Investigation of optimum growth conditions of InAlN for application in distributed Bragg reflectors

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Abstract. In$_{x}$Al$_{1-x}$N is an alloy system which can be lattice matched to GaN, with many potential applications including distributed Bragg reflectors. This work investigated the effect of changing the ammonia flux during metal organic vapour phase epitaxy growth. The indium incorporation increases as the ammonia flux is increased from 44.6 mmol/min to 179 mmol/min, but remains constant for further increases up to 446 mmol/min. The surfaces across this range generally have low surface roughnesses suitable for use in distributed Bragg reflectors. However, some samples have significant surface pit densities, although these are not linked to similar rises in threading dislocations generated at the GaN / InAlN interface.

1. Introduction
The growth of InAlN lattice matched to GaN is an interesting recent development in the area of group-III nitrides. Lattice matching occurs at approximately 17.5 % In [1, 2]. One of the main applications for the material is for strainless distributed Bragg reflectors (DBRs) [1]. However InAlN has been perceived as difficult to grow using the two main techniques used in the group-III nitrides, metal organic vapour phase epitaxy (MOVPE) and molecular beam epitaxy (MBE), due to the very different growth conditions of the system end members. For example in MOVPE AlN is usually grown at a high temperature of approximately 1100 °C [3], but InN is usually grown at a low temperature of approximately 550 °C [4]. It has also been suggested that the system may have a spinode which would lead to spontaneous phase separation [5].

Unfortunately there are few systematic published studies of the effects of different growth conditions in MOVPE on the composition, morphology and defect structure of InAlN, although there has been some work exploring growth conditions in MBE [6]. Studies of this sort are needed not only because they shed light on an interesting alloy system with very different end members but more importantly because they will make it easier for others to optimise InAlN growth, allowing more work to be done on the material and increasing the likelihood that it will be incorporated into devices. In particular there has been little investigation of the material by transmission electron microscopy (TEM) or by atomic force microscopy (AFM).

Here we study the effect of ammonia (NH$_3$) gas flux rate in MOVPE on the system by investigating 6 different ~100 nm thick epilayers grown on two different occasions, using AFM, X-ray diffraction (XRD) and TEM.
2. Experimental methods

Our growth was carried out in a Thomas Swan 6 × 2” close-coupled showerhead MOVPE reactor, on pseudo-substrates consisting of ~6 µm of GaN grown on c-plane (0001) sapphire at 1020 °C, following deposition of a 30 nm GaN buffer at 560 °C. All the InAlN epilayers were deposited at 800 °C, employing a trimethylindium (TMI) flux of 16.1 µmol/min, a trimethylaluminum (TMA) flux of 15.8 µmol/min, and a pressure of 50 Torr using N₂ as the carrier gas. The ammonia (NH₃) flux was varied between 44.6 mmol/min (a flux of 1 slm) and 446.4 mmol/min (a flux of 10 slm).

XRD was carried out using a Philips P’analytical PW3050/65 XPert PRO HR horizontal diffractometer using Cu Kα radiation with an asymmetric Ge (220) 4-bounce primary monochromator and analysed using X’pert Epitaxy and Smoothfit 4.0 Philips Analytical B.V. It was used to measure the average lattice parameters of both the epilayers and the pseudo-substrates, using the positions of both 002 symmetric and 105 antisymmetric reflections. These were then used to calculate the epilayer compositions, assuming Vegard’s law to hold for InₙAl₁₋ₙN, isotropic strain in the c-plane of the samples [7] and using literature values of the lattice parameters and elastic constants of AlN and InN [8]. Whilst Vegard’s law may not hold in this case, the precise nature of any deviations has yet to be studied in detail, so we cannot greatly improve on this approach. Fringe peak spacings were used to find the epilayer thicknesses, and defects were investigated using ω-scans of the 002 and 101 peaks.

Surface morphology was assessed by AFM using a Veeco Dimension 3100 with analysis using WSxM freeware [9].

TEM was carried out using a Philips CM30 operated at 300 kV, and samples were prepared by grinding and polishing to ~30 µm and then thinned to electron transparency using a Gatan precision ion polishing system.

3. Results and discussion

3.1. Overview of samples

The two sample series studied here were grown on separate occasions a few months apart, and each investigates a different range of NH₃ fluxes. The first, series A, used flows of 1 slm, 2 slm and 4 slm, and the second, series B flows of 4 slm, 7 slm and 10 slm. However due to process drift the growth temperature may not have been exactly 800 °C, and may have moved slightly between experiments. This is due to several practical reasons, for example the build up of material on the susceptor used and the difficulties of temperature calibration to an accuracy of a few degrees at 800 °C.

3.2. Initial investigation of epilayers: compositions and surface morphology

Initial experiments on all 6 wafers measured the epilayer composition by XRD, and also assessed the surface morphology by AFM using 1 µm × 1 µm scans. The distinguishing features of the surface are broad ‘hillocks’ ~50 nm across and 2-3 nm high, and pits which we initially hypothesized to be linked to dislocations due to similar features being linked to dislocations in other nitride systems [10, 11]. Table 1 gives the compositions, roughnesses and pit densities for all samples. Example AFM scans for each sample are shown in Fig. 1.

### Table 1. The initial measurements for all samples investigated. All measurements from AFM scans (surface roughness and pit densities) come from the average of 4 scans with standard errors quoted.

| Sample | NH₃ flux / slm | In incorporation / % | Roughness at 1 µm / nm | Pit density / µm⁻² (from 1 µm scans) |
|--------|----------------|----------------------|------------------------|-------------------------------------|
| A1     | 1              | 12.9 ± 1.4           | 0.29 ± 0.01            | 3.0 ± 1.0                           |
| A2     | 2              | 15.1 ± 1.4           | 0.44 ± 0.05            | 3.5 ± 0.3                           |
| A3     | 4              | 17 ± 1.45            | 0.77 ± 0.11            | 68.5 ± 9.3                          |
| B1     | 4              | 14.6 ± 1.4           | 0.70 ± 0.04            | 4.5 ± 0.9                           |
| B2     | 7              | 14.4 ± 1.35          | 0.87 ± 0.08            | 31.3 ± 2.6                          |
| B3     | 10             | 14 ± 1.4             | 0.64 ± 0.06            | 16.5 ± 2.9                          |
The existence of process drift becomes apparent when comparing samples A3 and B1, which are grown under nominally identical conditions with a NH$_3$ flux of 4 slm. The difference in composition could be explained if our reactor was running slightly hotter for series B, by a difference of approximately 10 °C. Despite this some growth trends can be distinguished, the indium incorporation increases for smaller NH$_3$ fluxes between 1 slm and 4 slm, but there is no further increase as the flux is increased beyond 4 slm. The surface roughnesses are of a comparable order of magnitude for all samples, with a slight increase from 1 slm to 4 slm which levels out at higher flows. However other details do not have an obvious explanation, particularly the pit density. If the pits are linked to threading dislocations it seems odd that sample A3 should have an order of magnitude increase in dislocation density as it is the closest to lattice matching. It also seems strange that there is an increase in pit density from B1 to B3, as there is no change in composition or epilayer strain state and therefore no reason for extra threading dislocations to be generated in the epilayer. These questions warranted further investigation.

![Figure 1. The surface morphology of all samples measured by AFM at 1 µm, showing nanoscale pits and hillocks.](image)

3.3. The further investigation of defects by XRD and TEM

The first method used to see if there was additional dislocation generation in the InAlN epilayers was to compare the XRD peak widths of the 002 and 101 peaks for both the GaN pseudo-substrate and the InAlN epilayers using $\omega$-scans. The 002 peak would be broadened by the introduction of screw and mixed type threading dislocations and the 101 by edge, mixed and screw. However, no significant difference was seen between the InAlN epilayer and the GaN peaks for any sample, and all samples had similar values (Tab. 2). Samples A3, B1 and B3 were then investigated by TEM using weak beam dark field (WBDF) imaging to view dislocations in cross section. However, no threading dislocations created at the GaN / InAlN interface were observed in B1 or B3, and only a few in A3 (Fig. 2). Our cross section samples had a volume corresponding approximately to an area of surface 100 nm by 5 µm, so if all the pits observed in AFM were created by such threading dislocations we would expect to see more in our sample.

| Sample | 002 peak | 101 peak |
|--------|----------|----------|
| A1     | $228 \pm 7 (244 \pm 7)$ | $399 \pm 7 (383 \pm 7)$ |
| A2     | $221 \pm 7 (239 \pm 7)$ | $404 \pm 7 (386 \pm 7)$ |
| A3     | $227 \pm 7 (242 \pm 7)$ | $394 \pm 7 (381 \pm 7)$ |
| B1     | $220 \pm 7 (241 \pm 7)$ | $397 \pm 7 (378 \pm 7)$ |
| B2     | $210 \pm 7 (240 \pm 7)$ | $366 \pm 7 (376 \pm 7)$ |
| B3     | $230 \pm 7 (242 \pm 7)$ | $399 \pm 7 (380 \pm 7)$ |
The evidence from X-ray peak widths and from TEM both strongly suggest that some of the additional pits observed in AFM are not caused by threading dislocations originating at the GaN / InAlN interface. While the threading dislocations observed do cause pits it may be the case that in some samples with large pit densities there are other pit generating mechanisms operating. It may also be the possible that dislocation loops are generated in the thickness of the film too close to the surface to be observed in our TEM studies to date.

![Figure 2. TEM of samples A3 and B1. WBDF images using g vectors of 0002 and 1120 respectively leave the screw and edge components of a threading dislocation visible.](image)

4. Conclusions

In$_x$Al$_{1-x}$N is a useful and interesting alloy system with many applications. Here we have investigated the effect of NH$_3$ flux on 100 nm In$_x$Al$_{1-x}$N epilayers. Indium incorporation increases between 1 slm (44.6 mmol/min) and 4 slm (179 mmol/min), but remains constant thereafter up to 10 slm (446 mmol/min). Under certain conditions the surfaces showed a significant number of pits similar to those caused by threading dislocations, but the mechanism for their generation remains an open question.

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