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THE EFFECT OF HYDROGEN CARRIER GAS ON THE
MORPHOLOGICAL EVOLUTION AND MATERIAL PROPERTIES OF
GaN ON SAPPHIRE

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ABSTRACT

In-situ optical reflectance is used to monitor the morphological evolution of the two-step GaN growth on sapphire. The amount of H₂ carrier gas used in the growth is observed to strongly influence the morphological evolution of the low temperature buffer layer and the subsequent high temperature nucleation behavior, which in turn affects the structural and electrical properties of the GaN epitaxial films. The optical reflectance transients correlate with the sizes and distributions of nuclei as observed by AFM.

INTRODUCTION

Despite the large lattice mismatch between sapphire and GaN, device quality GaN/AlGaIn/InGaIn has been made possible largely through the two-step nucleation procedure as proposed by Amano et al. in 1986 [1]. Briefly the procedure calls for the growth of a low temperature (LT) AlN buffer layer on the sapphire at 600°C, followed by a temperature ramp to above 1000°C for the high temperature (HT) nucleation of GaN on the AlN buffer layer. The buffer layer reportedly increases the nucleation site density and promotes the lateral growth of GaN at high temperature [2]. A model was proposed whereby the GaN nucleates on the LT buffer layer which consists of high density of columnar fine crystals, and then undergoes a geometric selection where the nuclei which grow along the c-axis grow the fastest and emerge as islands. These islands then coalesce and form a continuous surface with quasi-2-dimensional uniform growth [3]. The two-step nucleation procedure has enabled the growth of GaN with smooth morphology and far superior electrical and optical properties than GaN directly nucleated on sapphire. Subsequently Nakamura et al. also proposed the use of low temperature GaN buffer layer which is reported to be functionally identical to the AlN buffer layer [4]. Using a variety of microscopy techniques, subsequent studies on the growth mechanism revealed an evolution of the LT buffer layer starting from its deposition through the temperature ramp and anneal. These studies showed that the degree of crystallinity, the average grain size and root mean square (RMS) roughness of the buffer layer increased at the end of the process [5, 6, 7].

While such ex-situ studies revealed detailed but static information about the crystal structures and surface morphology of the growing film, an in-situ diagnostic capability in an MOCVD reactor will provide a more dynamic picture to the growth evolutionary processes. Using a pyrometer monitoring the infrared (IR) transmission through the sapphire substrate and GaN epitaxial film, Nakamura et al. was able to correlate the pyrometric output with the surface roughness of the GaN [8]. The observed decay and recovery of the IR intensity was supportive of the above mentioned model of growth evolution. Here, a real-time in-situ optical reflectance monitoring set-up has been implemented which has allowed us to make the same observations, as well as to dynamically explore the effects of various growth parameters. In addition to the parameters which have been reported

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Figure 1: Reflectance (—) and temperature (---) traces of a typical two-step growth of GaN on sapphire.

in the literature, namely the buffer layer thickness and growth temperature, V/III ratio, nitridization and etc, we have found that the concentration of hydrogen in the gas composition exerts a pronounced effect which has not been previously addressed.

EXPERIMENT

Growth of GaN is conducted in an inductively heated rotating disk reactor (RDR) with ammonia (NH_3) and trimethylgallium (TMGa) as precursors. Hydrogen (H_2) is used as the carrier gas as well as the make-up flow to supplement the total flow rate required to maintain a matched laminar flow pattern in an RDR. Temperature is monitored by an IR pyrometer calibrated by the melting of Ge at 934°C . *C*-plane (0001) sapphire substrates are degreased and thermally treated in-situ in H_2 at 1100°C for 10 minutes before the LT GaN buffer deposition. The temperatures for the buffer deposition and growth are kept fixed at 560°C and 1075°C respectively for this series of study. The susceptor is rotated at 1200 rpm. The rotating speed in combination with the reactor pressure and susceptor temperature determine the total gas flow rate required to maintain a matched flow pattern [9]. Flow rates of TMGa and NH_3 are kept at $130\mu\text{mol}/\text{min}$ and 6.0slm respectively to give a high temperature growth rate of approximately $2.3\mu\text{m}/\text{hr}$.

A normal incidence reflectometer ($\lambda = 550.5\text{nm}$) has been implemented using a tungsten lamp as a white light source. A lock-in amplifier is used to suppress background signals from radiation of the heated susceptor. Hall measurements are performed using Ti/Al metal contacts. (0002) symmetric X-ray diffraction (XRD) rocking curves are used to assess structural quality. A Digital Instrument Nanoprobe III atomic force microscope (AFM) operating in the tapping mode is used to study the surface morphology.

RESULTS AND DISCUSSIONS

A typical trace of reflectance intensity (solid line) in relation to growth temperature (dashed line) is shown in Figure 1. Distinct stages of the two-step GaN growth on sapphire, as highlighted by the alternating white and gray background, can be identified. Starting

with a sapphire surface pretreated in H_2 , the growth proceeds as followed: (i) LT buffer deposition (reflectance rises due to increase in refractive index); (ii) temperature ramp and the accompanying morphological evolution; (iii) HT nucleation, lateral growth, and further surface roughening; (iv) island coalescence, as indicated by the recovery of reflectance intensity; and (v) quasi 2D growth, with sustained reflectance oscillations due to increase of thickness. With the in-situ reflectance monitor providing realtime feedback on the growth evolution, the effect of various growth parameters on different stages of the growth can be investigated. Of particular interest are the effect on the extent of the initial HT roughening (stage iii), and the rate of subsequent recovery (stage iv), since these two stages are seemingly more sensitive to changes in the growth conditions and are susceptible to a wide range of variations. Qualitatively, we have observed that a higher HT nucleation temperature and a higher NH_3 flow (relative to a fixed TMGa flow rate) promote lateral growth, therefore reducing the amount of initial roughening and enhancing the recovery rate. A thinner buffer layer, down to about 150Å, generally leads to a similar effect. We note that similar observations have been reported in the literature based on both in-situ and ex-situ studies [10, 11, 12].

In addition to the above mentioned parameter, we have also investigated the effect of H_2 in the gas composition on the evolution of the morphology during nucleation. It has been reported that H_2 carrier gas stabilizes the (0001) GaN surface and suppresses lateral growth, but its exact role during nucleation has not been clarified [13]. In our experiment, to exclude the effect of the NH_3 from that of the H_2 , the partial pressure of NH_3 was kept approximately constant while the partial pressure of H_2 was varied. The total reactor pressure thus varied accordingly. Computer modeling showed the fluid flow to be similar in all cases. Nominally all other growth condition parameters were maintained invariant. Figure 2 shows the reflectance traces from three representative growths of GaN on sapphire in the studies. The growth conditions are summarized in Table I. Among the three samples, sample B represents our baseline conditions and its reflectance transient (Figure 2b) can be regarded as a reference trace with all the distinct stages of growth evolution readily described by the two-step process, while samples A and C respectively represent the cases with less and more H_2 . By reducing the H_2 flow and partial pressure of sample A to 5.1slm and 38Torr respectively, Figure 2a shows that the initial roughening is completely removed and the mean reflectance quickly attains a consistent level. In contrast, increasing the H_2 flow and partial pressure of sample C to 9.6slm and 105Torr respectively, results in prolonged initial roughening and more gradual recovery (Figure 2c). All three samples showed similar specular surface morphology with well defined step edges under AFM imaging at the end of approximately 2μm deposition. To ascertain that the above observations are not an indirect consequence of the different total reactor pressures employed in the growths, a control experiment growth was performed with the growth conditions of sample B, but with most of the H_2 replaced by N_2 to supplement for the partial pressure. The result demonstrates nearly identical reflectance transient to that of sample A.

Table I summarizes the structural and electrical properties of the above three samples. With little or no roughening and consistently strong reflectance intensity, sample A shows the broadest (0002) X-ray diffraction (XRD) linewidth, and the lowest electron mobility. The broad XRD linewidth indicates the presence of high density of dislocations with a screw component, as confirmed by cross section TEM to be in the region of mid to high $10^9 cm^{-2}$. Sample C which undergoes prolonged roughening has the narrowest XRD linewidth, but

Figure 2: Reflectance traces from three GaN growths with varying H_2 flow rates and partial pressures.

Table I: Selected growth parameters and characterization results of Samples A–C.

Sample	H_2 flow rate	Pressure (H_2)	Pressure (total)	μ ($\text{cm}^2/\text{V} - \text{sec}$)	n (cm^{-3})	X-ray FWHM
A	5.1 slm	38 Torr	100 Torr	227	1.34×10^{17}	350"
B	7.6 slm	78 Torr	140 Torr	512	1.42×10^{17}	300"
C	9.6 slm	105 Torr	170 Torr	435	2.57×10^{17}	290"

Figure 3: AFM images of the morphology of nucleation layers similar to those of (a) Sample A and (b) Sample B.

lower mobility and higher background carrier concentration than sample B. Despite better structural quality, the electrical properties of sample C may have been compromised by relatively insufficient amount of NH_3 in the gas composition.

To gain further insight into the underlying mechanism through which the H_2 manifests its effect on the growth evolution, we prepared two LT GaN buffer layers under identical growth conditions to those of samples A and B respectively, but interrupted right before the HT nucleation for AFM studies. Figure 3 shows the two $10\mu\text{m} \times 10\mu\text{m}$ AFM images of the samples surface morphology. It is clearly seen that the higher amount of H_2 in the gas composition of sample B results in increased grain size to between 1.5 to $2\mu\text{m}$, as compared to approximately $0.8\mu\text{m}$ in sample A. The RMS roughness of sample B also increases accordingly, from $45\text{\AA}$ to $90\text{\AA}$, based on the $10\mu\text{m} \times 10\mu\text{m}$ area.

Based on a combination of these in-situ and ex-situ characterizations, a modification to the growth model of GaN on sapphire can be inferred which enables one to make finer distinction between nucleation conditions which produce GaN of better or poorer quality. It is helpful to think of the deposition of the LT GaN buffer layer and the subsequent temperature ramp/anneal as a process of preparing a nucleation template. This nucleation template determines the nuclei density during high temperature nucleation. In the initial stage of nucleation, twisting and tilting could conceivably occur among the nuclei in order to accommodate the strain energy, which results in the formation of low-angle twist and tilt boundaries during and after island coalescence. With smaller grain sizes and higher nuclei density as in sample A, it is expected to take less time for the island coalescence to occur and the overall surface will never reach a high degree of roughness. But the higher density of incidence of coalescence also leads to more structural defects at the coalescence boundaries. In contrast, larger grain sizes and lower nuclei density as in samples B and C allow more room for individual islands to develop before they start to coalesce. Larger intermediate islands result in rougher surface morphology, but eventually the increased average volume of defect-free columnar domains and lower density of coalescence boundaries, leads to improved structural and electrical properties.

CONCLUSIONS

We have demonstrated that, in addition to the growth parameters which have been reported in the literature, the amount of H₂ carrier gas used in the MOCVD growth of GaN on sapphire can profoundly affect the morphology of the LT buffer layer. This in turn strongly influences the subsequent morphological evolution of the growth and the resulting material quality. A combination of in-situ reflectance and ex-situ electrical and structural characterizations suggest that a finite roughening and incubation period during the initial HT GaN growth, as a consequence of larger grain sizes and lower nuclei density, is necessary to achieve better GaN material quality.

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