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Lattice distortions in GaN on sapphire using the CBED–HOLZ technique

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ABSTRACT

The convergent beam electron diffraction (CBED) methodology was developed to investigate the lattice distortions in wurtzite gallium nitride (GaN) from a single zone-axis pattern. The methodology enabled quantitative measurements of lattice distortions (α, β, γ and c) in transmission electron microscope (TEM) specimens of a GaN film grown on (0, 0, 0, 1) sapphire by metal-organic vapour-phase epitaxy. The CBED patterns were obtained at different distances from the GaN/sapphire interface. The results show that GaN is *triclinic* above the interface with an increased lattice parameter c. At 0.85 μm from the interface, $\alpha = 90^\circ$, $\beta = 89{\cdot}05^\circ$ and $\gamma = 119{\cdot}66^\circ$. The GaN lattice relaxes steadily back to hexagonal further away from the sapphire substrate. The GaN distortions are mainly confined to the initial stages of growth involving the growth and the coalescence of 3D GaN islands.

1. Introduction

Gallium nitride (GaN) is an important optoelectronic material in short wavelength devices [1]. It is widely grown on c-plane sapphire by epitaxial thin film deposition techniques, and it crystallises with the wurtzite structure (space group: $P6_3mc$, $a = 0.31884 \pm 0.00002$ nm and $c = 0.51850 \pm 0.00002$ nm at 25 °C [2]) under standard growth conditions. Due to large lattice mismatch of about 16% with c-plane sapphire and substantial difference in thermal expansion coefficients, the GaN crystal experiences lattice distortions. The spatial variation of these distortions across the GaN film is an important issue for the growth of good quality epitaxial layers and their physical properties. Conventionally, high-resolution X-ray diffraction (HRXRD) has been used for studies of lattice distortions; however convergent beam electron diffraction (CBED) provides significantly higher spatial resolution. The positions of higher-order Laue zone (HOLZ) lines, which appear in the bright-field disc of a CBED pattern, are highly sensitive to changes in the crystallographic parameters (a, b, c, α, β, and γ) and therefore suitable for examining the lattice distortions on a nanometre scale [3–8].

In this paper, we describe a new CBED–HOLZ methodology that can quantitatively determine the changes in α, β and γ as well as the changes in lattice parameter along the c-direction of GaN from a single zone-axis pattern. The methodology proposed has been experimentally validated and the strains in GaN adjacent to a sapphire substrate have been measured in electron microscope specimens. We have not taken into account the thin film relaxation effects using finite element analysis to relate our measured strains to those in bulk specimens, but we focus on the methodology of measuring strains in an electron microscope specimen from a single zone-axis pattern.

2. CBED–HOLZ methodology

The [0, 0, 0, −1] projection of a GaN crystal is shown in Fig. 1. We have used JEMS software [9] for the HOLZ simulations. For the reader's convenience, both three-index and four-index notations are used wherever possible.

Extensive dynamical HOLZ simulations for different zone axes indicated that the [25, 14, 0] (or [12, 1, −13, 0]) zone axis of GaN is particularly suitable for identifying lattice distortions (for conditions, refer to Table 1). This orientation can be obtained by tilting the crystal about the c-axis by either method: (a) 4° from the [2, 1, 0] (or [1, 0, −1, 0]) zone axis or method (b) 0.33° from the [9, 5, 0] (or [13, 1, −14, 0]) zone axis (Fig. 2a). As expected, both methods gave the same HOLZ lines positions obtained by CBED calculations using a Bloch wave approach. However, in the case of method (b), the positions of HOLZ lines calculated using kinematic conditions with a 'HOLZ shift' were found to be in close agreement with those from dynamical calculations using a Bloch wave approach. In the case of kinematic HOLZ lines, the positions of the HOLZ lines are given by Bragg's Law. The 'HOLZ shift' is a feature

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given in the JEMS program for a first-order correction that partly takes into account the dynamical diffraction effects [9]. In the Bloch wave approach, the positions of the HOLZ lines are accurately calculated using dynamical diffraction theory. Thus a careful selection of the incident beam direction permits the generation of a HOLZ pattern in only a few minutes of computing time on a standard desktop computer facilitating the qualitative exploration of variations in crystallographic parameters on the positions of the HOLZ lines.

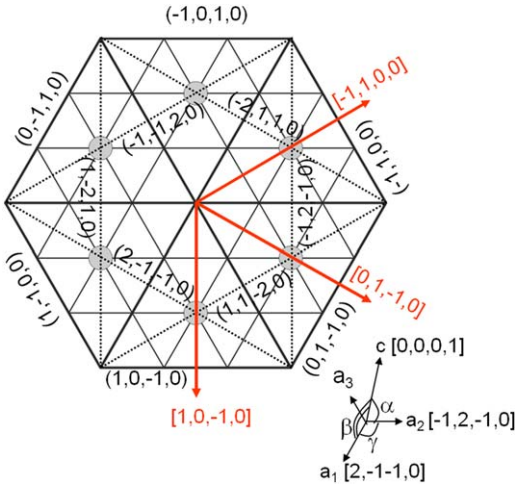

Fig. 1. [0, 0, 0, −1] projection of GaN crystal.

Table 1
Parameters used for room temperature HOLZ simulations using the Bloch wave approach in JEMS.

GaN crystal:	
$Ga(\frac{2}{3},\frac{1}{3},0), Ga(\frac{1}{3},\frac{2}{3},\frac{1}{2}), N(\frac{2}{3},\frac{1}{3},0.3772), N(\frac{1}{3},\frac{2}{3},0.8772)$	
Space group:	$P6_3mc$
a:	0.3173–0.3204 nm
c:	0.5173–0.5203 nm
α, β:	88.01–92.04°
γ:	119.68–120.32°
Zone axis:	[25,14,0] (refer Section 2)
Laue zone no.:	4
Acceptance angle:	50 mrad
Camera length:	3000 mm
Half-convergence angle:	3.85 mrad
Voltage:	200 kV
HOLZ threshold:	75 mV
Specimen thickness:	450 nm
Number of beams:	50
Atomic form factors (PRDW) and D–W factors used were the default values given in JEMS	

Initially, the HOLZ simulations were performed using kinematic theory with a 'HOLZ shift' for a qualitative examination of the positions of the HOLZ lines with varying lattice distortions (method (b)). Then, the simulations were performed using the Bloch wave method and quantitative measurements were performed on these HOLZ patterns. Fifty beams were used in the Bloch wave calculations (see Table 1) since no significant changes were observed when 100 or 200 beams were used. Also, the default values given in JEMS were used for the electron atomic form factors (AFFs), viz., the AFF values of Peng, Ren, Dudarev and Whelan (PRDW) [10] and the Debye–Waller (D–W) factors. Using different theoretical AFFs and D–W factors for GaN did not notably influence the positions of the HOLZ lines.

A representative simulated HOLZ pattern of an undistorted GaN crystal is shown in Fig. 2b with relevant HOLZ indices and intersections (A–E) marked in Fig. 2c. Fig. 2c is made from Fig. 2b by carefully drawing straight lines over the HOLZ lines. This pattern exhibits a vertical mirror perpendicular to the c-axis, passing through the HOLZ intersections A and B. An analysis of similar calculated HOLZ patterns for distorted GaN showed the following:

(a) The distance between A and B (defined as AB) remains nearly constant for small lattice distortions; hence AB can be used for the calibration of the HOLZ patterns.
(b) With α and β distortions, the mirror symmetry of the HOLZ pattern is lost (Fig. 3), and the line connecting C and E is non-parallel and parallel to AB, respectively. For identifying different intersections of the HOLZ lines, we have considered a relatively larger portion of the bright-field disk (Fig. 2b is a representative region), and straight lines were drawn over the HOLZ lines as shown in Fig. 2c. In Fig. 3, we have presented the cropped images showing only the necessary HOLZ intersections. For quantification, we define θ (with sign conventions indicated in Fig. 3) as the angle between AB and BC. $\theta-\alpha$ and $\theta-\beta$ have linear relationships (listed in Table 2, sl. no. 1). Also, θ exhibits ten times more sensitivity to changes in α than in β. For example, an error of 1° in the measurement of θ translates to an error of about $\pm 0.014°$ in α and $\pm 0.14°$ in β. A reversal in accuracy on a similar scale (i.e., β is more sensitive than α, relationships listed in Table 2, sl. no. 2) can be obtained by orienting the crystal along the

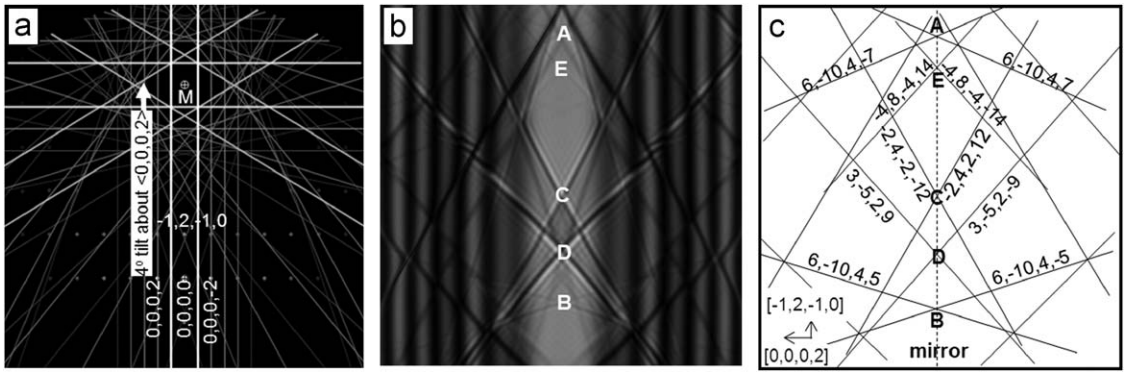

Fig. 2. Conditions employed for HOLZ simulations: (a) [25, 14, 0] (or [12, 1, −13, 0]) zone axis obtained by tilting the GaN crystal from the [2, 1, 0] (or [1, 0, −1, 0]) zone axis, about the c-axis, by 4°. This pattern has been simulated with a low camera length of about 1500 mm and Laue zone = 0 so that the Kikuchi bands and the centre of the Laue circle (M) can be clearly observed. (b) Portion of the HOLZ pattern obtained by dynamical calculations using the Bloch wave approach (GaN crystal, $a = 0.3189$ nm, $c = 0.5185$ nm; conditions given in Table 1); the positions of respective HOLZ lines are shown in (c) with important HOLZ intersections labelled as A–E.