Lattice distortions in GaN thin films on (0001) sapphire

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Abstract. The GaN/sapphire microstructure and lattice distortions in GaN were investigated using convergent beam electron diffraction (CBED) and electron backscatter diffraction (EBSD). CBED studies showed small scale (<1°) triclinic strain in the electron microscope specimens with a tetragonal expansion (~0.001 nm) of the lattice along the GaN growth direction, which has been observed in continuous films as well as isolated or partly coalesced islands. The strains extend up to ~ 2 µm from the GaN/sapphire interface. EBSD studies show a clear mosaic structure. The largest component of the mosaic structure corresponds to a twist around the [0001] axis, perpendicular to the interface.

1. Introduction
Over the last decade, the group III-Nitrides AlInGaN have become an important materials system with optical applications in the visible and ultraviolet range of the spectrum. When heteroepitaxially grown on substrates such as the basal-plane (0001) of sapphire or silicon carbide, the resulting GaN film contains a high density of threading dislocations (TDs) ranging between 10⁷ to 10⁹ cm⁻². The large lattice mismatch stresses between GaN and the substrate are not accommodated by these TDs, but are believed to be relieved by an array of misfit dislocations at the GaN/substrate interface [1,2]. The origin of the TDs remains a topic of debate with basically two schools of thought: (a) the TDs are formed at the coalescence boundaries of slightly misoriented grains, i.e., the ‘mosaic tilt-and-twist’ model [1,2], and (b) the TDs originate from other defects in the highly imperfect region near the GaN/substrate interface, leading to spatially random TD arrangements with some (low-energy) linear array formation as a result of dislocation mobility at the GaN growth temperature (typically >1000°C) [3-5]. In view of these contrasting ideas, further work is needed to improve our understanding of the interplay of threading dislocations, strain distribution, low angle grain boundaries and other lattice distortions at the GaN/substrate interface. In this study, the GaN/sapphire microstructure and strain distribution were investigated in detail using convergent beam electron diffraction (CBED), and electron backscatter diffraction (EBSD) techniques. CBED is a TEM based technique offering the quantitative study of crystallographic parameters with high spatial resolution (~ few nm), whereas EBSD is a SEM based technique indicating the crystallographic tilts on a larger scale (~ several µm). In this paper, we discuss the findings of our studies.

2. Experimental
The GaN growth was carried out in a 6x2” close-coupled showerhead metal-organic vapour-phase epitaxy (MOVPE) reactor and the details are described elsewhere [6]. Four different samples were
grown with a GaN nominal deposition thickness of ~0.75 µm (sample A), 1.2 µm (sample B), 1.65 µm (sample C), and 4.5 µm (sample D), on c-plane sapphire substrates with a nominal miscut of 0.25º to (11-20). Samples were grown with the GaN surface in different stages of coalescence, viz., islands (sample A), partly coalesced islands (samples B, C), and a fully coalesced continuous film (sample D). EBSD studies were performed using a FEI Helios Nanolab FIB/SEM equipped with an Oxford Instruments EBSD system. Orientation maps were acquired over regions approx. 30 µm square with a sample spacing of 75 nm and an accelerating voltage of 20 kV, a current of 1.4 nA and a sample tilt of 70°. Relatively slow scan speeds were used to obtain high data quality and indexing of close to 100% of the collected patterns. The data was processed using a self-written Matlab routine. Cross-sectional TEM specimens were prepared by argon ion beam thinning, and energy-filtered CBED experiments were performed using a Tecnai F20 TEM with a GIF 2000 at room temperature. The zone axis chosen was [12,1,-13,0], as this orientation was found to be sensitive to the changes in the lattice parameters (c and α, β, and γ) of GaN [7]. HOLZ simulations were performed using the JEMS [8] program and the conditions are described elsewhere [7]. In the calculations of lattice distortions, thin foil relaxation effects, due to having a thin TEM specimen, were not considered.

3. Results
3.1 Microstructure
TEM studies showed that isolated GaN islands exhibit {1-101} facets (Fig.1a). Mixed dislocations were bent towards the island coalescence boundary due to image forces at the free surface (TEM image, Fig.1b). Also, the AFM studies (Fig.1c) showed that the GaN island distribution appeared spatially random. The threading dislocation density at the surface of sample D was determined using TEM and AFM to be ~ (3.7±0.3) × 10^8 cm^-2 [9].

![Figure 1](image1.png)

**Figure 1.** Dark field TEM images showing dislocations with edge component in (a) sample A, and (b) sample B. (c) An AFM image showing the surface morphology of sample B.

3.2 CBED
A simulated HOLZ pattern in the bright-field disk of a [12,1,-13,0] CBED pattern is shown in Fig.2(a), which exhibits mirror symmetry about the a-axis. This symmetry is broken by strains which change α and/or β; for example, a simulated pattern with a change in β is shown in Fig.2(b). In this case, β is the angle between [2-1-10] and [0001] of the GaN crystal. The experimental HOLZ patterns obtained from GaN islands in sample B showed breaking of this mirror symmetry, (representative image, Fig.2c), indicating strains of this type in the GaN electron microscope specimen. Detailed analysis of the positions of HOLZ lines in these patterns using the methodology described in [7] gives the distortions in α, β, γ, and c, for example, the respective estimated values are ~ (90.0±0.03)°, (119.9±0.01)°, (0.5191±0.0002) nm for the HOLZ pattern in Fig.2(c). It is surprising to observe the presence of such large scale β-strains, however they were also visible in selected area diffraction patterns (Fig.2d), which showed that the angle between g_{002} and g_{2110} deviated by about 1° from 90°. Therefore, our results show that in our electron microscope specimen GaN is subject to a triclinic strain in the initial stages of growth. There is also an expansion along the [0001] growth direction. Similar phenomena were observed in samples A and C.
A careful study [7] of the HOLZ patterns recorded at different locations across the fully coalesced GaN film (sample D), as a function of distance from the GaN-sapphire interface towards the surface, indicated that: (a) lattice distortions are predominantly confined to the lower ~2 µm of the film, corresponding to growth where the GaN islands are not fully coalesced, and (b) the GaN lattice relaxes steadily further away from the substrate.

Figure 2. Simulated [12,1,-13,0] HOLZ patterns for (a) GaN and (b) triclinic distorted GaN (β=80°). (c) Experimental HOLZ pattern, (d) [01-10] SAD pattern of GaN near the GaN-sapphire interface.

Figure 3. EBSD data from sample C showing θ, twist $r_{12}$ about [0001], tilt $r_{13}$ about [10-10] and tilt $r_{23}$ about [-12-10]. Histograms of the different components are shown on the left, a section of the data taken along line A-A’ and an optical (Nomarski) image of the surface are shown on the right.

3.3 EBSD

The raw data, describing crystal orientation using Euler angles, were converted into matrix form and transformed by applying the inverse of the average rotation of the dataset. The resulting data show small rotations away from the average and can be displayed in several ways (Fig.3). The magnitude of the rotation θ can be obtained from the trace of the rotation matrix $R$, since $\text{tr}[R] = 1 + 2\cos\theta$. Individual components, corresponding to a twist about [0001] ($r_{12}$); rotation (tilt) about [10-10] ($r_{13}$) or rotation (tilt) about [-12-10] ($r_{23}$) can also be obtained. Examination of Fig.3 and similar data from the other samples shows that (a) a clear mosaic structure can be seen, with regions – presumably corresponding to individual islands prior to coalescence – having more or less constant orientation distinct from surrounding material and (b) the component which shows the largest variation is the twist $r_{12}$. Note that the linear features with large tilts (~0.5°) present in the θ and $r_{23}$ images are artefacts caused by surface relief, as can be seen in the Nomarski optical micrograph of Fig.3.
4. Discussion
The two techniques employed here sample complementary parts of the deformation of the material. CBED measures strain and is insensitive to a rigid-body rotation of the crystal, whereas EBSD measures rigid-body rotations and is not particularly sensitive to the strain in the material. These can be described as the symmetric (strain) and antisymmetric (rotation) parts of the full deformation tensor $e_{ij}$. The two are spatially related in that a strain (either elastic or plastic) must be present to accommodate rigid-body rotations in a continuous film. The initial GaN nucleation layer is in the form of islands each of which has a distinct orientation, and the EBSD data here show that the mosaic structure persists through the film. We found that the variation in tilt decreases with increasing layer thickness, similar to the EBSD study of Trager-Cowan [10], although in their study the $r_{12}$ component was not extracted from the data. It is known that TDs are mainly mixed/edge-type rather than screw-type [11], which is consistent with the accommodation of a larger twist component $r_{12}$. The CBED studies show the strain in the material produces a triclinic distortion in the electron microscope specimens of the initial stages of GaN growth. This phenomenon could be due to a variety of parameters, such as substrate miscut, GaN-sapphire lattice misfit, thermal stresses and strain relaxation mechanisms in thin electron microscope specimens. The interplay of these parameters is complex, and modelling of the initial stages growth of GaN on sapphire is necessary to understand the origin and evolution of the lattice distortions. Interestingly, preliminary DFT calculations [12] indicate that a triclinic strain, of the scale observed, only results in a small lowering ($\sim 10-50$ meV) of the energy band gap $E_g$ of GaN. Cathodoluminescence (CL) studies are planned to verify these findings.

5. Summary
We have investigated the lattice distortions in $c$-plane GaN grown on sapphire using CBED and EBSD. The studies show a mosaic structure where twist is the major component and there is a triclinic distortion in our electron microscope specimens of the GaN in the initial stages of growth. The origin of these distortions is due to a complex interplay of several parameters, which requires modelling for a better understanding.

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