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Indium Nitride and Indium Gallium Nitride layers grown on nanorods

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Abstract. Molecular beam epitaxy has been used to grow InN layers on both Si and SiC substrates and In_{0.5}Ga_{0.5}N layers on Si substrates using a nanorod precursor array. Transmission electron microscopy (TEM) studies show that nanorods grown first under N-rich conditions, and then under more metal-rich conditions to promote lateral growth are free of dislocations until coalescence occurs. At coalescence, dislocations are introduced at grain boundaries. These are predominantly twist boundaries, with better epitaxial alignment seen on SiC substrates. The lateral growth of In_{0.5}Ga_{0.5}N is shown to be cubic, tentatively ascribed to the growth of basal plane stacking faults at the start of the lateral growth and the low growth temperatures used.

1. Introduction

In order to develop high efficiency solar cells the choice of material becomes very important. The majority of current multi-junction solar cells combine different materials systems that are lattice-matched and each with a different band gap range. This has advantages over a single junction solar cell as band gaps of the materials used may be tuned to increase the amount of absorption at different regions of the solar spectrum.

In_xGa_{1-x}N has been proposed as a single materials system for a multi-junction solar cell as the band gap varies from 0.7eV ($x=0$) to 3.4eV ($x=1$) which spans the whole visible spectrum. Previous attempts to grow In_xGa_{1-x}N solar cells had low efficiencies [1] when compared to theoretical models [2] as films grown had suffered from high defect densities, and possibly phase separation at high In contents. Molecular beam epitaxy (MBE) is an effective method of growing high In content alloys of reasonable quality, but requires low growth temperatures as the In-N bond is quite weak. This means that at temperatures greater than 450°C indium desorbs more readily leading to variations in indium content [3].

One way to overcome defects is to start growth as nanorods before coalescence. Previous work on GaN grown on sapphire has shown that it is possible to first grow nanorods which are defect free, with dislocations only forming as a continuous film is grown on top of the rods [4].

This paper demonstrates the ability to grow InN and In_xGa_{1-x}N nanorods on silicon (Si) and silicon carbide (SiC) and suggests growth mechanisms. By varying the growth conditions it is shown that



coalescence of nanorods result in overgrowths which have grains of the order 500nm wide which span multiple nanorods.

2. Experimental Method

InN and $\text{In}_x\text{Ga}_{1-x}\text{N}$ nanorods have been grown directly on Si substrates by plasma assisted MBE in a Varian ModGen II system. Nanorods were grown for 5hrs at temperatures between 350-400°C in N-rich conditions. For lateral growth the In and Ga fluxes were increased for a growth time of 3hrs. InN samples were grown on SiC using similar conditions in a custom built MBE system with a RF plasma nitrogen source and a K-cell for the indium flux.

TEM was performed on a Philips EM430 at 200kV using imaging, selected area diffraction (SAD), convergent beam electron diffraction (CBED) and energy dispersive X-ray spectroscopy (EDX) techniques. Cross section and Plan view samples were prepared for TEM by mechanical polishing by diamond lapping films to less than 100µm thickness, then ion thinned using a Gatan PIPS, first at 5kV until a small hole began to form and finishing with a 15 minute polish at 3kV to reduce surface damage.

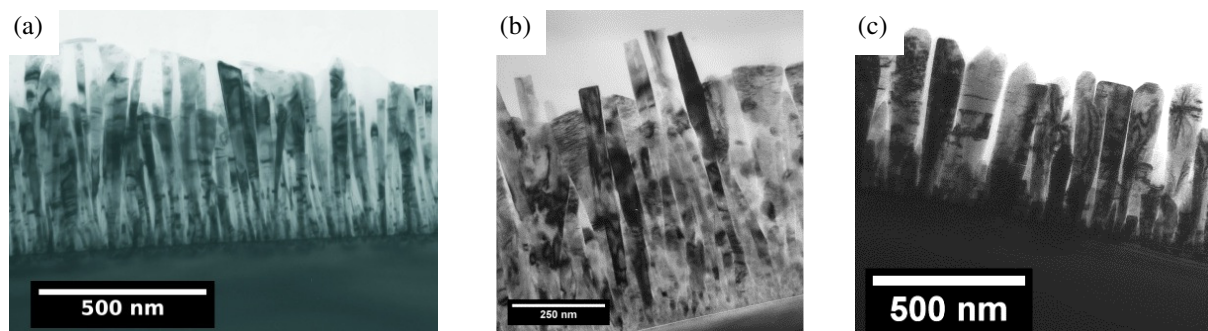


Figure 1. Shows cross-section TEM of N-rich growth of (a) InN on Si, (b) InN on SiC and (c) $\text{In}_x\text{Ga}_{1-x}\text{N}$ ($x=0.5$) on Si.

3. Results

Figures 1(a) and 1(b) show TEM of InN growth on Si and SiC grown under N-rich conditions at 400°C. On Si (Figure 1(a)) the InN nanorods grown have lengths of $250\pm 30\text{nm}$ and have a degree of lateral growth. These nanorods have no threading dislocations, with the only visible contrast being bend contours. In comparison the TEM of InN nanorods grown on Si-polar SiC (Figure 1(b)) show a dense forest of short nanorods $\sim 600\text{nm}$ long which have some lateral growth and some nanorods which grow 100nm longer. CBED has shown that nanorods of both lengths have In-polarity; consequently the difference in length has been attributed to competitive growth. Figure 1(c) shows TEM of $\text{In}_x\text{Ga}_{1-x}\text{N}$ nanorods grown on Si substrates. These were grown in N-rich conditions at a temperature of 400°C. They show evidence of some tapering as seen in the InN and do not have dislocations.

Plan view TEM of a sample grown on Si first under N-rich conditions to promote nanorod growth, and then under In-rich growth to promote lateral growth, is shown in Figure 2(a). Coalescence of the InN nanorods occurred, forming a nearly continuous film. However, significant rotational misalignments of the nanorods about the c-axis prior to coalescence caused the formation of large and small angle grain boundaries. Figure 2(b) shows a cross-section TEM image of InN grown on SiC with the same growth processes as on Si, which resulted in coalescence of the nanorods occurring right down to the substrate. In some areas where coalescence has occurred screw and mixed dislocations are generated, however some grains have no dislocations present and are 500nm wide which indicates that a good epitaxy of the nanorods was achieved before coalescence on the SiC substrates. $\text{In}_x\text{Ga}_{1-x}\text{N}$ coalescence is shown in figure 2(c). It is clear here that the lateral overgrowth

has led to coalescence, creating large defect-free grains wider than 500nm which span multiple nanorods.

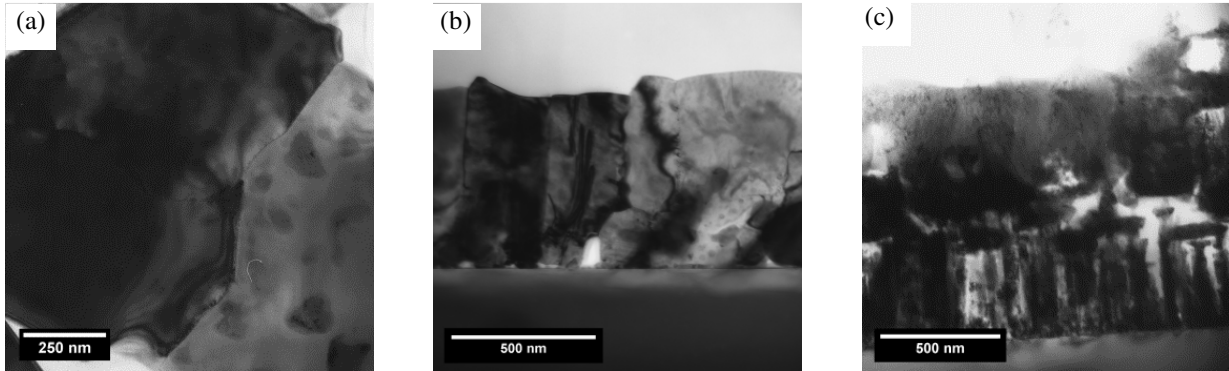


Figure 2. TEM of N-rich growth followed by metal-rich growth. (a) Plan view of InN on Si (b) cross-section of InN on SiC (c) cross-section of $In_xGa_{1-x}N$ ($x \sim 0.5$) on Si.

Further investigation of the overgrown films is shown in Figure 3. A [110] SAD pattern confirms that the overgrowth region is cubic $In_xGa_{1-x}N$ grown on hexagonal nanorods. Also visible in Figure 3 are multiple stacking faults at the base of the overgrowth region which extend to the edge of the crystals. Towards the top of the lateral growth region, where the cubic material is present, there are {111} stacking faults inclined at 70° to the basal plane after which the phase reverts to hexagonal. In Figure 4, EDX has been taken from the base and the top of a nanorod grown in N-rich conditions. At the bottom of the nanorod the indium content is 9%, which increases to 55% at the top of the nanorod.

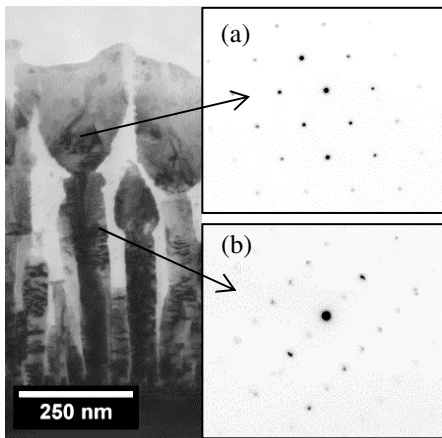


Figure 3. TEM of an $In_{0.5}Ga_{0.5}N$ nanorod grown on Si, first with N-rich conditions then metal rich conditions with insets (a) a [110] SAD pattern and (b) a $[11\bar{2}0]$ SAD pattern.

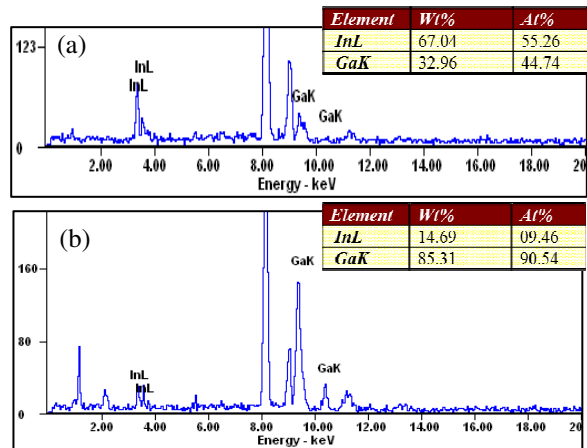


Figure 4. EDX taken from a nanorod grown under N-rich conditions. (a) EDX from the top of a nanorod has a composition $x=0.55$ and (b) EDX from the bottom of a nanorod has a composition $x=0.09$.

4. Discussion

When considering the growth of InN, the general trend is for the nanorods to taper as the growth progresses. This was seen on both Si and SiC substrates. One possible growth mechanism is that at temperatures as high as 400°C, the In atoms have enough energy to desorb during the start of growth increasing the concentration of In atoms at the growth surface. As the growth progresses, the increased indium concentrations promote more lateral growth, which results in tapering of the nanorods.

The growth of $\text{In}_x\text{Ga}_{1-x}\text{N}$ is slightly more complicated by the addition of Ga atoms. However at temperatures as low as 400°C it may be assumed that once a Ga atom adsorbs it is not likely to desorb, when compared to In atoms. In this case a similar model may be proposed as for the InN. As the growth begins, very few In atoms adsorb without subsequently desorbing, and when they desorb can be replaced by a Ga atom. As the growth continues, the concentration of In increases, which increases the likelihood of a desorbed In atom being replaced by a second In atom. This results in Ga-rich material at the start of the growth and more In-rich material towards the end of the growth. This has been confirmed by the EDX in figure 4(b) which shows an increase of In content as the growth progresses and is in agreement with other work on $\text{In}_x\text{Ga}_{1-x}\text{N}$ nanorods [5].

The coalesced regions with cubic regions may grow due to the stacking faults, which extend to the edges of the crystal, locally changing the stacking sequence from hexagonal (...ABAB...) to a cubic sequence (...ABCABC...), thus providing a cubic substrate for subsequent growth. Other work has shown it was possible to grow cubic GaN at low temperatures (~680°C)[6], this could possibly explain why the cubic growth continues.

5. Conclusion

The growth of $\text{In}_x\text{Ga}_{1-x}\text{N}$ nanorods over the composition range $x=0$ to $x=0.5$ has been demonstrated. Over this range the nanorods grown show no evidence of dislocations, only bend contours. After lateral growth, defects form due to rotational misalignment of the nanorods. This is more prevalent on Si than on SiC where InN nanorods show better alignment. $\text{In}_x\text{Ga}_{1-x}\text{N}$ nanorods show a segregation of the In content, with In-rich material at the tops of the rods. In the lateral growth of $\text{In}_{0.5}\text{Ga}_{0.5}\text{N}$ material, we have confirmed the presence of cubic zinc blende material which is seeded by basal plane stacking faults and continued due to low growth temperatures, before reverting to the hexagonal phase.

Acknowledgements

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