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The utility of x-ray diffraction measurements in characterizing the tilt that occurs during lateral growth of GaN on a stripe-patterned SiO_2 mask is demonstrated. These measurements, in conjunction with additional structural characterization, have led to the minimization of 'wing' tilt via careful control of stripe cross-section. In addition, *in situ* grazing-incidence x-ray diffraction studies have revealed that 'wing' tilt evolves early during lateral growth, thus demonstrating the importance of initial growth conditions in determining tilt magnitude.

1. Introduction

The lateral epitaxial overgrowth (LEO) technique consists of coating a heteroepitaxial thin film with a patterned mask and regrowing such that material emerges from openings and grows laterally over the mask. When performed on heteroepitaxial GaN thin films, the resulting reduction in threading dislocation density has directly led to improved performance in (Al,In)GaN-based opto-electronic devices such as blue lasers,[1-4] LEDs[5] and p-n junctions[6] with lowered leakage current, as well as low-dark-current, sharp-cutoff solar-blind photodetectors.[7] Although LEO is indispensable in improving device performance, some difficulties remain in controlling the structural quality of the overgrown material. In particular, since the crystal planes in the 'wings' (overgrown GaN) are typically tilted with respect to those in the 'window' (in the mask opening) regions, coalescence of wings from neighboring stripes takes the form of a tilt boundary.[8,9] This is shown schematically in Fig. 1, which depicts pure-edge dislocations forming at window edges, accompanied by a sub-boundary at the coalescence front composed of dislocations of opposite sign. Although initial studies have shown that the tilt boundary may be suppressed with increasing film thickness,[8,10,11] a more sound approach is to minimize or eliminate wing tilt altogether.

Wing tilt may be semi-quantitatively measured in transmission electron microscopy (TEM) using selected-area diffraction or analysis of dislocation spacing.[8,9] However, it is more readily quantitatively measured using x-ray diffraction, for example by measuring an ω rocking curve about the GaN 0002 peak.[10-12] In this case, the scattering plane is oriented perpendicular to the stripes, such that wing tilt is observed as a peak splitting: the central peak from the window and underlying seed regions has 'wing' peaks on either side belonging to the overgrown regions.

In this paper we demonstrate that wing tilt on a SiO_2 mask is empirically correlated with the wing aspect ratio in cross-section, specifically the ratio, **r**, of overgrowth (w) to stripe height (h).[13] By controlling this aspect ratio, low tilt values (<0.1°) may be obtained, which lead to little or no extended defect generation during coalescence. Additionally, we report on recently performed *in situ*, real-time x-ray diffraction measurements *during* LEO of GaN over a SiO_2 mask that indicate that wing tilt emerges during overgrowth at high temperature, which furthers the understanding of the origins of wing tilt.

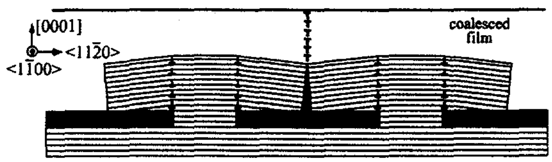

Fig. 1 Simplified schematic of sub-boundary formation due to wing tilt. Note opposite sign of dislocations at window edges vs. those at coalescence front.

2. Correlation of Wing Tilt with LEO Stripe Cross-Section

In the study described in this section, LEO of GaN was performed simultaneously on $\langle 1\bar{1}00\rangle$-oriented

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stripe patterns of various 'fill factors' (FF: ratio of stripe opening width in mask to stripe period), since it has been shown that the fill factor has a direct influence on stripe morphology.[14] The growth parameters of temperature and input V/III ratio were also varied, but the effects of these parameters on wing are described in detail elsewhere.[13]

The LEO GaN was grown by metalorganic chemical vapor deposition (MOCVD) on patterned, 2 μm-thick GaN on c-plane Al_2O_3 substrates. Samples selected for patterning were coated with ~200 nm SiO_2 deposited at 250 °C by plasma-enhanced chemical vapor deposition (PECVD). Subsequently, nominally 5 μm-wide stripes oriented in the $<1\bar{1}00>_{GaN}$ direction[15] were patterned using conventional photolithography, and etched using buffered HF. The stripe period was varied between 10 and 100 μm, yielding a FF range of 0.50 to 0.05. The approximate surface temperature during LEO was 1065 °C (measured by optical pyrometry), the reactor pressure was 76 Torr, and TMGa (58 μmol/min) and NH_3 (1.8 slpm) were used as precursors in a H_2 carrier.

The stripe cross-sections that resulted from these LEO conditions are shown in Fig. 2. The sidewall formation of these stripes was similar to that observed previously,[14] with inclined ($\{11\bar{2}n\}$, where n ≈ 1-2.5) facets appearing at low FFs. Vertical $\{11\bar{2}0\}$ facets dominated at FFs higher than 0.10. In this FF range, the equivalent planar thickness (the stripe cross-sectional area multiplied by the repeat period) remained constant, indicating that all the reactants impinging on the sample during growth were incorporated into the LEO stripes.

The tilts of wings away from window regions are clearly discernable as a peak splitting in a rocking curve, as shown in Fig. 3(a). This splitting becomes larger as the FF increases (*i.e.*, as stripes become more closely spaced). In addition, the wing peaks themselves become broader with higher FF, such that the approximate full width at half maximum (FWHM) of the wing peaks for FF = 0.05 is 0.10°, whereas the FWHM becomes ~0.52° for FF = 0.50 wings. The observation that wing tilt increases with increasing FF is useful, but more importantly, wing tilt increases as the wing width-to-height ratio (**r**) increases (Fig. 3(b)).[13] The monotonic increase of wing tilt with increasing **r** indicates the importance of stresses at the wing-mask interface, and may be likened to a cantilever beam whose deflection under a given load depends on its cross-section. By finding growth conditions which minimize **r**, we have reproducibly obtained wing tilt values less than ~0.1° on a SiO_2 mask.[11] In this case, few (if any) extended defects were formed at coalescence fronts, enabling successful laser fabrication on them.[3]

3. *In Situ*, Real-Time Measurement of Wing Tilt

Although the x-ray diffraction measurements described above proved useful in measuring and minimizing wing tilt, they were *ex situ* measurements performed at room temperature, and as such did not elucidate the point at which wing tilt evolved during the LEO process. The study described below utilized *in situ*, real-time x-ray diffraction **during** LEO to further the understanding of the origins of wing tilt.

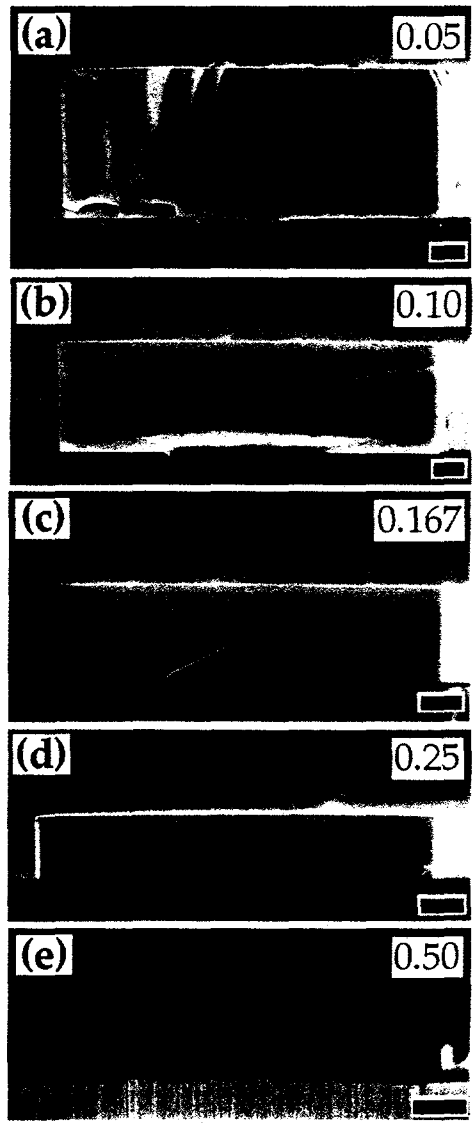

Fig. 2 SEM micrographs of stripe cross sections for fill factors of (a) 0.05, (b) 0.10, (c) 0.167, (d) 0.25, and (e) 0.50. Bar size is 1 μm; note that magnifications are not the same for all micrographs.

The *in situ* x-ray diffraction measurements were conducted at the BESSRC undulator beamline 12-ID-D in the Advanced Photon Source (APS). A vertical two-flow MOCVD chamber mounted on a 'z-axis' surface diffractometer enabled the use of grazing-incidence scattering, as described in detail elsewhere for the real-

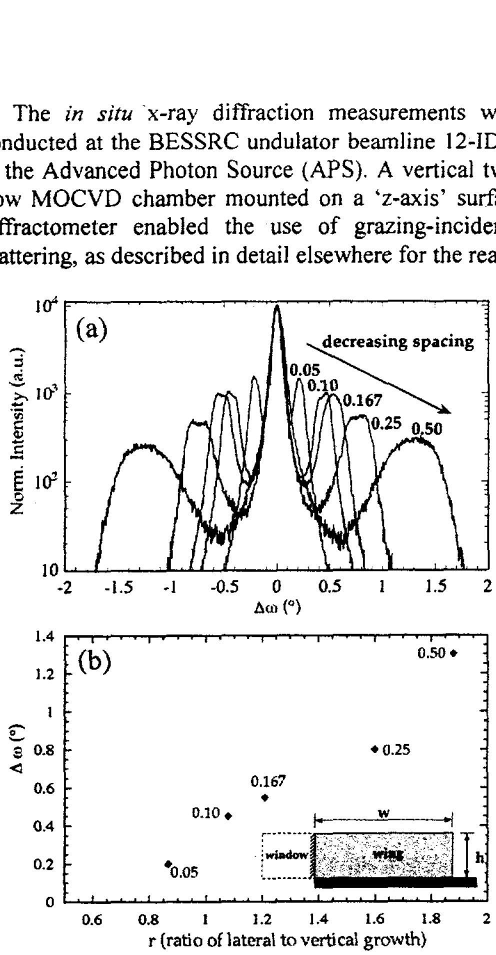

Fig. 3 (a) X-ray rocking curves labelled with fill factors from 0.05 to 0.50. (b) Wing tilt (Δω) vs. r, the ratio of wing width to height. Fill factors are noted next to each point.

time observation of GaN surface structure in the MOCVD environment.[16,17] Each sample consisted of a 2-3 μm GaN layer on sapphire grown by MOCVD at UCSB. The GaN was coated with ~200 nm of SiO_2 and patterned with <1 $\bar{1}$ 00>$_{GaN}$–oriented stripes of ~5 μm opening width and 20 μm period.[11] Samples were heated in the APS MOCVD chamber to an approximate surface temperature of 1060°C in flowing NH_3 and H_2/N_2 (1:3 ratio), and LEO was initiated as soon as this temperature was reached. The LEO conditions, described in detail elsewhere,[18] were optimized with respect to obtaining LEO stripes bound by (0001) and {11$\bar{2}$0} facets). This stripe morphology has been extensively studied,[9,11,14] and is one in which little or no dislocation redirection into the wings occurs, thus simplifying the analysis of wing tilt.

We monitored the profile of the GaN 10$\bar{1}$3 peak in reciprocal space, which has an in-plane azimuth parallel to the stripes. The splitting in this diffraction peak due to wing tilt was detected by scanning in a line oriented perpendicular to the stripes (in this case, [$\bar{1}$2$\bar{1}$0] for [10$\bar{1}$0]-oriented stripes). The tilt angle was calculated using the expression $tilt = \tan^{-1}(\Delta q/|q_{0003}|)$, where Δq was the measured distance in reciprocal space from the central 10$\bar{1}$3 peak and $|q_{0003}| = 3.634$ Å^{-1}. After growth, the sample was cooled in steps of approximately 200°C in flowing NH_3 and N_2/H_2 in order to make tilt measurements during cooldown.

The development of wing tilt during GaN LEO is evident in Fig. 4(a), which consists of a series of repeated line scans measured during growth at 1060°C. Each scan lasted approximately 100 s, with alignment checks done occasionally between scans early in the growth. The emergence of features distinct from the central (*i.e.*, underlying bulk GaN) 10$\bar{1}$3 peak was evident for times as early as 100 s, and by ~300 s, distinct wing peaks had evolved, with a peak position corresponding to a tilt of ~0.9°. Subsequently, the wing peaks narrowed and gradually increased in separation from the central peak, reaching a tilt of ~1.19° (with a full width at half-maximum of ~0.18°) after 3600 s growth. As shown in Fig. 4(b), upon cooldown to room temperature, the wing tilt did not change significantly, increasing from 1.19° at 1060°C to only 1.36° at room temperature. This indicates that although stresses were induced by thermal expansion mismatch with the sapphire substrate upon cooldown to room temperature, they had only a minor effect on the magnitude of wing tilt, contrary to what may be inferred from recent modelling studies.[19]

Since wing tilt develops early in lateral overgrowth, thermally activated processes must be considered as possible origins. It is likely that the SiO_2 mask undergoes physical and/or chemical changes early during LEO. Processes such as densification, etching, and formation of reaction products such as SiO_xN_y probably occur when the SiO_2 mask is subjected to the LEO growth environment (elevated temperature, as well as NH_3 and H_2, which are potential reducing agents). Additionally, the change in SiO_2 mask stress state in relation to the underlying film upon heating to 1060°C should be considered, as it has been postulated to be important in GaAs LEO on a SiO_2 mask.[20] One or more of the above factors could cause stresses sufficient to induce plastic deformation during LEO, since the Peierls stress in GaN is likely lowered at the growth temperature.[21] Further experimental studies are underway to establish the relative influence of each of these factors on wing tilt.

4. Conclusions

We have demonstrated the valuable role of x-ray diffraction in measuring and minimizing wing tilt that occurs during lateral ovegrowth of GaN on an SiO_2 mask. In addition to conventional rocking curve measurements at room temperature, we have utilized *in situ*, real-time grazing-incidence x-ray diffraction measurements in the MOCVD environment to directly observe the emergence and evolution of wing tilt during LEO. Although thermally induced stresses during cooldown appear to have little effect on wing tilt, factors such as changes in mask physiochemical properties and stress state early in growth must be considered as origins of wing tilt.

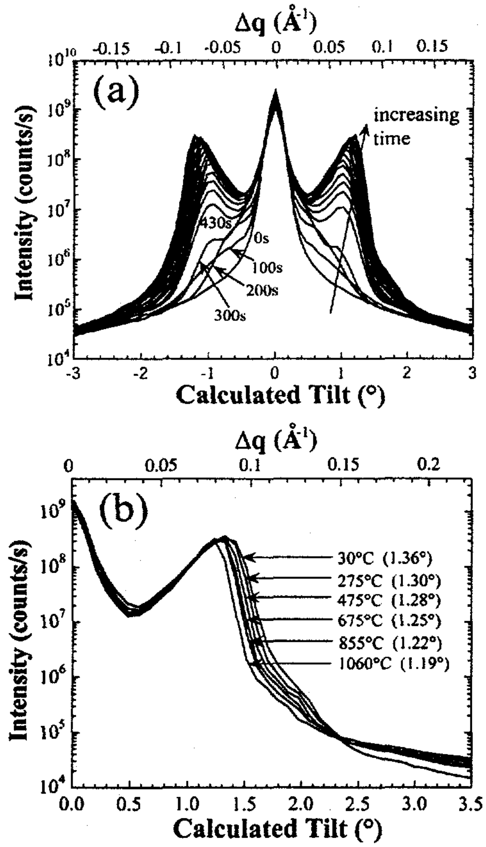

Fig. 4 (a) Intensity vs. calculated tilt angle (bottom x-axis) and Δq (top x-axis) during 3600 s of LEO growth at 1060°C. For clarity, early scans are labelled with their starting time in seconds. (b) Intensity vs. calculated tilt angle and Δq during cooldown from 1060°C to room temperature, shown over half of the range measured.

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