GaN devices based on nanorods

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Abstract. Transmission and scanning electron microscopy are used to examine the role of an intermediate nanorod layer in reducing threading defect densities in GaN/(0001)sapphire. Films grown by molecular beam epitaxy under N-rich conditions showed Ga-polar nanorods growing out of a more compact N-polar layer. The nanorods sometimes contained extended threading defects, which were faulted dipoles lying on \{10\textsuperscript{-10}\} planes with a displacement vector of \(\pm 1/2[0001]\), which act as sources for spiral growth. By overgrowing the nanorods under Ga-rich conditions, continuous epilayers were formed with threading defect densities down to \(10^8\) cm\(^{-2}\). In a second approach, nanorods produced by etching through a self-organised layer of Ni islands were overgrown by metal-organic chemical vapour deposition, to produce overlayers with defect densities down to \(5\times10^7\) cm\(^{-2}\). In both cases, the mechanisms by which the nanorod layer reduces the threading defect density are identified.

1. Introduction

A major issue which currently affects the efficiency of GaN devices is the high density of threading defects (up to \(10^9\) cm\(^{-2}\)), which arise through growing epilayers on highly mismatched substrates. Epitaxial lateral overgrowth (ELOG) is one solution to this problem. In ELOG, “seed” columns of GaN, defined by masking and etching procedures, and usually several micrometers across, are induced to grow laterally until a continuous overlayer is formed. However, although threading defect densities in the laterally-grown “wings” can be reduced to around \(10^6\) cm\(^{-2}\), threading defect densities remain high in the seeds, which might comprise 20\% of the layer. The wings also often exhibit grain rotations, typically up to 0.5-1º, which lead to new grain boundary defects [1]. In addition, the mask deposition, patterning and etching steps add to overall costs. In order to circumvent some of these problems, we have investigated alternative “nano-ELOG” approaches where the seed columns are a “compliant” layer of nanorods, whose diameters and spacing are in the range 20-100nm, i.e. about two orders of magnitude less than in conventional ELOG.

In this paper, transmission and scanning electron microscopy (TEM and SEM) are used to examine two different nano-ELOG methods. In the first method, molecular beam epitaxy (MBE), at the University of Nottingham, has been used to nucleate and grow nanorods under strongly N-rich conditions on (0001)sapphire. Lateral overgrowth is then initiated under more Ga-rich growth conditions to form a continuous overlayer. The morphology of growth, and the nature and role of a new type of threading defect present in the nanorods will be explained. When the nanorods grow
together further threading defects are generated. The origin of threading defects present in the final overlayer will be examined and the factors controlling their density will be discussed. In the second nano-ELOG method, carried out in the University of Bath, nanorods are produced by etching a continuous GaN epilayer through a self-organised mask. Following removal of the mask material, metal-organic chemical vapour deposition (MOCVD) is used to produce lateral growth of the GaN to form a continuous layer. Again, the role of the nanorod layer in reducing the threading defect density will be discussed.

2. The MBE method
The growth of GaN nanorods by MBE has been investigated quite extensively (see [2] for a review). It is clear that, in general, nanorod growth is favoured under strongly N-rich conditions, is sensitive to surface preparation (particularly the presence or absence of an AlN interlayer) and can be non-catalytic. In addition, growth under Ga-rich conditions favours a transition to 2-dimensional growth.

In our case, samples were grown by plasma-assisted MBE in a Varian ModGen II MBE system with active nitrogen provided by an HD25 RF activated plasma source. An AlN buffer layer 2-10nm thick was grown at a temperature of 600°C prior to GaN growth which was carried out at substrate temperatures in the range 700-800°C. Growth was carried out for up to 6hrs under strongly N-rich conditions, and subsequently for 0-6hrs under Ga-rich conditions.

![Figure 1. SEM image of nanorods grown by MBE under N-rich conditions](image)

2.1 N-rich growth
Figure 1 shows a SEM image taken after N-rich growth, indicating an array of high aspect ratio nanorods. Figure 2 shows a more detailed view of the microstructure. The cross-sectional TEM image in Figure 2a reveals a bimodal microstructure in which parallel sided nanorods showing bend contours but little evidence of defects emerge from a more compact but rough layer. This compact, or intermediate, layer contains high densities of threading defects, some of which are in contrast in Figure 2a along with high densities of basal plane stacking faults which are out of contrast. From the SEM image in Figure 2b, it can be seen, at least qualitatively, that the nanorods mostly grow out of depressions or holes in the intermediate layer. The image in Figure 2c shows a plan-view sample obtained by coating the deposit in epoxy resin, and backthinning to reveal the tops of nanorods. It can be seen that the nanorods are facetted (with {10-10} surfaces), and that where nanorods have touched during growth, a liquid-like coalescence has taken place. The nanorods appear mostly perfect, but some have contrast indicative of threading defects, as discussed below.
Convergent beam electron diffraction (CBED) patterns, taken with the 0002 systematic row symmetrically excited, show strong asymmetry between the ±(0002) reflections, where [0001] is defined as the Ga-N bond direction. The asymmetry, which depends on double diffraction between systematic row reflections, can be used to distinguish the polarity of the GaN layer. CBED analysis showed that the nanorods in Figure 2 were Ga-polar (growing in the [0001] direction), while the intermediate layer was N-polar (see [3] for more details).

Figure 2. Samples grown under N-rich conditions (a) TEM cross-section revealing nanorods growing out of holes in a more compact layer, (b) SEM image suggesting that the nanorods emanate from holes in the compact layer, (c) TEM image of a plan-view sample intersecting the tops of nanorods. This shows good alignment of the nanorods, a liquid-like coalescence where nanorods are starting to merge. There is contrast in some of the nanorods suggesting the presence of threading defects.

2.2. Defects in nanorods
More in-depth studies of the structure of the nanorods suggested that, while many were perfect single crystals, others contained threading defects. Figure 3 compares perfect nanorods in Figure 3a with an imperfect nanorod in Figure 3b. The imperfect nanorod is seen to have an extended threading defect and terminates in a faceted growth surface, while the perfect nanorods appear to have relatively flat (0001) growth surfaces. By tilting the imperfect nanorods about their axes, it was found that the threading defects were planar. This is illustrated in Figure 4, which shows two nanorods which have
been rotated by about 70° about [0001]. In both cases, 0002 bend contours cross the nanorods, and are displaced on crossing the threading defects. The left hand nanorod shows an extended defect which is obliquely inclined in one image and edge-on in the second (the projection of the defect is indicated by arrows), whereas, coincidentally, the defect in the second nanorod shows the opposite behaviour.

![Figure 3.](image)

(a) perfect nanorods, (b) a nanorod with a threading defect

![Figure 4.](image)

Nanorods with faulted planar defects, seen in images which are relatively rotated by about 70° about a vertical axis. The fault plane for nanorod 1 is approximately edge-on in the left hand image, while the fault in nanorod 2 is edge-on in the right hand image. The bend contours, which are of 0002 type, are displaced on crossing the bounding partials, which are arrowed.

Tilting experiments suggested that the defects in Figures 3b and 4 lie on {10-10} planes. The displacement of 0002 bend contours on crossing these defects suggests relaxation due to a displacement field parallel to [0001]. In contrast, the defects showed little contrast in a-type reflections.
suggesting that displacements in the basal plane were either zero or very small. In fact, an analysis of how the 0002 contours were displaced on crossing the defect suggested that the defects were bounded by a dislocation dipole with Burgers vector ±c/2 [4]. This implies that these threading defects were faulted, with a change in stacking sequence across the fault plane from ABABAB to BABABA.

Although this type of fault is unusual, it has been previously reported in GaN epilayers, for example by Tanaka et al [5] on the basis of high resolution images. We believe that the faults in nanorods provide a mechanism of nanorod growth as illustrated in Figure 5. This shows a top view in which the opposite c/2 partials associated with the fault provide a step on the (0001) growth surface one Ga-N layer in height, i.e. a single surface step, where two such steps represent a unit cell height (0.52nm). This step provides a spiral growth mechanism, with intermediate stages in a single repeat illustrated in the diagram. The faceted (and, in fact, vicinal) nature of the top surface can be readily explained by step flow from the source.

![Figure 5](image.jpg)

**Figure 5.** Schematic suggesting how the threading defects present in nanorods initiate a spiral growth mechanism. Viewed from the top, A and B are the emerging partials connected by a single c/2 step on the growth surface. Addition of adatoms to the step causes step flow in the sequence 1-4, whereupon the step is re-formed and the sequence repeats.

Close examination suggested that the faults emanated from points close to the sapphire surface. An example is illustrated in the two dark field images in Figure 6. The top image shows a general view. Threading defects are present along the length of the two arrowed nanorods. These nanorods are bent, such that the diffraction conditions change along their length. However, in places, the pairs of bounding partials associated with the threading defects can be seen quite clearly. These partials appear to converge to points close to the sapphire surface; the bottom image has been taken at a slight tilt to the top image to enhance the contrast of the partials in the boxed region as the sapphire surface is approached. However, the exact nucleation site remains unclear at present. One possibility is that nucleation takes place at steps on the sapphire surface, where a single step height of 0.22nm compares quite closely to the fault displacement of c/2 = 0.25nm. However, further work is needed to confirm this possibility.
2.3. Ga-rich growth

In samples subjected to N-rich followed by Ga-rich growth, it was found that the nanorods grew laterally to eventually form a continuous overlayer. Figure 7 shows an intermediate stage, where the nanorod diameter has increased, but nanorods remain mostly separate. Magnified images show the regions A and B more clearly. The region A shows two threading defects. One of these defects is seen to terminate. This could be due to the defect meeting the surface of the thin foil. However, termination of the threading defect is also consistent with recombination of the bounding partials, which have opposite Burgers vectors, to form a closed dislocation loop. In the region near B, it is apparent that the second defect has opened into a void. Again, this effectively terminates the threading defect, since there is no net dislocation, or fault, in the void. Thus, overgrowth of the void, should it eventually occur, would lead to perfect crystal with no net threading defect.

Figure 6. Dark field images in $g = 0002$. The top image shows two nanorods (arrowed) which contain threading defects. The defects extend right along the length of the nanorods, but are only partly visible due to bending of the nanorods (the bounding partials can be seen quite clearly in places). The image at the bottom, which is taken at a slight tilt to the top image, shows the boxed region at a higher magnification, and tracks the bounding partials (arrowed) to a point close to the sapphire interface.
Figure 7 shows a sample grown by MBE under N-rich conditions followed by a period of growth under Ga-rich conditions. The top image is a general view showing that, under Ga-rich conditions, lateral growth is faster towards the tops of nanorods (i.e. towards the top left). The bottom images show expanded views of the regions near A and B.

Figure 8 shows an example where Ga-rich growth has led to a continuous overlayer. The image is taken with an $a$-type reflection strongly excited, to highlight misorientations due to relative rotations about the growth axis. The image shows clearly that many neighbouring nanorods coalesce to form regions of perfect crystal without threading defects. However, on a larger scale, there are grain boundaries present which contain threading defects. Overall, the density of threading dislocations was around $10^8 \text{ cm}^{-2}$, up to two orders of magnitude less than in continuous epilayers of an equivalent thickness.
Figure 8. TEM cross-section showing a sample grown for 5hrs under N-rich conditions, followed by 5hrs under Ga-rich conditions to form a near-continuous overlayer. The image is taken under near 2-beam conditions with an \( a \)-type reflection operating, such that grain boundaries (arrowed) representing relative rotations about the growth directions are clearly visible. Basal plane stacking faults visible in the central region indicate that lateral growth extends about 1\( \mu \)m. Growth direction is upwards.

3. The MOVCD method

MOCVD has proved less effective than MBE for the growth of \( c \)-oriented nanorod arrays, but is well-established as a means of achieving lateral overgrowth. Thus, in this case, arrays of nanorods were fabricated by post-processing of continuous (0001)GaN epilayers grown on (0001)sapphire substrates by MOCVD. These epilayers, grown and subsequently processed in the University of Bath, generally had densities of threading defects in the range 1-3\( \times \)10\(^8\) cm\(^{-2}\). Nanorods were then produced in the epilayers by wet and dry-etching through a self-organised mask of Ni islands produced by annealing a thin vapour-deposited Ni layer [6]. Arrays of nanorods produced in this manner are illustrated in Figure 9, the density of nanorods being about 10\(^9\) cm\(^{-2}\). Since the density of nanorods was greater than that of the threading defects, most nanorods were free of threading defects, in contrast to the situation with optically defined masks.

Continuous overlayers produced by lateral overgrowth of the nanorod arrays by MOCVD showed a significant reduction in the density of threading defects compared with the original epilayers. In some cases lateral overgrowth preserved the airgaps between nanorods, while in other cases, complete closure of the airgaps resulted depending on the pre-processing treatment. Figure 10 shows an example of the latter, indicating extensive lateral migration of threading defects and, thereby, the region of lateral growth. Figure 11 shows an example where nanorods were etched partially through the GaN epilayer, and the airgaps have been partially closed by lateral overgrowth. Although the detailed structure in the region containing the nanorods is not clear, it can be seen that the overlayer has fewer defects than the original epilayer. Threading defects are seen to migrate laterally in the region just above the nanorods indicating again the region of lateral growth. In this region, it was found that dislocation reactions occurred, reducing the density of threading defects to 5\( \times \)10\(^7\) cm\(^{-2}\), i.e. by nearly an order of magnitude compared with the original epilayer.
Figure 9. SEM image showing nanorods produced by etching through a mask of Ni islands.

Figure 10. This shows nanorods produced by etching, which have been subsequently overgrown by MOCVD, generating extensive lateral migration of threading dislocations.

Figure 11. TEM cross-section, showing a sample where nanorods have been etched part-way through a GaN epilayer, and then overgrown by MOCVD. The overlayer shows some lateral migration of dislocations and a lower average density of threading defects than in the original epilayer.
4. Conclusions
The results show that both the MBE and MOCVD methods, although quite different, produce significant reductions in the density of threading defects. In the MBE approach, the nanorods provide seed columns, which are either perfect single crystals or contain threading defects. Some of the threading defects in nanorods terminate by recombination of the partials or in voids, such that relatively few of these defects are found in the final overlayers. There is a high density of threading defects in the imperfect intermediate layer, but these terminate at an airgap and do not propagate into the final overlayer. The majority of threading defects are formed when a continuous overlayer is formed, describing the misorientations and relative displacements between nanorods which must exist at nucleation. As groups of neighbouring nanorods coalesce to form defect-free regions, we must suppose that some of these displacements are accommodated elastically. However, elastic accommodation becomes less likely as the coalescence regions increase in size, leading eventually to the larger scale grain boundaries seen in Figure 7. The MOCVD approach shows some similar features. Although there are additional processing steps, the nanorods are again relatively defect-free. Lateral overgrowth in this case tends to close the airgaps, but lateral migration of the threading defects towards the tops of the nanorods again reduces the overall density of threading defects.

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