Title
Polarity determination of a-plane GaN on r-plane sapphire and its effects on lateral overgrowth and heteroepitaxy

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Authors
Wu, F
Craven, M D
Lim, S H
et al.

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I. INTRODUCTION

Despite the advances being made with nitride-based semiconductor devices, current state-of-the-art device structures are ultimately affected by polarization-induced electrostatic fields since these structures are grown along the polar c-axis of the wurtzite crystal structure. Polarization discontinuities along the growth direction generate fixed sheet charges at interfaces and surfaces which, in turn, produce electric fields which affect the operation of III-N-based devices. For example, internal fields are responsible for the large sheet charge densities present in AlGaN/GaN heterostructures and the quantum confined Stark effect observed in GaN/AlGaN and InGaN/(In)GaN quantum well structures. The most direct means of eliminating internal field effects in the nitrides is through the growth of wurtzite device structures with nonpolar orientations or zinc blende device structures along a ⟨001⟩ direction. The metastability of the zinc blende phase of nitrides creates challenging issues for consistent and reproducible device growth. The most attractive means, therefore, of producing internal field-free nitride device structures is the growth of nonpolar wurtzite GaN.

Recently, we characterized (11̄0) a-GaN films heteroepitaxially grown on (1̄10̄2) r-plane sapphire substrates and the subsequent lateral overgrowth of these films (see Refs. 11 and 12). In order to explicitly define the film/substrate epitaxial relationship and to accurately describe growth features and surfaces, the polarity of the nonpolar GaN film must be determined since the GaN c-axis lies in the growth plane, as shown schematically in Fig. 1. In this article we describe the convergent beam electron diffraction (CBED) experiments used to determine the polarity of a-GaN grown on r-sapphire. CBED is a well-established method which has been widely used to determine the polarity of polar c-plane GaN films. The results of the CBED experiments performed here are used to accurately index crystallographic growth features and surfaces observed in nonpolar GaN heteroepitaxial films and lateral overgrowth samples. Once identified, we can develop a general understanding of how polarity affects nonpolar GaN growth.

In particular, polarity effects are most clearly observed in the lateral overgrowth characteristics of a-GaN films. The heteroepitaxial a-GaN films have proven to be excellent candidates for dislocation reduction via lateral epitaxial overgrowth (LEO) since their microstructure consistently contains a large density of threading dislocations (≈2.6 × 10¹⁰ cm⁻²) with line directions parallel to the [11̄0] growth direction. Stacking faults also appear in the a-GaN microstructure with a fault density of ≈3.5 × 10^⁸ cm⁻¹. Polarity dictates the lateral overgrowth from rectangular mask openings aligned along [11̄0]GaN (perpendicular to the c axis) since the stripe sidewalls are c-planes with opposing surface polarities. Note that convention identifies the (0001) surface (+c-plane) as the Ga-face and the (000̄1) surface (−c-plane) as the N-face. Additionally, polarity affects the formation of large-scale, crystallographic pits in heteroepitaxial a-GaN films grown under nonoptimized conditions. These surface pits contain two inclined facets that terminate at a c-plane facet with a common polarity.

II. EXPERIMENT

The polarity of an uncoalesced, laterally overgrown nonpolar GaN sample grown via metalorganic chemical vapor deposition (MOCVD) was determined using CBED. Convergent beam electron diffraction analysis was used to determine the a-GaN polarity to explicitly define the film/substrate relationship, and subsequently to identify various growth features and surfaces observed throughout our studies of a-plane GaN. In particular, the effects of polarity on (1) lateral overgrowth from mask stripe openings aligned along [1̄10̄0]GaN and (2) pit formation in heteroepitaxial films grown under nonoptimized conditions were investigated. The fundamental differences between the polar surfaces are clearly observed; analysis of the lateral epitaxial overgrowth stripes revealed that (0001) surfaces grew faster than (0001̄) surfaces by approximately an order of magnitude, and these stable, slow-growing (0001̄) surfaces are a likely cause of pitting in a-GaN films. The growth features under investigation were imaged using scanning and transmission electron microscopy. © 2003 American Institute of Physics.

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deposition (MOCVD) was determined using CBED. The conventional LEO technique employed here involves two growth steps and one intermediate processing step. Initially, a specular (1120) α-plane GaN film was heteroepitaxially grown on an (1012) r-plane sapphire substrate using a low temperature GaN nucleation layer prior to high temperature epitaxial GaN growth (see Ref. 11). The as-grown nonpolar GaN film was coated with ~200 nm of SiO₂ which was subsequently patterned with parallel 5 μm wide mask openings (windows) spaced 15 μm apart and crystallographically aligned parallel to [1100]GaN, perpendicular to the c-axis. After patterning the mask, the sample was subjected to brief MOCVD regrowth using the same growth conditions as those used for the high temperature heteroepitaxial growth.12 During selective epitaxial regrowth, the GaN grows vertically through the mask windows and laterally over the mask, however, the regrowth was kept brief so that coalescence of adjacent stripes was avoided and the uncoalesced stripe morphology could be studied. We employ specific GaN crystallographic indexing throughout this article with the (1120) GaN growth surface as the reference index due to the reduced symmetry of the nonpolar GaN surface.

The morphology and microstructure of the as-grown heteroepitaxial GaN film and subsequent LEO samples were observed using a JEOL 6340F scanning electron microscope (SEM) operated at 5 kV and a JEOL 2000FX transmission electron microscope (TEM) operated at 200 kV, respectively. As-cleaved sample cross sections were imaged for morphology analysis while cross-sectional foils were prepared for TEM analysis by wedge polishing and subsequent ion milling with a Gatan precision ion milling system (PIPS). In addition to TEM imaging, the LEO sample foils were used for polarity determination via CBED using the TEM foil imaged in Fig. 3(a). In CBED patterns obtained for a range of TEM foil thicknesses and compared with patterns simulated using the DESKTOP MICROSCOPIST 2.0 software package. The software generates intensity patterns calculated using the dynamical Bloch wave method. Furthermore, selected area electron diffraction patterns from the TEM foil confirmed the a - GaN/r-sapphire in-plane orientation previously determined using x-ray diffraction measurements (see Ref. 11).

III. RESULTS AND DISCUSSION

A. GaN LEO stripe morphology and microstructure

GaN LEO stripes aligned along [1100]GaN possesses rectangular cross sections for the growth conditions employed, as shown in Fig. 2(a). Since the polar GaN c-axis is perpendicular to the direction of the stripe, the vertical sidewalls of the stripe are {0001} c-plane facets with opposing polarities while the top horizontal facet is the nonpolar (1120) growth surface. Figure 2(a) also reveals substantial asymmetry in the extent of lateral overgrowth to the left and right of the mask window (the regions of growth over the mask are referred to as "wings"). The right wing of the stripe is roughly an order of magnitude wider than the left wing, indicating a significant difference in the growth rates of the polar surfaces. This lateral growth rate asymmetry for [1100]GaN stripes is consistently observed for various LEO samples grown to date. LEO dramatically reduces the substantial threading dislocation (TD) density present in the heteroepitaxial α-GaN films via mask blocking for this particular crystallographic orientation as shown in Fig. 2(b), a cross-sectional TEM image of the window/mask region. TDs extend vertically through the mask window but do not propagate laterally into the wings; therefore, the wing regions are considered dislocation free. Note that this TD reduction behavior is unique to the [1100]GaN stripe orientation since TDs have been observed to bend into the overgrowth for other crystallographic α-GaN LEO stripe orientations.12

B. Polarity determination via CBED

To determine the polarity and strictly define the a = GaN/r-sapphire crystallographic relationship, CBED experiments were performed on the LEO wing which exhibited a larger growth rate using the TEM foil imaged in Fig. 3(a). Note that since there is a 180° rotation between the image...
and the diffraction patterns due to the microscope geometry, the TEM images shown in Fig. 3 have been rotated along the diffraction pattern so that the pattern alignment matches the associated image. A series of CBED patterns corresponding to a range of foil thicknesses was obtained on a single foil as a result of wedge polishing. Figures 3(b) and 3(c) show two of the experimental CBED patterns obtained for the nonpolar sample. The center disk of the patterns exhibits mirror symmetry whose contrast provides information on the thickness of the sample. The asymmetry in the contrast of the 0002 fringes is dependent on the thickness and polarity of the sample. The asymmetry whose contrast provides information on the thickness of the sample since inversion domains were not observed. For these simulations, the positions of the atoms were defined according to Fig. 1 and the absorption coefficient was set as 0.05. Additionally, 33 zero order electron beams were selected for many-beam dynamical calculation in the (1100) zone axis. Note that Zhao and co-workers used similar simulation conditions to determine the polarity of GaN grown on Si (111).

To confirm the accuracy of the polarity analysis described above, the analysis was repeated for a c-plane GaN film grown on a c-plane sapphire substrate via MOCVD. A TEM image of the polar GaN film cross section is shown in Fig. 3(f). The experimental CBED patterns that correspond to the thickness of the simulated patterns used for the nonpolar GaN CBED comparison are shown in Figs. 3(g) and 3(h). The CBED analysis identifies the surface of the MOCVD-grown c-GaN film as Ga-face, which is consistent with a large body of work. The CBED patterns from the polar sample confirm the validity of the a-GaN polarity determination.

C. Polarity effects: LEO

The nonpolar GaN LEO sample analyzed herein provides a unique opportunity by which to study and compare the growth of (0001) and (0001-1) surfaces on the same wafer under identical growth conditions. The TEM image shown in Fig. 2 clearly reveals the substantial effects of polarity on the lateral overgrowth. The CBED analysis determined that the positive c-axis points in the direction of faster lateral growth, which corresponds to the direction of enhanced lateral growth rate. To complement the CBED analysis and confirm the epitaxial relationship, the selected area diffraction pattern shown in Fig. 4 was obtained in the vicinity of the a-GaN/r-sapphire interface. The GaN +c-axis coincides with the r-sapphire substrate’s c-axis surface projection, and produces the following in-plane relationship: [0001]GaN||[1100]sapphire and [1100]GaN||[1120]sapphire. To confirm the accuracy of the polarity analysis described above, the analysis was repeated for a c-plane GaN film grown on a c-plane sapphire substrate via MOCVD. A TEM image of the polar GaN film cross section is shown in Fig. 3(f). The experimental CBED patterns that correspond to the thickness of the simulated patterns used for the nonpolar GaN CBED comparison are shown in Figs. 3(g) and 3(h). The CBED analysis identifies the surface of the MOCVD-grown c-GaN film as Ga-face, which is consistent with a large body of work. The CBED patterns from the polar sample confirm the validity of the a-GaN polarity determination.
thus the (0001) surface grows faster than the (0001$\bar{\text{1}}$) surface by approximately an order of magnitude for the growth conditions employed. The polarity dependence of the growth rate is not entirely unexpected since fundamental differences exist between the (0001) and (0001$\bar{\text{1}}$) surfaces of the GaN crystal. According to density functional theory calculations, the energetically stable polar nitride surfaces independent of the chemical environment are cation stabilized, thus each Ga atom on a (0001) surface has one dangling bond while each Ga atom on a (0001$\bar{\text{1}}$) surface has three dangling bonds. Regardless of the applicability of the above calculations to a MOCVD growth environment, different surface terminations are manifested in vastly different surface morphologies: characteristically rough (0001$\bar{\text{1}}$) surfaces are inferior to smooth (0001) surfaces in terms of GaN crystal growth.

An investigation of the sidewall surface morphology provides further evidence of the sidewall polarity and subsequent growth rate asymmetry. As has been shown for polar c-GaN growth on a variety of substrates, high temperature (e.g., T$_{\text{surface}}\approx 1050–1100^\circ\text{C}$) MOCVD growth conditions consistently produce smooth Ga-face (0001) GaN surfaces that grow via a step-flow mode.$^{13–16,19,22}$ Although the current morphological analysis cannot confirm the presence of atomic steps, the SEM image shown in Fig. 5(a) of the (0001) surface reveals a smooth morphology that is interrupted by a sidewall facet. These facets regularly appear due to crystallographic stripe misorientation and have been observed in misaligned polar GaN LEO stripes.$^{23}$ The sidewall facets are indexed as (1102) surfaces according to the interplanar angle measured between the sidewall facet and the (0001) sidewall in Fig. 5(b), a plan view SEM micrograph of a [1100]$_{\text{GaN}}$ LEO stripe. Note that the sidewall facet is closely related to [1101], a common nonbasal facet encountered in MOCVD growth of GaN.$^{24,25}$ Conversely, instead of sidewall facets, areas of smooth (0001$\bar{\text{1}}$) sidewall morphol-
ogy are interrupted by hexagonal hillocks, features which have been reported for N-face GaN MOCVD growth.\textsuperscript{26,27} The hillocks nucleate at specific locations along the mask/wing interface, as shown in Fig. 5(c), and appear to be the step source for the (0001) sidewall growth. We speculate that the terraces associated with the hexagonal hillocks and the small amount of lateral overgrowth in the [0001] direction account for the absence of periodic sidewall faceting on the (0001) surface.

In addition to the relative lateral growth rates and sidewall morphology, the crystal polarity dictates the planar defect reduction which is observed in the lateral overgrowth of \(\{\bar{1}100\}\)\textsubscript{\text{GaN}} LEO stripes. As previously mentioned, the microstructure of the heteroepitaxial \(a\)-GaN contains stacking faults on the (0001) plane. Plan-view TEM foils of an individual LEO stripe were prepared according to the schematic included with the images shown in Fig. 6. The planar fault density in the Ga-face LEO wing is dramatically reduced with increasing distance from the window region; after \(\sim 1\) \(\mu\)m of lateral overgrowth along [0001], no stacking faults are present in the wing. The observation that we can “grow out” stacking faults via lateral overgrowth along [0001] indicates that our stripe sidewalls have sufficient atomic step sources which allow fault-free growth. This is supported by the SEM observation of misorientation facets shown in Fig. 5(a), which implies the existence of nonsingular (0001) sidewall surfaces. In contrast to the Ga-face wing, planar defect reduction is not observed for the N-face wing shown in Fig. 6. We speculate that the absence of fault density reduction in the [0001] overgrowth is a consequence of both a short wing and the surface morphology of the polar facet.

**D. Polarity effects: Heteroepitaxial growth**

The \(a\)-plane GaN films used as template layers for LEO were grown heteroepitaxially on \(r\)-plane sapphire using a two-step growth technique similar to that used for \(c\)-plane GaN growth on \(c\)-plane sapphire. If the growth conditions for either the low temperature nucleation layer or the high temperature epitaxial growth are not optimized, large crystallographic pits decorate the film. The general form of the triangular pits is shown schematically in Fig. 7(a): inclined facets terminate at vertical \(c\)-plane facets which possess a common polarity across the sample. Although the vertical \(c\)-plane surfaces of the pits are clearly apparent in the TEM cross section (imaged perpendicular to the \(c\)-plane) shown in Fig. 7(b), the pit surfaces themselves are faceted and the intersection of the pit surfaces with each other and the \(a\)-plane surface are not sharply defined. An additional perspective of the pits is provided by the SEM plan-view micrograph shown in Fig. 7(c). Despite the range of pit shapes observed, the overall orientation and geometry of the pits are uniform. According to the CBED analysis and measurements of the TEM and SEM images, the vertical pit surface is aligned with (0001) and the two inclined facets are \{10\(\bar{1}\)1\}.

Note that the N-face polarity of the vertical \(c\)-plane pit surface can be identified using the polarity-dependent characteristics of the GaN LEO stripe in Fig. 7(d).

The crystallographic pits observed throughout the ongoing development of \(a\)-GaN growth assume the general form of \(V\) defects, which are characteristically observed under conditions of kinetically limited growth of \(c\)-plane GaN and its alloys.\textsuperscript{25} For a \(c\)-plane film, the \(V\) defect is an open, inverted pyramid which is bound by the pyramidal \{10\(\bar{1}\)1\} facets commonly encountered in GaN MOCVD growth. Due to the nonpolar orientation, the \(V\) defect appears in an \(a\)-GaN film on its side (\(<\) defect), as shown schematically in Fig. 7(e). Unlike the \(V\) defects observed in \(c\)-plane growth, the \(<\) defect is not created by an individual threading dislocation.

![Figure 6](image6.png)

**FIG. 6.** Plan-view TEM micrographs of a \(\{\bar{1}100\}\)\textsubscript{\text{GaN}} LEO stripe revealing the planar defect reduction achieved with lateral overgrowth. The stacking fault density is reduced to zero after \(\sim 1\) \(\mu\)m of lateral overgrowth along [0001] whereas no stacking fault reduction is observed for [0001] overgrowth. The location of the TEM foil with respect to the LEO stripe is shown schematically on the left. Since the plan-view foil was taken from the lower portion of the LEO stripe close to the SiO\(_2\) mask, voids that exist at the interface between the GaN wing and the SiO\(_2\) mask are visible in the micrographs.

![Figure 7](image7.png)

**FIG. 7.** Schematic representation (a) of the pits encountered in \(a\)-GaN films grown under nonoptimized conditions. (b) Cross-sectional TEM and (c) plan-view SEM images illustrating the deviations in pit geometry from the basic form shown schematically. Despite the minor variations in pit shape, the orientation of the pits is consistent across the sample: two inclined \{10\(\bar{1}\)1\} facets terminate at a vertical (0001) facet. (d) The polarity of the \(c\)-plane pit facet is clearly visible in the plan-view SEM image of a LEO sample that contains a pit in the underlying template layer. The \(a\)-GaN pits adopt the form of a characteristic \(V\) defect, shown schematically (e), which is commonly observed in \(c\)-plane growth.
located at the base of the defect. We speculate that these pits form upon island growth and coalescence at the early stages of high temperature growth. Since the pits do not close up throughout the course of the high temperature growth, the pit facets are extremely stable and slow growing. This conclusion is reasonable considering the observation of the slow growing (0001) surface present during LEO from [1100] stripes. Future developments in the heteroepitaxial growth will focus on preventing the formation of these slow growing facets through strict control of the nucleation layer and initial high temperature a-GaN growth conditions.

**IV. CONCLUSION**

CBED analysis conclusively determined the polarity of (1120) a-plane GaN on (1102) r-plane sapphire thereby explicitly defining the epitaxial film/substrate relationship: [0001]_GaN// [1101]_sapphire and [1100]_GaN// [1120]_sapphire. The CBED analysis made it possible to identify the effects of polarity on both heteroepitaxial growth and LEO. They are as follows.

(1) Polarity clearly affects LEO from mask windows aligned along [1100] (perpendicular to the c-axis) since these stripes possessed vertical (0001) and (0001) sidewall surfaces. The (0001) surface appeared smooth on SEM length scales and grew approximately an order of magnitude faster than the (0001) surface which showed characteristic hexagonal hillocks.

(2) The polarity of the LEO stripe also plays a role in planar defect reduction since the Ga-face wings exhibited a dramatic reduction in stacking fault density after ~1 μm of lateral overgrowth whereas no reduction was observed for the short N-face wings.

(3) The formation of crystallographic pits on heteroepitaxially grown film surfaces can be attributed to polarity effects. The triangular pits characteristically contain a facet which is either exactly (0001) or slightly misoriented from the c-axis. We speculate that exposed (0001) facets are the source of the stacking faults present in the heteroepitaxial films. Future studies of a-plane growth will focus on preventing these stable surfaces from forming during the early stages of high temperature epitaxial growth.

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