Curvature and bow of bulk GaN substrates

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(Received 1 April 2016; accepted 3 July 2016; published online 19 July 2016; corrected 15 August 2016)

We investigate the bow of free standing (0001) oriented hydride vapor phase epitaxy grown GaN substrates and demonstrate that their curvature is consistent with a compressive to tensile stress gradient (bottom to top) present in the substrates. The origin of the stress gradient and the curvature is attributed to the correlated inclination of edge threading dislocation (TD) lines away from the [0001] direction. A model is proposed and a relation is derived for bulk GaN substrate curvature dependence on the inclination angle and the density of TDs. The model is used to analyze the curvature for commercially available GaN substrates as determined by high resolution x-ray diffraction. The results show a close correlation between the experimentally determined parameters and those predicted from theoretical model. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).

I. INTRODUCTION

Gallium Nitride (GaN) is a binary III–V direct band-gap semiconductor material with a Wurtzite crystal structure. Its unique physical properties have made it advantageous for practical applications in optoelectronic and high power/frequency electronic devices. GaN-based device structures have traditionally been grown heteroepitaxially on sapphire or silicon carbide substrates. However, large lattice mismatch between GaN and these foreign substrates yields threading dislocation (TD) densities (TDDs) up to 10^8–10^9 cm^{-2}. Studies have shown that TDs in GaN are electrically active and detrimental to carrier transport in emission properties of semiconductor heterostructures. Therefore, there is a strong need to get bulk GaN material for homoepitaxial growth of device structures with low TDD.

II. BACKGROUND

A. Substrate curvature and bow

However, despite the high quality of HVPE materials, the freestanding substrates have significant curvature. The curvature present in freestanding GaN is caused by internal stresses from within the substrate. The stresses undergo a change in sign and change from the bottom to the top of the substrate, where they change from compressive to tensile stress.

In this report, the TDD and the radius of curvature (Rc) for commercially available bulk GaN substrates were recorded and compared alongside a derived theoretical model for dislocation inclination in freestanding c-plane GaN. From this, a relationship between the stress gradient induced curvature in c-plane GaN to the TDD and threading dislocation inclination (TDI) is demonstrated.
FIG. 1. Example of bow and curvature in a substrate. (a) Flat substrate with no curvature of the crystal planes. (b) Bowed substrate surface with no crystal plane curvature. (c) Bowed substrate with crystal plane curvature. (d) Flat surface with crystal plane curvature.

lattice elastic strain. For example, Figs. 1(c) and 1(d) have the same curvature, but only Fig. 1(c) is bowed. Therefore, to accurately measure the RC of a substrate, regardless of which bowing state is present, XRD is the preferred method.

B. Derivation of curvature in bulk multi-layered structure

To accurately analyze the curvature of a bulk crystal substrate, a multi-layered structure model is used as shown in Fig. 2(a). The total thickness \( H \) is divided into \( N \) layers with arbitrary thickness \( h_i \). The reference length, \( L \), included aref) with no elastic strain, stresses, or bending. The layers are then separated as shown in Fig. 2(b), such that they are unconstrained and each layer undergoes a lattice transformation, expanding, or contracting with respect to its respective reference length. The number of lattice parameters within \( L \) remains the same, introducing transformation strain \( \varepsilon_i^T \), also known as stress free (eigen) strain,\(^{17} \) expressed as

\[
\varepsilon_i^T = \frac{a_i - a_{\text{ref}}}{a_{\text{ref}}}, \quad i = 1 \ldots N,
\]

where \( a_i \) is the lattice parameter after undergoing the lattice transformation.

Coherent layers deform in a compatible manner, meaning a 1:1 correspondence between the lattices at the interface of contacting layers such that during the lattice transformation, the lattice maintains continuity across the interface. The lateral strains in the layers will have a \( z \)-dependence, with the coordinate origin at the bottom of the first layer. The compatible deformation of the multilayer structure with the respective coordinates is shown in Fig. 2(c), and the required conditions of the compatibility of total strain \( \varepsilon^T \) are shown as\(^{18} \)

\[
\frac{\partial^2 \varepsilon^T(z)}{\partial z^2} = 0.
\]

The general expression for total strain can then be expressed as

\[
\varepsilon^T(z) = A + Bz,
\]

where \( A \) and \( B \) are unknown coefficients used to derive an expression for curvature. Given that each layer is subjected to a different magnitude of self-strain, the compatible total deformation mentioned above, \( \varepsilon^T \), is maintained by the layer’s elastic strain layers, \( \varepsilon_i \), written as

\[
\varepsilon_i = \varepsilon^T - \varepsilon_i^T.
\]

The elastic strain in the \( i \)th layer will have a \( z \)-dependence as the compatible total deformation and self-strain have a \( z \)-dependence; therefore, \( \varepsilon_i \) can be rewritten as

\[
\varepsilon_i(z) = A - \varepsilon_i^T(z) + Bz.
\]

The elastic bending shown in Fig. 2(d) accompanies non-uniform elastic strain. The inherent strain in the individual layers of the structure causes the multilayered structure’s curvature, thus, to develop an expression for curvature, an analysis of the stress state is required. The \( z \)-dependent elastic strain is related to mechanical stress by assuming Hooke’s Law and plane stress state in layers as shown

\[
\sigma_i(z) = M_i(z)\varepsilon_i(z), \quad i = 1 \ldots N,
\]

where \( \sigma_i(z) \) and \( M_i(z) \) are the \( z \)-dependent mechanical stress and the biaxial elastic modulus of the \( i \)th layer, respectively. The two unknown coefficients, \( A \) and \( B \), can then be determined by using mechanical equilibrium conditions for force and bending moment to yield

\[
A = \frac{J_1 I_k - J_2 I_0}{I_0 J_2 - I_1^2}, \quad (7a)
\]

\[
B = \frac{J_0 I_k - J_1 I_0}{I_0 J_2 - I_1^2}, \quad (7b)
\]

where \( I_k \) and \( J_k \) are defined as

\[
I_k = \int_0^H z^k M_i(z)dz, \quad k = 0, 1, 2, \quad (8a)
\]

\[
J_l = \int_0^H z^l M_i(z)\varepsilon_i(z)dz, \quad l = 0, 1. \quad (8b)
\]

This is done in order to simplify the expressions for the coefficients. Assuming that the self-strain and biaxial moduli are known for the system, the curvature \( \kappa \) can be found using the relation
\[ \kappa = \frac{\varepsilon^T(H) - \varepsilon^T(0)}{H}, \]  
(9)

which reduces to
\[ \kappa = -B. \]  
(10)

The approach described above is consistent with the results presented in the literature.\cite{19-21}

### C. Inclined threading dislocations and stress gradients in III-nitride structures

This report attributes the compressive to tensile stress gradient in freestanding GaN to curvature caused by threading dislocations\cite{22} and their inclination within the material. Cantu et al. reported relaxation of compressively strained Si-doped (0001) Al\(_{0.49}\)Ga\(_{0.51}\)N deposited on Al\(_{0.62}\)Ga\(_{0.38}\)N, where TEM discovered threading dislocations with initial [0001] lines directions in Al\(_{0.62}\)Ga\(_{0.38}\)N inclining toward the (1100) directions within Si doped Al\(_{0.49}\)Ga\(_{0.51}\)N.\cite{23}

These threading dislocations have an effective misfit dislocation (MD) component that generates stress, which may be in the same sense or different senses to other stresses in the layer. Furthermore, a model describing the conditions for dislocation inclination and relaxation due to “effective climb” of edge dislocations by Romanov and Speck\cite{24} explains the mechanism behind the relaxation of compresses stresses in Al\(_x\)Ga\(_{1-x}\)N. Further growth with “frozen-in” inclined dislocations is shown to generate a tensile strain gradient as demonstrated by Cantu et al.\cite{25} which is responsible for cracking at a critical thickness.\cite{26} Accord et al. and Manning et al. further demonstrated in situ growth wafer curvature measurements revealing compression to tension stress transition in compressed Al\(_x\)Ga\(_{1-x}\)N.\cite{27,28}

The mechanism by which stresses relax/generate and form gradients are related to the inclination of the TD with respect to their original direction. The most common method of observed relaxation in lattice-mismatched semiconductor heterostructures is misfit dislocation (MD) formation at the interfaces, perpetuated by dislocation glide. This gliding process can proceed by bending the pre-existing threading dislocations or by generation of new dislocations. There is, however, a glide-free mechanism of elastic stress relaxation related to the inclination of the TD with respect to their original direction and their effective climb as shown by Romanov et al. (2003), where the total biaxial far-field plastic relaxation at the top layer surface resulting from the triangular MD grid \(e_{pl}^3\) is given by
\[ e_{pl}^{top} = \frac{3}{2} e_{pl}^1 = \frac{3}{2} b \rho_{TD} L, \]  
(11)

where \(b\) is the Burgers vector, \(\rho_{TD}\) is the threading dislocation density, and \(L\) is the projected length directly related to the layer thickness \(h\) and the inclination angle \(x\), such that \(L = h \tan(x)\). The inclination angle, \(x\), is formed when the straight dislocation originating from the substrate bends at the substrate/layer interface and becomes an angular dislocation as shown in Fig. 3. The lines of these dislocations are inclined with respect to the [0001] growth direction, with \(x\) as large as 20°. The TD inclines toward the (1100) direction. From this, the average plastic relaxation for a layer is then

\[ R_C = \frac{x_2 - x_1}{\omega_2 - \omega_1}, \]  
(13)

where \(x_1\) and \(x_2\) are respective positions along the x-direction and \(\omega_1\) and \(\omega_2\) are the omega peak positions at those positions.

The threading dislocation density was determined using cathodoluminescence (CL) imaging. CL was performed using an FEI Scanning electron microscope (SEM) with a
The assumption is then made that the arc length, \( \gamma \), from the chord defined by \( \omega_1 \) and \( \omega_2 \) in the curved surface is the circumference of a circle with radius, \( R \), a geometric relationship can be established between the two scan points, 1 and 2, which are \( \Delta x \) apart on the lab horizontal frame. The angle of the incident beam with the lab horizontal is \( \omega \), the angle between the lab and crystal horizontals is \( \Phi \), and the angle between the crystal vertical and lab horizontal is \( \delta \).

Since the same peak in reciprocal space is being observed, it can be assumed that the Bragg’s condition is identically satisfied at both \( x \) positions, that is, \( \omega_1 = \omega_2 \) in Fig. 5 for all positions on the surface when observing a single peak in reciprocal space. The relation between Bragg peaks can be expressed as the angle, \( \beta \), between the scan positions relative to the center of the \( R_C \):

\[
\beta = \omega_1 - \omega_2. \tag{14}
\]

The assumption is then made that the arc length, \( \gamma = R\beta \), is equal to the horizontal shift of the sample stage. This assumption holds true as long as the deviation of the arc from the chord defined by \( \Delta x \), \( \sigma_r \), is negligible. Mathematically, this condition would be satisfied if \( \Delta x \gg \sigma_r \) or \( \Delta x \ll R \). Both are reasonable assumptions considering that radii of curvature are generally on the order of 10 m–1000 m, while translations are on the order of 5 mm. This makes the following:

\[
\gamma = R\beta = R(\omega_1 - \omega_2) = \Delta x. \tag{15}
\]

Our initial analysis of this system for a convex system was done with the lab frame using the same Cartesian unit vectors as the \( R_C \) frame. However, upon shifting to a concave derivation, the \( R_C \) frame had to be rotated about the \( y \)-axis to maintain a positive \( R \) vector. This shift changes the sign on the \( \Delta x \) term as the \( x \)-axis unit vector rotates about the \( y \)-axis. This can now be simplified to:

\[
R = \frac{-\Delta x}{\omega_1 - \omega_2} = \frac{x_2 - x_1}{\omega_2 - \omega_1}, \tag{16}
\]
as previously described by Fewster and thus derived regardless of the radial frame of reference used.

**IV. RESULTS**

**A. XRD curvature measurements**

The radii of curvature for the commercial bulk GaN crystals are displayed in Table I. Substrate A had the largest \( R_C \) with the largest standard error, 8.08 \( \pm \) 0.97 m. Substrate B and substrate C had similar radii of curvature, 5.42 \( \pm \) 0.23 m and 6.44 \( \pm \) 0.16 m, respectively. The smallest \( R_C \) was recorded from substrate D with 2.75 \( \pm \) 0.17 m.

As mentioned previously, \( R_C \) was measured through a series of omega scans displaced along the \( x \) direction, parallel to the source and detector of the diffractometer. The radii of curvature of the bulk GaN crystals further indicate that the substrates were bowed concave up as shown in Fig. 6. With the substrate bowed concave up, the incident beam would reach the Bragg condition at reduced angles as it traversed the curved surface. This would, in turn, result in a Bragg peak at a smaller \( \omega \) for one-dimensional translations where \( \Delta x > 0 \), hence a negative slope in Equation (13) is realized. For simplicity, the \( R_C \) was represented as an absolute value.

**B. Cathodoluminescence measurements**

Panchromatic CL images for the commercial bulk GaN crystals along with the corresponding evaluated TD density...
are displayed in Fig. 7 and Table II, respectively. The CL images show predominantly bright areas corresponding to band gap luminescence from defect free GaN as well as dark spots corresponding to non-radiative regions around defects that have been shown to be associated with threading dislocations, as in Rosner et al. The CL images show a fairly uniform concentration of TDs along the region of the imaged surface, independent of length scale, while the corresponding SEM images of the bulk crystals all exhibited planar smooth surfaces (not shown).

Substrate A, shown in Fig. 7(a), had the lowest number of TD per scanned area, with a TDD of 4.16 × 10^6 cm^-2. Substrate B shown in Fig. 7(b) had a TDD of 1.35 × 10^6 cm^-2, with a low standard error compared with the rest of the sample set. The TDs in substrate B are distributed in small groups or clusters. Substrate C shown in Fig. 7(c) had a TDD of 1.26 × 10^6 cm^-2; however, this value is associated with a larger standard error, due to large variations in the distribution of dislocations at larger scales (not shown). Substrate D shown in Fig. 7(d) had the highest density of threading dislocations, at 2.92 × 10^6 cm^-2.

V. THEORETICAL MODEL AND DISCUSSION

To model the dislocation inclination in freestanding GaN, a relationship between curvature, dislocation density, and threading dislocation inclination angle must be established. The curvature of a bulk multi-layered structure was derived to be equal to the unknown coefficient B in Eq. (10), which takes into account the thickness dependence of the self strain $\varepsilon_i^s(z)$, defined as the lattice transformation that each layer undergoes as it expands or contracts with respect to its reference length. The self strain subjected to each layer of the structure is equivalent to the average plastic relaxation resulting from the glide free mechanism of stress relaxation related to the inclination of TD from Romanov et al., as shown below

$$\varepsilon_i^s(z) = \bar{\varepsilon}_{pl} = \frac{1}{4} b \rho_{TD} h \tan(\alpha).$$  

Equation (17) can then be used to solve for the unknown coefficient B, so that the curvature can be determined. This result derives an expression where the curvature can be proportional to the TDI and TDD. This statement assumes that the biaxial modulus no longer has $z$-dependence as $\varepsilon_i^s(z)$ is assumed to be the $z$-dependent plastic relaxation. After appropriate calculations are made, the curvature is

$$\kappa = \frac{1}{4} b \rho_{TD} \tan(\alpha).$$  

The thickness dependence is eliminated, and the curvature developed by the average plastic relaxation is directly related to the TDD and the average inclination angle of the TDs. Figure 8 shows a three dimensional schematic for inclined TD in wafers grown in [0001] direction.

Dwilinski et al. reported on the growth of exceptionally high quality bulk GaN substrates using the ammonothermal method. They reported TDD of $\sim 5 \times 10^3$ cm^-2. Regardless of the crystal size, the $R_C$ was reported to be greater than or equal to 1000 m. Figure 9(a) plots the derived model for varying inclination angles alongside the ammonothermal bulk GaN crystal grown by Dwilinski et al. The substrate supported the model, with an estimated inclination angle of $\sim 15^\circ$.

Figure 9(b) shows the dependence of $R_C$ with respect to TDD for the GaN substrates investigated. The experimental

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**TABLE I. Radius of curvature recorded experimentally via X-ray diffraction for the commercial bulk GaN crystal substrates.**

| Substrate designation | Radius of curvature (m) | Standard error (m) |
|-----------------------|-------------------------|-------------------|
| A                     | 8.08                    | 0.97              |
| B                     | 5.42                    | 0.23              |
| C                     | 6.44                    | 0.16              |
| D                     | 2.75                    | 0.17              |
RC and TDD values are plotted alongside the derived theoretical model over varying inclination angles. All of the commercial bulk GaN crystals investigated correlated well with the theoretical relations derived, supporting the proposed model relating the curvature from stress gradients to TDD and TDI.

According to the derived theoretical relation, substrate A had a TDI angle of 20°, with minimal deviation in both the RC and TDD. Substrate B had an inclination angle of 10°. The standard error was negligible for the RC and TDD; therefore, this is the most precise data point of the batch. Substrate C crystal was speculated to have an inclination angle, but the standard error is significant along the TDD; therefore, it could have an inclination ranging from 10° to 17°. Substrate D is speculated to have an inclination angle between 8° and 10°. From here, we can conclude that the theoretical model holds for bulk GaN crystals of large and small TDD and RC.

The ranges of inclination angles (8°–20°) chosen for this study are based of the observed and measured inclination angles from the previous studies. TEM studies done by Cantu et al. reported a TDD of ~3 × 10^10 cm^-2, where the TD was inclined by 10°–25° in the 1100 direction and had a misfit component, thus relieving misfit strain. Observations of Follstaedt et al. for compressively strained AlGaN indicated that the TD inclination occurred prior to the introduction of Si, where the average inclination angle was ~19°, and the TDD is ~6.9 × 10^5 cm^-2. Follstaedt et al. further examined AlGaN films with increasing Ga content, reporting TD inclination angles ranging from 6.7° to 17.8° and TDD of 6–7 × 10^9 cm^-2.

At the TDD observed in this report for HVPE bulk GaN (10^8–10^9 cm^-2), it is difficult to measure TDI via cross-sectional TEM, as the TDD is too low. Another method to measure the TDI is via x-ray topography; however, the TDD observed is too high for this technique. TD can be observed at this density when using Electron Channeling Contrast Imaging as performed by Picard et al. and Carnevale et al.

The average radius of curvature for the commercial HVPE grown GaN substrates is approximately ~5 m, leading to a large disparity in vicinality from edge to edge over the substrate (e.g., ~0.6° for RC = 5 m for a 50 mm substrate). For comparison, the vicinality of GaN on sapphire is optimized for ~0.2° miscut in the m-direction of GaN which corresponds to 0.2° towards a-direction sapphire.

Therefore, even for GaN substrates suitable for LD and LEDs (TDD ~10^4 cm^-2–10^6 cm^-2), there still exists a large non-negligible curvature. From this, one can see the importance of lowering the TDD in the future so as to achieve the flattest, least bowed substrates possible and avoid the detrimental effects to carrier transport and emission properties of.

TABLE II. Threading dislocation density recorded experimentally via cathodoluminescence imaging for the commercial bulk GaN crystal substrates.

| Substrate designation | Threading dislocation density (cm^-2) | Standard error (cm^-2) |
|-----------------------|--------------------------------------|------------------------|
| A                     | 4.16 × 10^5                         | 1.14 × 10^5            |
| B                     | 1.35 × 10^6                         | 3.67 × 10^4            |
| C                     | 1.26 × 10^6                         | 6.05 × 10^5            |
| D                     | 2.92 × 10^6                         | 2.09 × 10^5            |

FIG. 8. 3D schematic of inclined TDs in wafers grown in [0001] direction.

FIG. 9. (a) The Ammono crystal is in agreement with the theoretical model with an estimated inclination angle of ~15°. (b) The RC with respect to the TDD for a range of TDI for the commercial Bulk GaN substrates plotted alongside the derived theoretical model.
TD have in semiconductor heterostructures as demonstrated by Rosner et al.

VI. CONCLUSIONS

The radius of curvature and threading dislocation density for commercially available freestanding GaN substrates were measured. Substrates with higher threading dislocation densities have smaller radii of curvature. Furthermore, a model was derived to investigate the dislocation inclination in GaN substrates and its dependence to TDD and $R_c$. The model was developed by assuming that the curvature is caused by internal stresses within the substrate that undergoes a change from compressive to tensile stress from the bottom to the top of the substrate. The experimentally determined $R_c$ and TDD for substrates with uniform distribution of TDD confirm and support the derived theoretical model.

ACKNOWLEDGMENTS

This work was supported by the King Abdullah Center for Science and Technology and King Abdullah University of Science and Technology (KAUST/KAUST) as well as the Materials Research Laboratory and California Nanosystems Institute at UC Santa Barbara for providing access and training to their laboratories. A.E.R. got the support for the conducting theoretical part of this study from Russian Science Foundation Project No. 14-29-00086.