

INITIAL STAGES OF MOCVD GROWTH OF GALLIUM NITRIDE USING A MULTI-STEP GROWTH APPROACH

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ABSTRACT

A multilayer buffer layer approach to GaN growth has been developed in which the thermal desorption and mass transport of low temperature buffer layer are minimized by deposition of successive layers at increased temperatures. High quality GaN with featureless surface morphology has been grown on (0001) sapphire substrate by metalorganic chemical vapor deposition using this multilayer buffer layer approach. The lateral growth and coalescence of truncated 3D islands (TTIs) nucleated on low temperature buffer layers at the initial stage of overlayer growth is affected by the thickness of the final buffer layer on which nucleation of TTIs takes place. The effect of the thickness of this buffer layer on the quality of GaN is studied by using scanning electron microscopy, van der Pauw geometry Hall measurements and cathodoluminescence and an optimum value of 400Å is obtained.

INTRODUCTION

The group III nitrides are attractive materials for application to optoelectronic and electronic devices. [1],[2]. A lattice matched substrate for these materials is still not commercially available and they are generally grown on lattice mismatched substrates like sapphire or SiC by a two step growth method. In this approach a buffer layer is grown at low temperature (~450°C) and the second layer is grown at high temperature (950°C - 1050°C) [3],[4]. Because of the lattice mismatch between the sapphire (0001) substrate and GaN, the growth mechanism has been found to proceed in three distinct evolution stages : (1) formation of three dimensional truncated islands (TTIs) on the low temperature buffer layer, (2) coalescence of the TTIs by lateral growth, (3) two dimensional growth after complete coalescence of the TTIs. [5],[6]. To enhance the formation of TTIs on the whole buffer layer and to increase the coverage of them, thermal desorption and / or mass transport of the buffer layer must be suppressed. Recently, we have found that using a multilayer buffer layer approach consisting of layers of GaN grown at different temperature suppresses the thermal desorption and mass transport of the buffer layer and enhances the lateral growth of the TTIs. In this paper, we report the lateral growth behavior of TTIs formed during the initial growth on the low temperature buffer layer and the surface roughness and quality of GaN overlayers grown by this multi-step buffer layer approach.

EXPERIMENTAL

GaN epitaxial films were grown by atmospheric pressure MOCVD using a closed space showerhead reactor. This reactor design utilizes a water-cooled multi-inlet gas distribution showerhead in which the group III and V sources are separately inlet directly (~1cm) above the substrate. Trimethylgallium (TMGa) and NH_3 were used with H_2 carrier gas. C-plane sapphire was used as the substrate. After degreasing the substrate and annealing it in the growth chamber for 30 minutes at 1080°C under H_2 flow, a thin (~100Å) buffer layer of GaN was grown at 450°C and the temperature was raised to 800°C to grow a second buffer layer. The second buffer layer thickness was varied from 0Å to 1600Å to observe the effect of the multi-buffer layer approach and the influence of the buffer layer thickness on the quality of the final overlayer. The overlayer was grown at 950°C with a V/III ratio of 933 and the growth time was varied from 10 minutes to 1 hour to observe the effect of the second buffer layer thickness on the macroscopic evolution of the overlayer. About $2.5\mu\text{m}$ of GaN was grown at 1000°C on the TTIs formed on the multi-step buffer layers to increase the relative lateral growth rate. This results in filling the pits on the surface that arise when the TTIs begin to coalesce and eventually in 2D growth. [7] To obtain a certain growth rate at 1000°C , the V/III ratio was decreased to 373 when the growth temperature was increased to 1000°C . Growth rates for overlayers were $4.4\text{\AA}/\text{sec}$ at 950°C and $2.2\text{\AA}/\text{sec}$ at 1000°C . The evolution stages and surface morphology were examined by SEM, the optical and electrical quality of the overlayer were examined by CL and van der Pauw method Hall effect, respectively. CL measurements were performed at 87K with the acceleration voltage of 15KeV.

RESULTS and DISCUSSION

Figs. 1 (a) - (f) show the cross-sectional and planview SEM pictures of samples grown at 950°C for 30 minutes on buffer layers with three different thicknesses of an 800°C buffer layer. In the planviews one observes that for all buffer layers, the dominant surface features are pyramidal shaped islands with flat tops - TTIs. Fig 1(a) shows the cross-sectional image and (b) shows the planview image for a sample without an 800°C buffer layer. The height of the TTIs is not uniform and the TTIs do not cover the whole area of the buffer layer. The black areas in the planview image are areas which are not covered by TTIs because of thermal desorption and/or mass transport of the low temperature buffer layers. This desorption of buffer layers and nonuniformity of the vertical size of the TTIs cause surface roughness and voids in the grown film even after growing a $4.0\mu\text{m}$ thick GaN film. Fig. 1(c) shows the cross-sectional image and (d) the planview

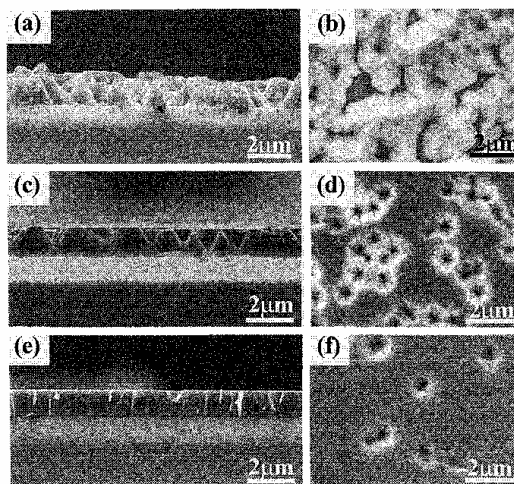


Fig. 1 Cross-sectional and planview SEM images of samples grown 10 minutes at 950°C on buffer layers. The thickness of the second buffer layer grown at 800°C is (a),(b) 0Å, (c),(d) 400Å, (e), (f) 1600Å

image for a sample with a 400Å thick 800°C buffer layer. The height of the TTIs are uniform, cover the whole area, and begin to merge with each other. This sample shows a smooth surface morphology after growing a 4.0µm thick GaN overlayer as shown in Fig. 2.(a). Fig. 1 (e) shows the cross-sectional and (f) the planview image for a sample with a 1600Å thick 800°C buffer layer. The TTIs have merged and a nearly continuous film covers the whole surface. However, after growing a 4.0µm thick GaN overlayer, this sample exhibits a rough surface morphology as shown in Fig. 2.(b). The height of the TTIs for each sample is the same except for the sample grown without an 800°C buffer layer. Comparison of these figure shows that the lateral growth rate of TTIs depends on the 800°C buffer layer thickness. As the thickness of 800°C layer increases, lateral growth of the TTIs and merging of the TTIs is enhanced, but the vertical growth rate does not change. However, beyond 400Å, as the thickness increases, surface roughness of the final film also increases.

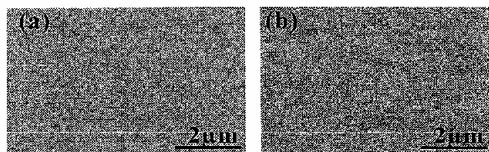


Fig. 2 Planview SEM image of surface of 4µm thick film grown on multi-buffer layer. The thickness of the second buffer layer grown at 800°C is (a) 400Å, (b) 1600Å

Fig. 3 shows the spatially integrated CL spectra of sample grown 4µm at 950°C/1000°C on the buffer layers, 800°C layer thickness of which is varied from 0Å to 1600Å. (a), (b), (c), (d) show the spectrum of sample, 800°C layer thickness of which is 0Å, 400Å, 800Å and 1600Å respectively. Only spectra for samples with 400Å and 800Å thick 800°C buffer layers show near band edge emission at 357nm, and spectrum for sample with the 400Å thick 800°C buffer layer shows higher intensity for the near band edge emission. Decreasing the V/III ratio to obtain higher growth rate during growth of the thick overlayer increases the intensity of the yellowband. By optimizing the growth conditions for the overlayer, the intensity ratio of band edge emission to yellow band emission can be increased to 2.3 in the CL spectrum for the sample with the 400Å thick 800°C buffer under the same conditions for measurement.

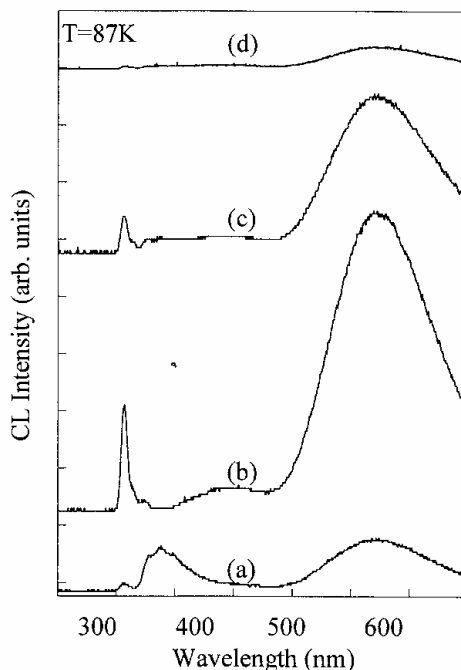


Fig. 3 CL spectra of 4µm thick GaN films grown with four different second buffer layer thickness. The thickness of the second buffer layer grown at 800°C is (a) 0Å, (b) 400Å, (c) 800Å, (f) 1600Å. The spectra were taken at 87K with acceleration voltage of 15KeV

Fig. 4 shows the electron Hall mobility as a function of the thickness of the 800°C buffer layer. The mobility increases as the thickness of the 800°C layer increases to 400Å, reaches a maximum and decreases as the thickness

increases from 400Å to 1600Å. The absolute values of the mobility for these samples are low because we selected relatively high growth rate conditions for growth of the thick film. However by growing at 950°C with a 400Å thick 800°C buffer layer, we obtained an electron mobilities as high as 531 cm²/V sec.

From the results of SEM, CL and Hall measurements, it is clear that 400Å is the optimum thickness for the 800°C buffer layer to achieve smooth surface morphology and good crystallinity. If no 800°C buffer layer is used, the desorption of the 450°C buffer

layer affects the initial nucleation of TTIs and prevents their coalescence. As a result, defects or grain boundaries are generated between the TTIs and these defects or boundaries lower the mobility and decrease the CL band edge emission. On the other hand, as the thickness of 800°C layer increases, the lateral growth rate of the TTIs increases. Because of this increased lateral growth rate misoriented TTIs become entrapped in the film after coalescence. As a result, the crystallinity of the films after TTI coalescence is reduced which affects the carrier mobility and the intensity of band edge emission. Further study is underway to verify this hypothesis. In summary, we show that a multi-step buffer layer is effective to avoid thermal desorption and/or mass transport of lower temperature buffer layers, to enhance lateral growth of TTIs and to generate TTIs which have uniform height and uniformly cover the buffer layers. The lateral growth rate of TTIs can be controlled by the thickness of the buffer layer grown at higher temperature. There is an optimum lateral growth rate to achieve the maximum mobility, smooth surface morphology and good crystallinity.

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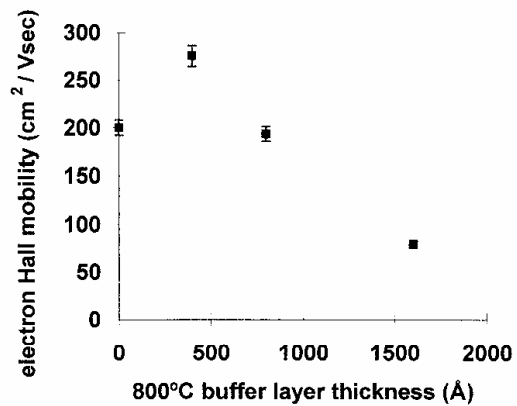


Fig.4. Electron Hall mobility as a function of 800°C buffer layer thickness

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