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Inversion domain nucleation in homoepitaxial GaN

P.D. Brown[1*], J.L. Weyher[2,3], C.B. Boothroyd[1], D.T. Foord[1], A.R.A. Zauner[2], P.R. Hageman[2], P.K. Larsen[2], M. Bockowski[3] and C.J. Humphreys[1].

[1]Department of Materials Science and Metallurgy, University of Cambridge, Pembroke Street, Cambridge, CB2 3QZ, UK.
[2]University of Nijmegen - RIM, Exp. Solid State Physics III, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands.
[3]High Pressure Research Centre, Polish Academy of Sciences, ul. Sokolowska 29, 01-141 Warsaw, Poland.

ABSTRACT: Homoepitaxial GaN grown by metal organic chemical vapour deposition on the N-polar surface of bulk GaN commonly exhibits gross hexagonal pyramidal features, typically 5 to 50μm in size, depending on layer thickness. The evolution of these defects is dominated by the growth rate of an emergent core of inversion domain (typically 100nm in size). The associated nucleation events are considered to be thin bands of amorphous gallium hydroxide (2 to 5nm in thickness) remnant from the standard KOH chemo-mechanical substrate polishing procedure used.

1. INTRODUCTION

Interest in homoepitaxial GaN continues, driven by its potential for high power optoelectronic device applications. In this context, the absence of microstructural defects afforded by homoepitaxial growth is considered beneficial, particularly in view of recent definitive evidence confirming that dislocations do indeed exhibit non-radiative recombinative properties (e.g. Sugahara et al, 1997). However, metal organic chemical vapour deposition (MOCVD) grown homoepitaxial GaN on chemo-mechanically polished (000$\bar{1}$), N-polar, substrates commonly exhibit gross hexagonal shaped hillocks that could be somewhat problematic for subsequent device processing. Here we use a range of microscopical techniques to characterise the nature of the cores of these hillock structures and the precise form of their associated nucleation events.

2. EXPERIMENTAL

The homoepitaxial GaN samples examined in this study were grown by MOCVD at 1050°C as detailed elsewhere (De Theije et al, 1999, Weyher et al, 1999a). The bulk GaN substrate material was grown under a high hydrostatic pressure of nitrogen (15 – 20kbar) from liquid Ga at 1600°C (Porowski, 1996). Prior to growth, the (000$\bar{1}$) surfaces were mechanically polished using 0.1μm diamond paste and then chemo-mechanically polished in an aqueous KOH solution (Weyher et al, 1997). Epitaxial growth was performed using trimethylgallium and NH_3 precursors with H_2 as the carrier gas, under a total pressure of 50mbar. Electron transparent samples were prepared in plan-view using conventional sequential mechanical polishing and ion beam thinning procedures, whilst cross-sectional samples were prepared using a Ga-source focused ion beam (FIB) workstation. The site

* Now at the Division of Materials Engineering and Materials Design, School of Mechanical, Materials, Manufacturing Engineering and Management, University of Nottingham, University Park, Nottingham, NG7 2RD.

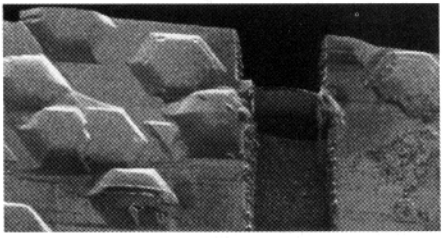

Fig. 1 Secondary electron image of homoepitaxial GaN/GaN(000$\bar{1}$) with an FIB prepared membrane through the core of one of the growth hillocks.

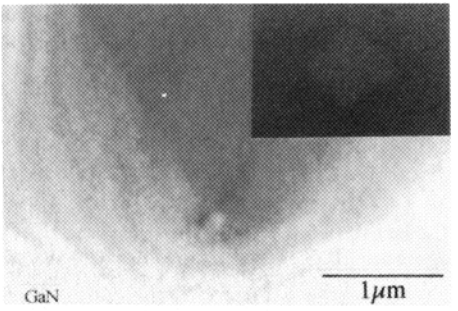

Fig. 2 Slightly tilted plan-view TEM image through a growth hillock revealing the central defect core. Enlarged <0001> image of hillock core inset.

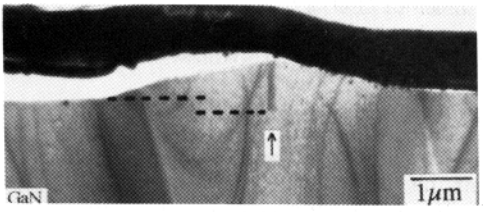

Fig. 3 Low magnification cross-sectional TEM image showing a growth hillock in profile (defect core arrowed).

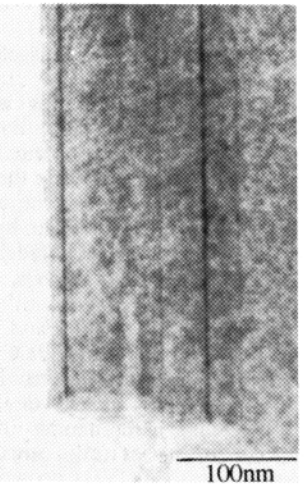

Fig. 4 TEM image showing the faceted structure of a hillock core in cross-section.

selectivity of the FIB technique enabled cross-sections through the emergent cores of the hillocks to be obtained, thereby allowing the nucleation events to be isolated and characterised.

3. RESULTS AND DISCUSSION

Fig. 1 shows a secondary electron image of the homoepitaxial GaN/GaN(000$\bar{1}$) growth hillocks recorded during FIB cross-sectional sample preparation. The hillocks are typically 5 to 50μm in size depending on layer thickness (time of growth). When prepared in plan-view geometry (Fig. 2), such hillocks exhibit a small faceted core structure at the centre, but otherwise the layers are generally found to be defect free. Fig. 3 shows a low magnification image of one such growth hillock from a very thin layer imaged in cross-section. The protective Pt-stripe deposited on a thin Au gold coating layer prior to FIB milling has become detached from the top surface of the sample and slightly obscures the hillock. Nevertheless, the hillock core structure can clearly be seen (arrowed). It is presumed that the defect originates at the line of the original epilayer / substrate interface since no other contrast delineating the region of this interface could be discerned. The hillock structure is enlarged and slightly tilted in Fig. 4 to emphasise its faceted structure. The core emanates from a thin platelet nucleus that is only ~100nm in size. Convergent beam electron diffraction (CBED) analysis confirmed the N-polar growth surface of the surrounding matrix material, but no clear CBED patterns could be obtained from this particular core structure since it was embedded within matrix material.

Fig. 5 shows a cross-section through a core structure from a thicker layer, although the nucleating event has been sectioned away during sample foil preparation in this instance. In

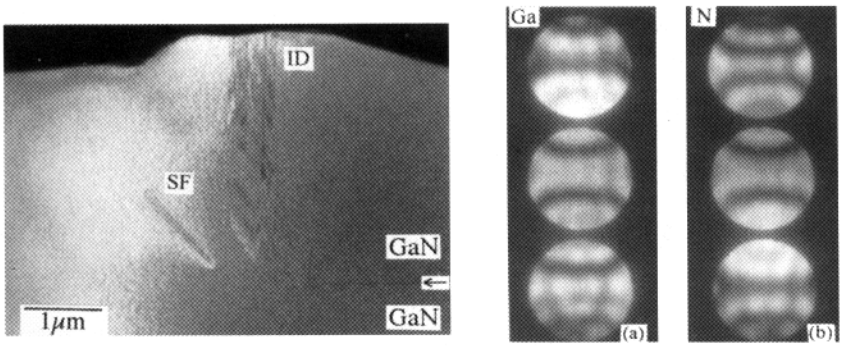

Fig. 5 Thinner section through a hillock core with two associated CBED patterns (a,b) recorded either side of the core boundary, corresponding to core and matrix material respectively. The reversal in contrast within the {0002} diffraction discs confirms the defect to be an inversion domain.

addition to the central core there is also an inclined stacking fault and a horizontal screw dislocation (arrowed) which, again, presumably delineates the line of the original epilayer / substrate interface. CBED patterns recorded either side of the boundary wall of the core immediately reveal a reversal of the contrast within the {0002} diffraction discs and as such confirm the defect core to be an inversion domain (ID) (e.g. Cherns et al, 1998). Correction for image rotation introduced by the electron microscope (using a polarity calibrated sample) demonstrates that the central core has a Ga-polar growth surface, while the surrounding matrix has a N-polar growth surface, commensurate with the polarity of the substrate used for growth. When compared with Fig. 3, it is clear that the cores of the inversion domains have a much higher growth rate (approx. x3) than the surrounding N-polar matrix, leading to the production of the 'circus-tent' hillock structures around them. Competition between the growth and desorption rates of Ga and N-polar surfaces allows the gross hexagonal pyramids to evolve. Interestingly, the sense of Ga-polar surface having a faster growth rate than the N-polar surface is contrary to that found for thin film growth of wurtzite CdS, where the S-polar surface has a higher growth rate than the Cd-polar surface (Halsall et al, 1988). The implication, therefore, is that the N-polar surface has a much higher desorption rate than the Ga-polar surface for the MOCVD growth conditions used here.

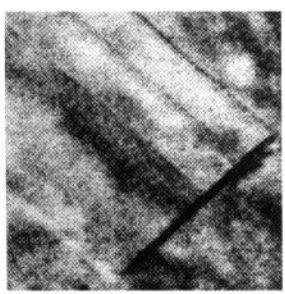

Fig. 6 HAADF image, confirming the presence of low atomic number material at the ID source.

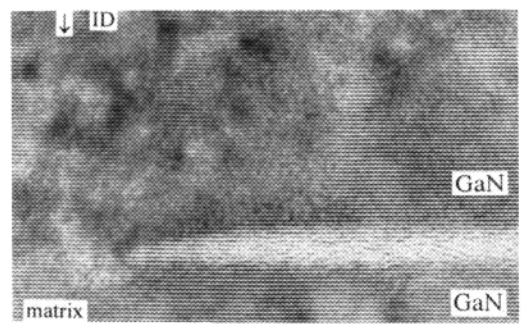

Fig. 7 HREM image confirming the presence of a narrow band of amorphous material at the ID source.

High angle annular dark field imaging (HAADF) imaging (Fig. 6) of the sample shown in Figs. 3,4, tilted slightly off a systematic row orientation to minimise the effects of diffraction contrast, shows dark contrast at the position of the inversion domain, and therefore confirms the presence of a low atomic number material associated with the nucleation event. Preparing samples thin enough for high resolution electron microscopy (HREM) confirmed the nature of such nucleation events to be narrow bands of amorphous material, 2 to 5nm in thickness (Fig. 7). Electron energy loss spectroscopy (EELS) confirms the presence of

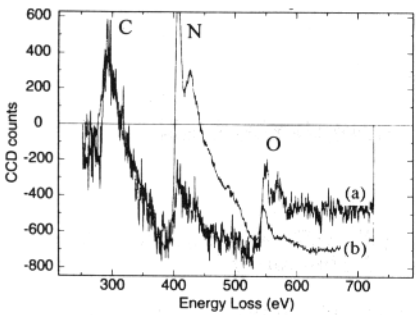

Fig. 8 EEL spectra confirming the presence of oxygen at the nucleating event. (a) Nucleation event. (b) Matrix.

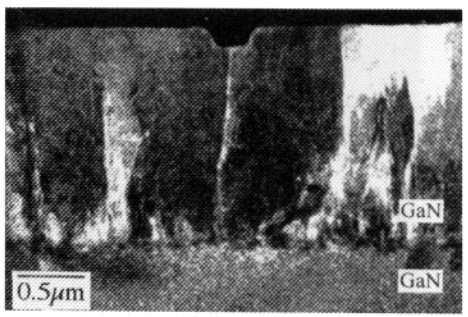

Fig. 9 Homoepitaxial GaN grown on a Ga-polar GaN substrate, showing the association of a grain boundary with a surface flat-bottomed pit.

oxygen (Fig. 8) within these narrow bands of amorphous material, and as such these features are attributed to remnant contamination from the chemo-mechanical polishing technique used to prepare the substrates prior to growth. The oxygen-containing residue is presumed to be gallium hydroxide, a likely reaction product of the KOH etchant with GaN. An improved surface preparation method incorporating a short deoxidising polishing procedure in an aqueous solution of NaCl (Ohkubo, 1998) led to a dramatic reduction of these nucleation sources and thus allowed N-polar homoepitaxial GaN films to be grown largely free of these gross hillock structures (Weyher et al, 1999a).

Preparation of (0001)Ga-polar substrates suitable for homoepitaxy proved to be a difficult problem since chemo-mechanical polishing procedures were found not to be effective. Thus, substrates were mechanically polished with 0.1μm diamond paste and received a reactive ion etching (RIE) treatment prior to growth. Such samples were found to contain a very high density of threading dislocations, with FIB prepared cross-sectional samples showing the association of grain boundaries, rather than inversion domains, with the location of flat bottomed surface pits (e.g. Fig. 9). Such highly faulted layers were again attributed to remnant surface roughness and contamination after the substrate preparation process. However, improved RIE procedures have recently enabled the preparation of GaN homoepitaxial layers on Ga-polar surfaces free of structural defects, and this will be reported on elsewhere (Weyher et al, 1999b).

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