shows a cross-sectional TEM image through this particular CVD grown, etched GaN/sapphire sample hillock and immediately reveals the extremely high density of threading dislocations and

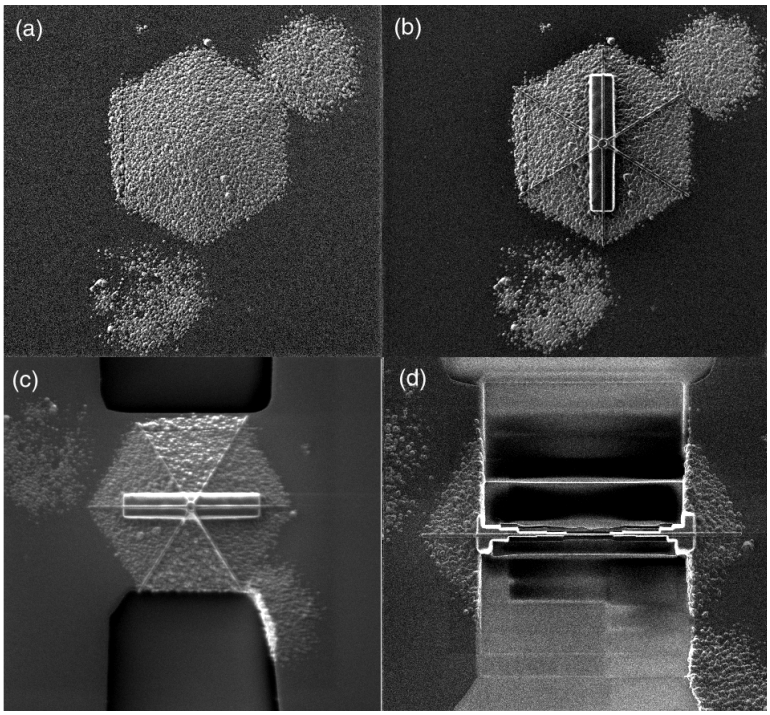


**Figure 2.** Secondary electron images showing the focused ion beam workstation procedure used to target buried defect sources beneath the emergent defect core of a growth hillock. (a) An etched CVD grown GaN/sapphire hillock (∼5 $\mu$m in size). (b) Platinum stripes are deposited along the hillock facet edges to retain sight of the defect core (∼100 nm in size). (c) High Ga flux is used to create an access trench to the defect. (d) Decreasing Ga flux is used to fashion the electron transparent membrane at the approximate position of the defect core.

domain boundaries that prohibit definitive identification of the particular defect and associated nucleation event specific to the hillock core in this instance. Further, no definitive statement can be made about the association of threading defects with the form of the surface profile.

Some measure of progress, however, was gained by sectioning through the core of a surface hillock of a homoepitaxial GaN sample grown by CVD on a bulk N-polar GaN substrate (figure 4). A columnar defect located below the apex of the hillock core is unambiguously identified as being an inversion domain, as evidence by convergent beam electron diffraction patterns (inset) acquired within and to the side of the defect shown. Image rotation correction of the electron microscope using a polarity calibrated sample confirmed that the matrix indeed exhibited an N-polar growth surface, whilst the emergent defect core displayed a Ga-polar growth surface. The inversion domain tapers off as less material is incorporated within the sample foil. Thus, the nucleation event responsible for the formation of this particular defect lies somewhere outside the plane of the defect foil, i.e. has now been sectioned away, effectively illustrating the predominant practical constraint arising from slight sample misorientation when sectioning for specific buried nucleation sites. This specimen had also been etched prior to examination, hence the faceted surface profile. However, no apparent structural defects were found to be associated with the facet steps.

The consideration of slight sample misorientation during FIB milling is further illustrated with reference to figure 5, showing a cross-sectional weak beam TEM image through a hillock

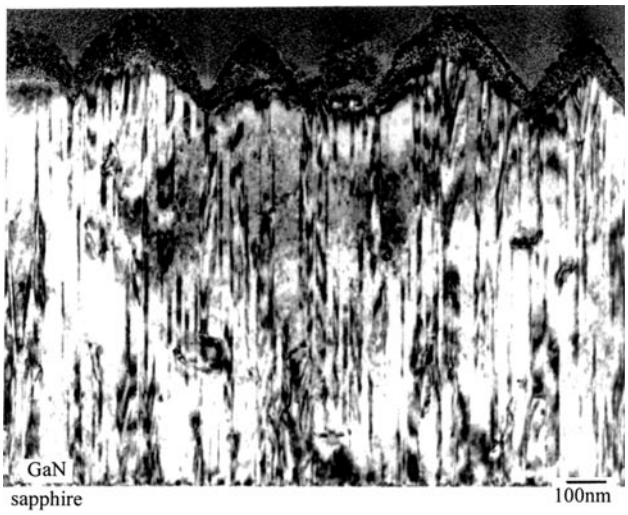


**Figure 3.** TEM image through the CVD grown GaN/sapphire sample sectioned in figure 2. The high threading defect density effectively prohibits unambiguous determination of the defect structure responsible for the hillock core.

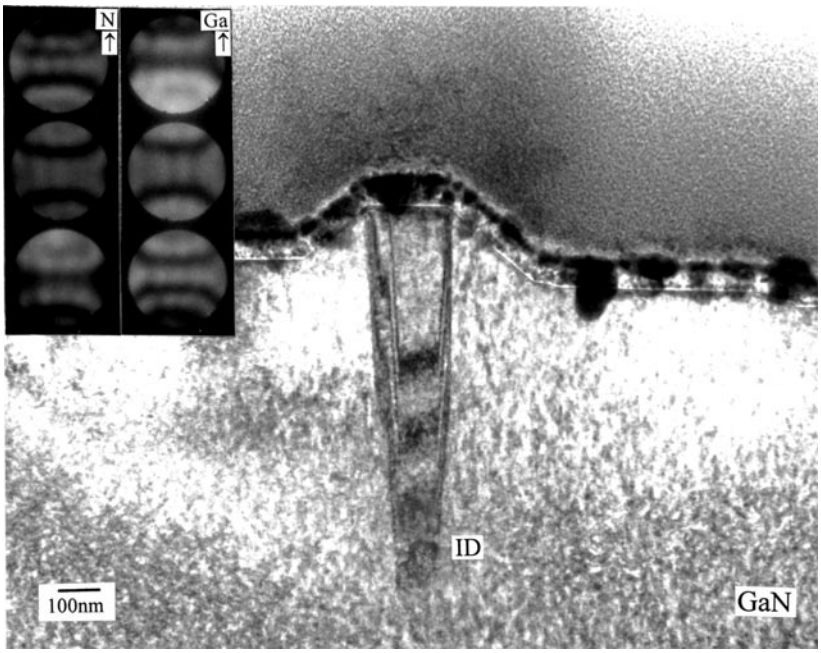


**Figure 4.** TEM image through the centre of a point-topped hillock (homoepitaxial CVD grown GaN on an N-polar GaN substrate chemomechanically polished using KOH prior to growth). The inset CBED patterns demonstrate reversal of contrast of the {0002} diffraction discs when moving across the defect boundary plane. Thus, the defect is an inversion domain with a Ga-polar growth surface emerging from the N-polar matrix.

core of homoepitaxial GaN on an N-polar substrate. Tapering fringe contrast due to the inversion domain is indicated along with the perceived line of the original epilayer/substrate

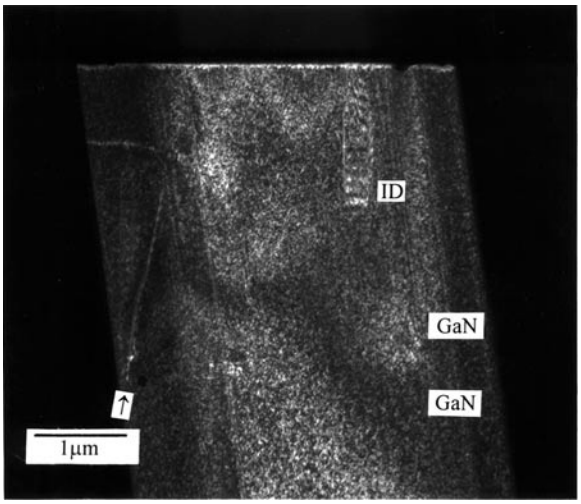


**Figure 5.** Weak beam TEM image through the centre of a point-topped hillock (homoepitaxial GaN grown on N-polar GaN) revealing the presence of an inversion domain plus an additional stacking disorder (arrowed) emanating from the line of the original epilayer/substrate interface.

interface ∼2.8 $\mu$m below the surface. Additional defects threading the layer (as arrowed) are occasionally found to be associated with such hillock microstructures. Unfortunately, the restricted sample tilt imposed by the nature of the FIB trench leading up to the electron transparent membrane prohibits large sample tilt and hence definitive diffraction contrast analysis of such defects. However, it is noted that they exhibit fringes under certain diffraction conditions and thus are presumed to be some non-prismatic plane stacking disorder. It is considered that such features, when emergent at the growth surface, correlate with the morphological perturbations exhibited by certain hexagonal pyramids (e.g. figure 1(b) in [5]).

Figure 6 shows a TEM image through an inversion domain located at a hillock core complete with a nucleation event. The lower (∼0.9 $\mu$m) sample thickness in this instance assisted greatly with the isolation of this defect, providing leeway of up to 3° misorientation between sample normal and milling Ga beam. The striped nature of the projected image is simply due to the form of the embedded hexagonal faceted structure within the GaN matrix. The fine detailed analysis of this particular nucleation event using complementary HREM and EELS techniques has previously been described [10, 11], necessitating thinning of the sample even further to isolate the material comprising the nucleating event free of the surrounding host matrix. The source of this inversion domain was thus found to comprise a narrow band 2–5 nm thick of amorphous oxygen-containing compound, believed to be gallium hydroxide remnant from the KOH chemomechanical polishing procedure [5, 10]. This is distinct from some well defined crystallographic event related to $GaO_2$ or $Ga_2O_3$ interlayers that are candidate structures for inversion domain nucleating events. A dramatic reduction of these remnant hydroxide deposits following etching in molten NaCl was subsequently demonstrated leading to the growth of near hillock free N-polar homoepitaxial layers [5]. TEM sections through such sample foils demonstrated the complete absence of any threading structural defects and further failed to reveal the line of the original epilayer/substrate interface. Present within figure 6 is another irregular stacking disorder (arrowed) similar to that shown in figure 5, again loosely associated with this hillock microstructure, the suggestion being that strain fields arising from the embedded inclusions during growth may be responsible for the initiation of these features and thus subsequent perturbation of the hillock surface morphology.

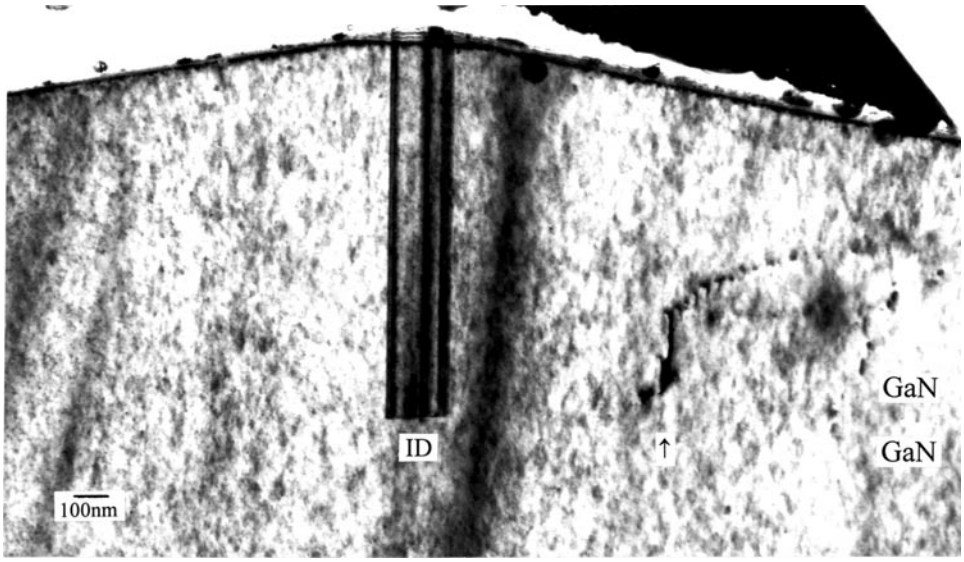


**Figure 6.** TEM image through the centre of a point-topped hillock (homoepitaxial GaN grown on N-polar GaN). An inversion domain complete with nucleation source considered to originate at the line of the original epilayer/substrate interface is shown. An additional stacking disorder is also present (arrowed).

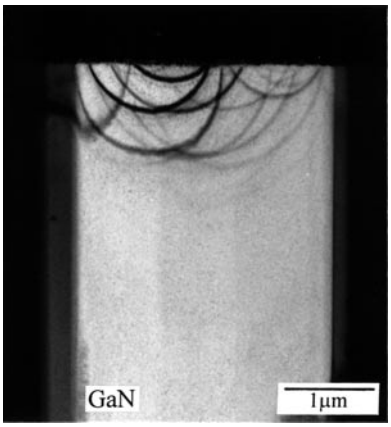


**Figure 7.** TEM image through the centre of a flat-topped hillock (homoepitaxial GaN grown on N-polar GaN) demonstrating the complete absence of threading defects and with no remnant contrast from the line of the original epilayer/substrate interface. The strain contrast at the top of the image arises from the Pt/Au protective coating.

TEM sectioning through the perceived centre of a flat topped hillock of homoepitaxial GaN grown onto an N-polar GaN substrate similarly failed to reveal any sub-surface defects (figure 7). Near surface strain contrast is apparent due to the effect of the Pt/Au protective coating, but there are no discrete defects evident within the layer itself. Further, no line of the original epilayer/substrate interface could be discerned. The suggestion is that flat-topped hillocks are formed by the overgrowth of N-polar matrix material onto a hillock originally defined by the presence of an inversion domain [5]. Locating such buried defects without any

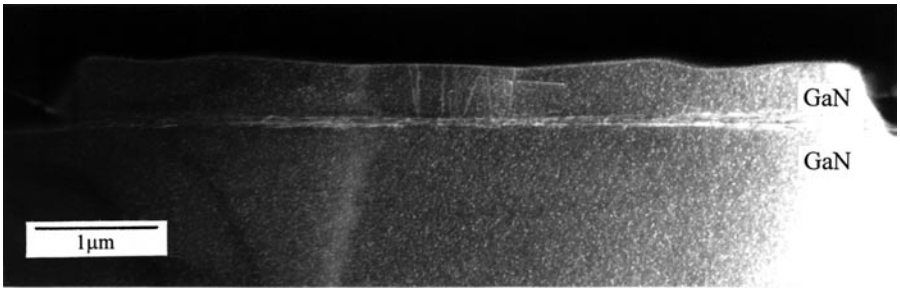


**Figure 8.** TEM image (homoepitaxial GaN grown on N-polar GaN) through a complete hillock that had formed at a surface scratch. A band of threading screw and edge dislocations at the centre mediate hillock growth.

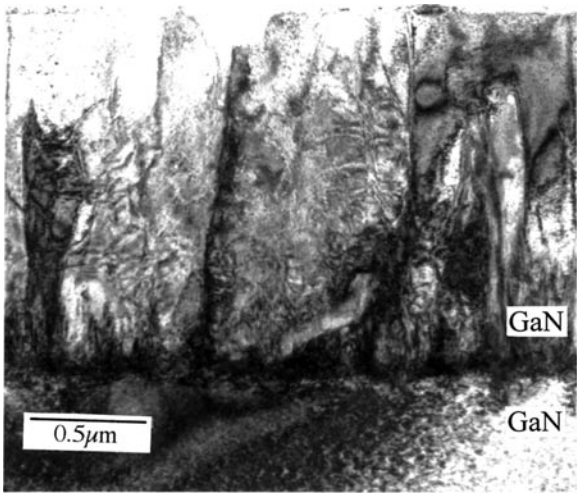


**Figure 9.** TEM image (homoepitaxial GaN grown on mechanically polished Ga-polar GaN) revealing a high density of threading dislocations and the association of a grain boundary with a flat-bottomed pit.

definitive clue to their presence from the surface topography, however, is likely to constitute quite a challenge.

Further insight into the modes of layer growth of CVD homoepitaxial GaN on N-polar substrates is demonstrated by figure 8, which shows the presence of a narrow band of dislocations at the centre of a discrete growth hillock that had nucleated at a surface scratch. The slight undulation of the surface profile of this particular hillock is merely an artefact of the FIB sample preparation process since the hillock surface was briefly exposed to high Ga flux prior to coating and sectioning. However, both screw and edge type threading dislocations are identified within the bundle at the hillock core and thus it is considered that these defects when emergent at the growth surface are associated with a step-flow mechanism for continued hillock growth [5].

The TEM assessment of homoepitaxial GaN samples grown onto Ga-polar substrates that had only received a mechanical polishing procedure prior to growth revealed a very high density of threading dislocations bounding a well defined columnar grain structure (figure 9). The presence of a grain boundary below a flat-bottomed pit was also observed, but with no evidence for presence of any inversion domain. It is considered that the Ga-polar surface is

more stable (i.e. faster growth rate/slower desorption rate) than the N-polar surface for the growth conditions used [5, 10]. Thus, inversion domains, once established, freely propagate for the case of N-polar growth, pulling up the hillock tentlike structures around them, while they do not become established for the case of Ga-polar homoepitaxy. TEM examination of a homoepitaxial GaN sample grown onto a Ga-polar substrate that had been subjected to an Ar–$SiCl_4$–$SF_6$ reactive ion etch following substrate mechanical polishing again failed to reveal the presence of any extended structural defects [8], in a manner similar to that shown in figure 7.

## 4. Summary

A range of GaN/sapphire and homoepitaxial GaN samples has been sectioned for TEM investigation using site-specific FIB procedures. The extremely high density of defects associated with heteroepitaxial GaN hinders the unambiguous identification of discrete nucleation events. However, careful sample orientation has enabled buried defect sources within homoepitaxial GaN grown on N-polar substrates to be isolated and characterized. Platelets of hydroxide $\sim$100 nm in size remnant from the KOH polishing procedure are considered to be responsible for the formation of inversion domains and hence growth hillocks within this system. Effective elimination of these defects was achieved by applying a deoxidizing aqueous NaCl etchant to these N-polar substrates prior to growth. A high density of threading dislocations was found to propagate through GaN layers grown on mechanically polished Ga-polar GaN substrates. These defects were effectively eliminated by applying an Ar–$SiCl_4$–$SF_6$ reactive ion etch procedure to the substrates prior to growth.

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