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Investigations of *in situ* reflectance of GaN layers grown by MOVPE on GaAs (001)

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ABSTRACT

The growth of low temperature GaN (LT-GaN) layers on GaAs (001) substrate was performed by metal organic vapor phase epitaxy (MOVPE) at growth temperature range of 500–800 °C. Laser reflectometry (LR) was employed for *in situ* monitoring of all growth steps. The simulation of experimental time reflectance traces shows that at the first growth stage, the surface roughness increases to reach a limit value depending on growth temperature. Due to surface roughness profile the growth rate time-dependence was found non negligible at the first growth stage. The investigations of *in situ* reflectance give more precise measurement of growth rates that yields to thermal activation energy close to 0.12 eV. The *ex-situ* analyses by spectral reflectance (SR) and Atomic Force Microscopy (AFM) showed that the better surface morphology was obtained when the GaN buffer layer is grown at lower temperature, while three dimensional (3D) growth mode was observed at higher temperature. A series of high temperature (800 °C) GaN (HT-GaN) layers were grown on different thicknesses of low temperature (550 °C) GaN buffer layer. The results showed that high density of nucleation sites enhances the initial growth rate and improves the morphological quality of GaN active layer.

1. Introduction

Since many decades, GaN material has found wide application in optoelectronic devices such as: UV detectors, LEDs, and LASER diodes [1–4]. However, the development of high-efficient GaN has been hampered by the absence of perfect and low cost substrates materials. In spite of the progress made in growth of GaN on sapphire [1–7], this substrate is not suitable for device processes such as cleaving, etching and forming electrodes. These objectives still the principal cause of several attempts using others substrates. Different substrates have been used for epitaxial growth of GaN such as Si [8–12], SiC [13–15], GaAs [16–18] and even GaN substrates [19–21]. In particular, GaAs substrate still has a low cost, a large wafer size, a high thermal conductivity, and an associated advanced technology [22,23]. The advantages of GaAs as a substrate for GaN epitaxy include its isoelectronic structure (i.e. both GaAs and GaN are III-V compounds), the shared element (Ga), the potential to convert the surface of GaAs to GaN, and parallel cleavage planes between the film and the substrate [24].The main goal

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to study the GaN epitaxy on GaAs substrate is to obtain pure zinc blende GaN [24–26]. In order to achieve smooth and flat GaN surface and GaN/GaAs interface, several growth processes must be adequately controlled [27]. These processes include prevention of GaAs decomposition and minimizing of dislocations density that can lead to improvement of GaN crystal quality [28,29]. It is well known that, due to As element loss, uncapped GaAs decomposes when it is heated at temperature above 600 °C. However, GaAs surface can remain stable, under AsH_3 flow. Nevertheless, due to its easy etching under NH_3, GaAs surface was found unavoidable at higher temperature [30]. This factor set a limit to the maximum growth temperature while high temperatures are more suitable for GaN growth. To overcome this limitation, a low temperature GaN buffer layer (LT-GaN) may be envisaged to protect GaAs surface during sublayer growth at relatively higher growth temperatures (>600 °C) [31–33]. The early stages of LT-GaN, especially islands nucleation control, are of crucial importance to achieve good quality of GaN sublayer. There are few results about investigation of wetting difficulty between GaN and GaAs. In this background, it is very useful to more study the first stages of low temperature GaN nucleation versus growth conditions. Generally, nucleation layer quality essentially depends on its thickness and growth temperature [34–36]. Sun et al. [36] reported that more slowly growth by metal organic vapor phase epitaxy (MOVPE) of GaN on GaAs (001) at higher temperature (850 °C) induces more stacking faults. Further, the GaN crystallization depends on diffusion lengths of adatoms (Ga and N) that can be controlled by growth temperature [37]. Thus, high quality of GaN layer requires a precise control and a perfect mastery of growth parameters. For this purpose, optical techniques such as: spectroscopic ellipsometry (SE) [38], laser reflectometry (LR) [39] and spectral reflectance (SR) [40] have received considerable attention for *in situ* monitoring of epitaxial growth. Their potential as a nondestructive, real-time monitoring tool present a considerable support to be used during the growth process. Among these tools, lasers reflectometry is the most successfully used in MOVPE, due to its easy installation [41,42]. This technique gives information's on both growth rate and surface roughness during growth [43–46].

In this work, based on investigation of *in situ* reflectance recorded during growth of low temperature GaN layers on GaAs (001) substrate, we study the kinetic growth at the first stage of GaN nucleation on GaAs (001) substrate. For this reason, we used a quantitative optical model that includes both a time-dependent growth rate and time-dependent surface roughness profiles, to simulate several *in situ* reflectance traces of GaN layers grown on GaAs (001) substrate at low growth temperature. The output simulations were correlated with *in situ* reflectance measurements and with *ex situ* analysis performed by atomic force microscopy (AFM) and by UV–Visible spectral reflectance (SR).

2. Experimental details

GaN layers were grown on semi-insulator GaAs (001) substrates by atmospheric pressure MOVPE. The heated reaction zone is obtained through induction heating and is monitored by S-type thermocouple and feedback system. Trimethylgallium (TMG: 8 μmol/min at low temperature and 40 μmol/min at high temperature) and (NH_3: 2slm) were used as precursors of Ga and N, respectively. H_2 and N_2 were used as carrier gas for the growths of buffer layer and sublayer, respectively. A first series of samples labeled from A to D were directly grown on GaAs at temperature varied in the range of 500–600 °C. These LT-GaN layers have almost the same thickness at around 0.5 μm. In order to study the buffer layer thickness effect on high temperature GaN sublayer, a second series of HT-GaN layers labeled from E to J was performed at 800 °C where the buffer layer thickness was varied in the range of 38–260 nm. Firstly, GaAs substrate surface was exposed to NH_3 flow during 10 min at temperature of 550 °C. After this nitridation phase, the low temperature GaN buffer layer was grown. Then, the temperature was set to 800 °C to grow GaN sublayer. All growth steps were *in situ* monitored by single wavelength (λ = 632.8 nm) He–Ne laser reflectometry operated at normal incidence. In addition to simulation results for thickness measurement we also used SR as *ex situ* tools. AFM analyses were carried out with a Nanosurf easyScan 2 Flex system in the dynamic force mode, with cantilever's resonance frequency of about 165 kHz. Commercially available tips (ACLA silicon AFM probes from AppNANO) with curvature radius lower than 6 nm were used.

3. Results and discussion

3.1. LT-GaN growth

In order to study the growth temperature effect, a series of GaN layers were directly grown on (001) GaAs substrates. Fig. 1 shows the plots of experimental time reflectance's during the growth of GaN at temperature ranging from 500 to 600 °C. The time reflectance curves were normalized to those of the substrates (R_s). As seen in Fig. 1, the reflectance curve of sample A that grown at lower temperature (500 °C) is characterized by regular oscillations with constant amplitude. When the temperature is increased to 550 °C (sample B), the reflectance signal drops and the maximum amplitude remains constant at a level close to 80% of substrate reflectance. This behavior was associated to surface degradation at the first stage of nucleation layer that leads to an important surface roughness causing light scattering. However, the reached maximum of reflectance keeps a constant value until the growth end. In addition, the amplitude oscillations decrease with the growth temperature increase. The extreme case was obtained for 600 °C (sample D) for which the absolute maximum reached by the reflectance amplitude from a critical thickness around 130 nm does not exceed 10% of R_s value. On one hand, this indicates that the surface damage originating from wetting between GaN and GaAs, take place in the first nucleation stage and on the other hand, the surface roughness reaches a limit value, which still independent on growing thickness.