AN ATOMIC FORCE MICROSCOPY STUDY OF THE INITIAL NUCLEATION OF GaN ON SAPPHIRE

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ABSTRACT

Preliminary results of a study of GaN nucleation and growth by molecular beam epitaxy using a nitrogen rf plasma source are presented. Nucleation layers and 3000 Å thick layers were investigated by atomic force microscopy and x-ray diffraction. Growth under gallium-rich conditions both increased nucleation island size and promoted two-dimensional growth.

INTRODUCTION

The potential applications of blue and ultraviolet optoelectronic devices based on GaN have been recognized for many years [1]. Recent advances in epitaxial GaN growth by metal organic chemical vapor deposition (MOCVD) have lead to commercially available devices from both U.S. (CREE Research, Inc.) and foreign companies (Nichia Chemical Industries). Rapid progress in this direction is also being accomplished by molecular beam epitaxy (MBE) growth using active nitrogen species [2]. The most common substrate for epitaxial growth is sapphire (α-Al_2O_3) due to its availability, low cost and robust nature. GaN layers grown on sapphire, however, typically contain a high density of defects, mainly threading dislocations, due to a large lattice mismatch and thermal expansion coefficient mismatch between the epilayer and the substrate [3,4]. In contrast to growth in other semiconductor systems, these high dislocation densities ($>10^9$ cm^{-2}) are reported to persist for up to 4 µm of growth [3,4]. While some devices are tolerant of such high densities [4], it is desirable to determine growth conditions for improved structural quality.

GaN typically nucleates and grows on sapphire by island formation. The use of low temperature buffer layers (450 - 600 ^{o}C) of AlN [5] or GaN [6,7] results in a dramatic improvement in layer morphology and electrical properties. Annealing prior to high temperature growth causes coalescence of the nucleation islands, resulting in low angle grain boundaries which create the observed dislocation arrays [3,4]. This subgrain structure is stable during growth under most conditions. Thus, subsequent crystal quality is strongly dependent on the nucleation layer.

The predominant growth mode is a further factor in dislocation reduction with increasing layer thickness. Two-dimensional growth results in the highest degree of structural perfection in epitaxial layer growth. Typical MBE and MOCVD growth conditions appear to promote three-dimensional growth [8]. This may lead to individual growth of the low angle grains, preventing dislocation recombination and annihilation. A recent study has reported MOCVD growth conditions resulting in two-dimensional step-flow growth [9], with a concomitant reduction in dislocation density to about 2×10^8 cm^{-2}. This paper presents the initial results of a study to determine conditions for MBE growth of GaN that result in buffer layers with increased grain size as well as determining the appropriate parameters to promote two-dimensional growth.

EXPERIMENT

The GaN layers for this study were grown at West Virginia University by MBE in a system similar to that described elsewhere [10]. Since we are interested in developing lower temperature growth of GaN by MBE, we have only investigated growth temperatures less than 700°C. A standard MBE source provided the Ga flux. A cryogenically-cooled rf plasma source (Oxford Applied Research CARS-25) operating at 500W was used to produce the active nitrogen flux. The layers were characterized using x-ray diffraction and atomic force microscopy (AFM) (Digital Instruments Nanoscope II).

RESULTS

Several studies [3,4,11] have indicated that the transition between buffer layer and "bulk" film structure occurs in the first 0.4 to 0.5 μm of growth. We grew a series of 3000 Å layers to determine both growth rates and growth modes in this transition region. We were also interested in the transition point between Ga-rich growth and conditions which produced Ga condensation as evidenced by the presence of Ga droplets. Figure 1 illustrates our growth rate for several conditions. Above 2.5 $\times 10^{-7}$ Torr Ga and 600°C, growth is limited by the amount of active nitrogen present as indicated both by the increase in growth rate with temperature at a fixed Ga flux, and by the relatively constant growth rate at a given temperature for increasing Ga flux. The decrease in growth rate between 2.5 and 5.0 $\times 10^{-7}$ Torr Ga at 660°C is apparently related to a change from a three-dimensional to a two-dimensional growth mode, as discussed later.

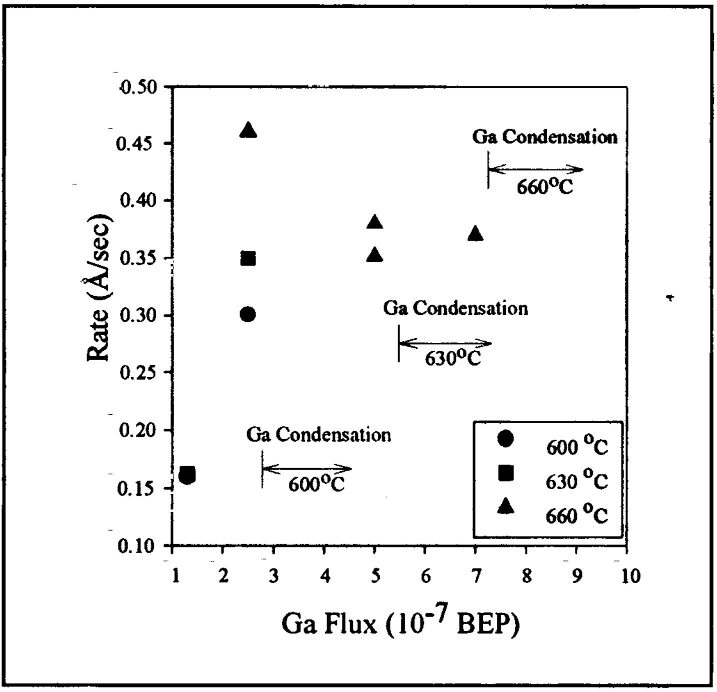


Figure 1. Growth rate of GaN for several temperatures and valus of Ga flux.

Early in this study we grew layers using nucleation conditions reported by others [2,5,6,7,12]. We found that exposing the sapphire substrate to an active nitrogen flux resulted in a fine-grained (<1000Å) island size. Growth of a low temperature nucleation layer at 450 $^{\circ}$C followed by an anneal at 660 $^{\circ}$C also resulted in a similar small grain size. Thus, we undertook a study of island size distributions in nucleation layers as a function of growth parameters. The nucleation layers were studied using AFM. Figure 2 is a micrograph of one such layer. Distributions were determined by taking ten AFM micrographs at points distributed across the sample surface. The islands were approximated as circular regions of various sizes by visual comparison with a template. A histogram of occurrence frequency vs. diameter was thus obtained. The distributions were adequately represented by poisson statistics, and the mean value was found by a least squares fit to the distribution.

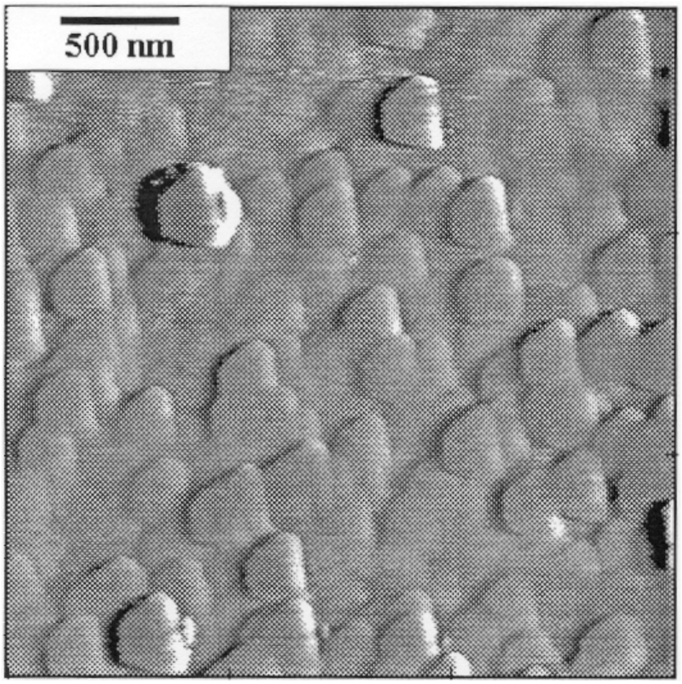


Figure 2. AFM micrograph of a GaN nucleation layer, 100Å thick, grown at 660 $^{\circ}$C under 6 sccm nitrogen flow and 5.0×10^{-7} Torr BEP Ga. The average surface roughness of this layer was about 10 Å. The mean island diameter was about 3000 Å.

The nucleation island diameter was found to increase somewhat linearly with temperature over the range investigated. However, in order to obtain this increase, the nucleation had to occur under Ga-rich conditions. Indeed, at a given temperature, the largest island size always occurred near the boundary for Ga condensation. Our results are summarized in Figure 3. For a fixed Ga flux and nitrogen flow rate, we observed a temperature corresponding to a maximum island size. Above this temperature, the nucleation islands became smaller. At a fixed temperature, island size could be increased by increasing the Ga flux, up to the occurrence of Ga condensation. In contrast, increasing the nitrogen flow rate resulted in smaller island size.

Further information was gained by examining layers grown by extending the nucleation growth to 3000 Å thick. Figure 4(a) is the AFM micrograph of such a layer grown at 660 $^{\circ}$C. The morphology appeared to consist of well-defined three-dimensional microcrystallites, with an average surface roughness of about 150 nm. These growth parameters appear to promote three-dimensional growth without early coalescence of the islands. Also, up to the 3000 Å thickness, there was no apparent change in average microcrystallite dimension from the original nucleation island size. X-ray diffraction measurements were consistent with single-crystal, hexagonal GaN for all layers measured. However, the full width at half maximum (FWHM) was fairly large, about 400 arc minutes for the sample depicted in Figure 4(a). Doubling the Ga-flux brought the growth to near-Ga-condensation conditions and gave an increase in island size, as seen in Figure

4(b). More importantly, however, is evidence of two-dimensional growth which was observed for all layers grown above 660 °C for Ga-rich conditions. Coalescence of the islands is occurring and the overall roughness between grains is reduced to about 20 nm The tops of the islands are fairly flat with well defined sub-nanometer steps corresponding to a few monolayers. We believe this indicates we are near the conditions needed for two-dimensional step-flow growth. The x-ray diffraction FWHM was reduced to about 120 arc minutes for this layer.

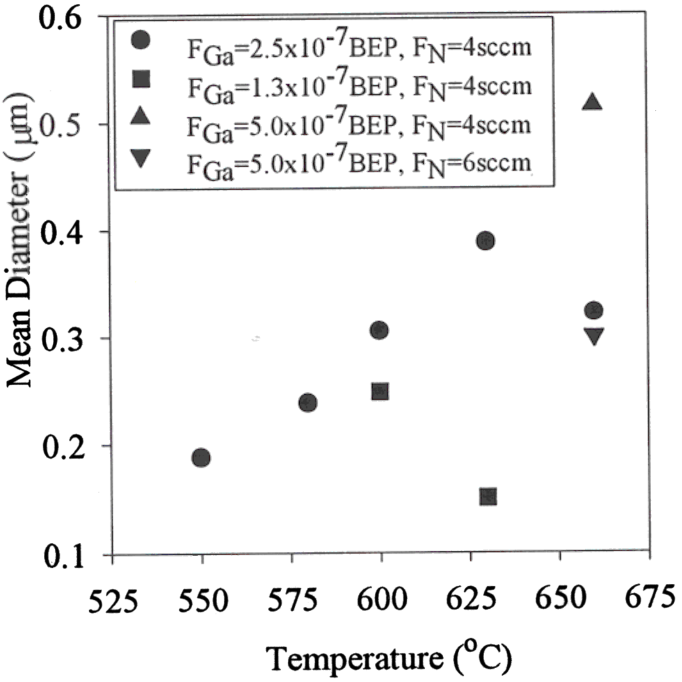
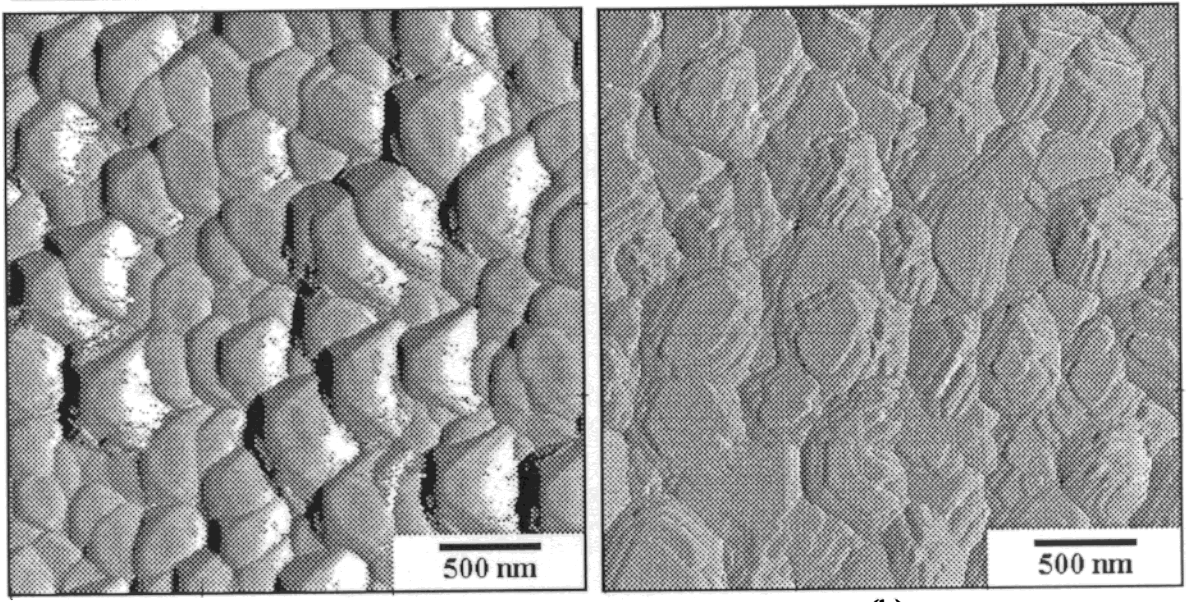


Figure 3 . GaN nucleation island diameter as a function of growth conditions.



(a) (b)

Figure 4 . AFM micrographs of GaN layers, 3000Å thick, grown at 660°C under 4 sccm nitrogen flow and (a) $2.5\text{x}10^{-7}$ Torr BEP Ga (b) $5.0\text{x}10^{-7}$ Torr BEP Ga flux.

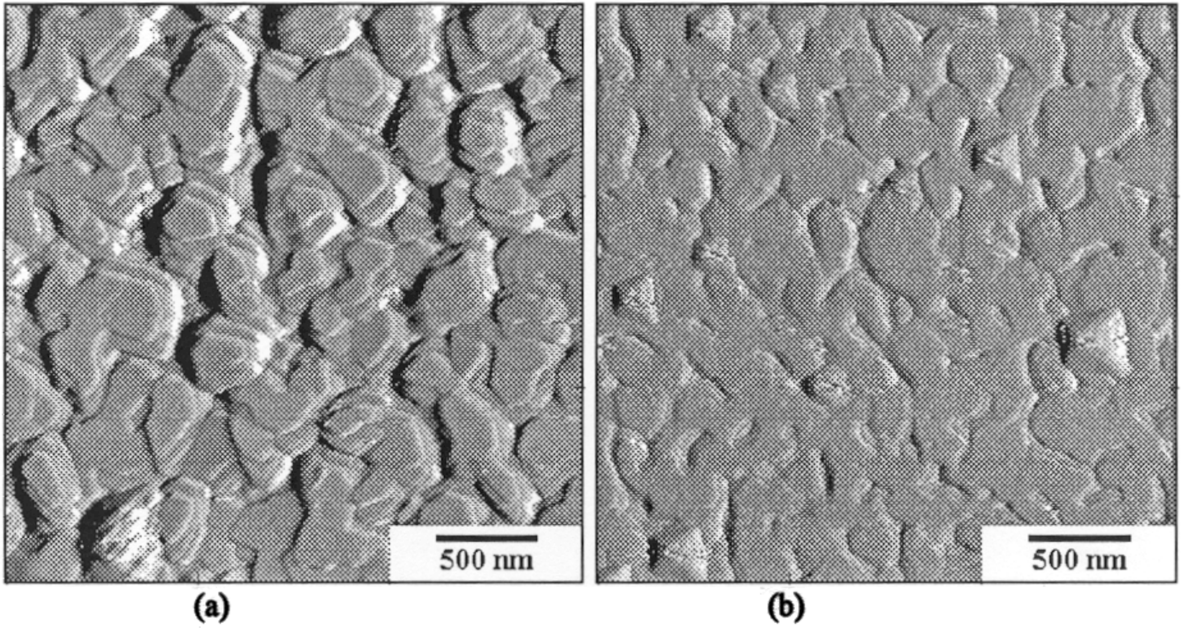


Figure 5 . AFM micrographs of GaN layers, 3000 Å thick, grown at 670°C under 6 sccm nitrogen flow and 5.0×10^{-7} Torr BEP Ga flux: (a) without a buffer layer and (b) with a 100Å thick buffer layer annealed at 670°C for 20 minutes prior to layer growth.

Only layers grown above 660 °C under Ga-rich conditions, where increasing the Ga-flux does not increase the growth rate as shown in Figure 1, exhibit characteristics of two-dimensional growth. All others exhibited a well defined, three-dimensional microcrystallite structure. Figure 5(a) is an AFM micrograph of a layer grown at 670 °C under increased nitrogen flow to move further away from the Ga-condensation point while maintaining Ga-rich conditions. Coalescence is not as evident as for the previous sample, with an average surface roughness of about 30 nm. However, the islands are again flat-topped and steps corresponding to monolayer growth are observed. Figure 5(b) is a 3000 Å layer grown under the same conditions except that growth was interrupted after the first 100 Å of growth. This nucleation layer was then annealed at 670 °C for 20 minutes under nitrogen flux, and growth was resumed. The resulting layer exhibited almost complete coalescence, with an average surface roughness of about 1.5 nm. Sub-nanometer terraces were present on the top of the "flat" regions, again indicating a predominantly two-dimensional growth mode. The x-ray diffraction FWHM obtained for this layer was 51 arc minutes. Further optimization should result in complete coalescence and two-dimensional growth.

CONCLUSIONS

Apparently Ga is the more mobile species under our growth conditions since we obtained larger island sizes and two-dimensional growth only under Ga-rich conditions. Lowering the Ga flux lead to three-dimensional growth with a nucleation layer consisting of smaller island sizes. The nitrogen may only incorporate at available sites near where it initially adsorbs, with little lateral motion before desorbing. The best conditions for growing the nucleation layer appear to be as Ga-rich as possible, near the limit for Ga condensation.

In this study, we have determined conditions for MBE growth of GaN that give large nucleation island size, and promote two-dimensional growth of layers. However, the results presented here are preliminary, as we have not yet performed detailed optical and electrical characterizations. We plan to continue this study by growing thicker layers on annealed buffer layers with the larger island size for further characterization of material properties.

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