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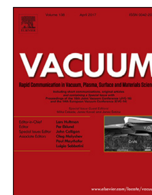

Study of cubic GaN clusters in hexagonal GaN layers and their dependence with the growth temperature

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ABSTRACT

The inclusion of cubic phase in MOVPE-grown hexagonal GaN on GaAs substrate and its dependence with the growth temperature are investigated by high-resolution X-ray diffraction (HR-XRD), scanning electron microscopy (SEM), atomic force microscopy (AFM) and cathodoluminescence (CL). It is observed that the GaN layers surface exhibits 3D-grains structure. The density and shape of these grains are largely dependent on the growth temperature. HR-XRD study reveals the presence of cubic GaN clusters in the hexagonal GaN layer. Using CL we show that the cubic inclusions are not localized at the substrate/epilayer interface but propagate throughout the film.

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1. Introduction

GaN wide band gap semiconductor and its alloys are useful materials for the fabrication of devices operating in blue and ultraviolet spectral regions [1,2]. The most stable crystalline structure of GaN is the hexagonal structure (h-GaN), thus devices including light emitting diodes are based on h-GaN. High-quality h-GaN films and heterostructures are usually grown either by metal organic chemical vapor deposition (MOVPE) or by molecular beam epitaxy (MBE) on sapphire (Al_2O_3) and 6H-SiC substrates [3,4]. However, h-GaN layer shows spontaneous and piezoelectric-induced polarization [5], which is generally a drawback for devices [6]. c-GaN results in more isotropic physical properties without detrimental polarization effects. Thus, growing GaN in pure cubic structure (c-GaN) or mixed cubic-hexagonal structure, as reported by Saengkaew et al. [7], is promising to achieve non-polar or semi-polar GaN layers. In addition, c-GaN layers offer easy cleavage along (001) facet orientation, superior electronic proprieties, and high doping efficiencies [8–10] as compared to h-GaN layers.

It has been reported that c-GaN layers can be grown on different substrates such as (001) GaAs [11], (001) Si [12] and 3C-SiC [13], but the most commonly employed substrate for this is GaAs (001). GaAs substrates are preferred because of their advantage for device fabrication due to easy cleaving, etching, and ohmic contact realization. Nevertheless, issues such as large lattice mismatch between the GaAs substrate and GaN layer, small difference between the formation energy of the two phases (hexagonal and cubic), and high decomposition rate of GaAs substrate in ammonia (NH_3) at temperature above 700 °C lead to high structural quality c-GaN layers growth challenging [14–17]. To improve the growth of c-GaN on GaAs substrate it is necessary to go further in the understanding of the link between the formation of the cubic phase and the deposition conditions. In a previous study, we have shown that a significant improvement of the GaN layer structural quality can be achieved using substrate nitridation and low temperature buffer layer deposition before the growth of the epitaxial GaN layer at higher temperature. These steps prevent the decomposition of the GaAs substrate surface and enhance the GaN nucleation to initiate the growth of the GaN layer [18]. We also have shown, with other authors [19–21], that to obtain and stabilize the cubic phase, the deposition conditions (V/III ratio, growth temperature, GaAs substrate orientation) have to be optimized. In this paper, we focus on the growth and characterization of GaN layers on GaAs (001) substrate and we study, using in-situ reflectivity measurements, scanning electron microscopy (SEM), atomic force microscopy (AFM), high-resolution X-ray diffraction (HR-XRD) and

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cathodoluminescence (CL), the formation of cubic inclusions in the hexagonal GaN layer as function of the growth temperature in the temperature range of 700–850 °C.

2. Experimental details

GaN layers were grown by atmospheric pressure metalorganic vapor phase epitaxy (AP-MOVPE) on (001) GaAs substrate. Ammonia (NH_3) and Trimethylgallium (TMG) were used as a precursor of nitrogen (N) and gallium (Ga), respectively. H_2 was used as a carrier gas with a total flow rate of 2 slm (standard liter per minute). The growth process starts by nitridation of GaAs substrate under NH_3. This step is followed by the growth of a GaN buffer layer at the same growth temperature used during the nitridation process. Four different GaN layers were then grown at different growth temperatures on top of the buffer layer. The different samples are labeled $^{\#}S_1$, $^{\#}S_2$, $^{\#}S_3$ and $^{\#}S_4$ and correspond to growth temperature of 700, 750, 800 and 850 °C, respectively. Details of the growth process of the samples can be found elsewhere [21]. The growth was in-situ monitored by single wavelength ($\lambda = 632.8$ nm) He-Ne laser reflectometry operated at normal incidence. The surface morphology is characterized by scanning electron microscopy (SEM) and atomic force microscopy (AFM) at a scale of $5 \times 5\ \mu m^2$. X-ray diffraction measurements were performed using a high-resolution diffractometer equipped with a fourfold Ge (2 2 0) monochromator, delivering a pure CuKα_1 line of wavelength ($\lambda = 0.154$ nm). Room temperature depth-resolved CL investigations are performed in a digital scanning electron microscope. Emitted light is detected via a parabolic mirror collector and analyzed by a spectrometer with a focal length of 320 mm using a 1200 grooves mm^{-1} grating and a spectral resolution of 0.06 nm. The signal is then recorded by a liquid N_2-cooled Horiba Jobin Yvon Instruments Symphony 1024 × 256 CCD detector.

The depth profile of residual impurities along the growth direction was obtained by secondary ion mass spectroscopy SIMS. The calibration of elemental concentration for this instrument was done by using relative sensitivity factors (RSFs) derived from the analysis of a GaN standard with known doses of impurity implants. SIMS measurements of negative and positive secondary ions were performed with the ultra-low energy conditions of 3 keV primary ion bombardment impact energy by Cesium and Oxygen respectively.

3. Results and discussion

Fig. 1 shows the reflectance curves recorded as function of the time during the growth of the GaN layers on GaAs (001) substrate for temperature varying between 700 °C and 850 °C. Four steps can be distinguished: (i) nitridation of GaAs substrate at 550 °C, (ii) low temperature (550 °C) growth of 65 nm thick GaN buffer layer, (iii) temperature ramp up, (iv) GaN layer deposition in the temperature range of 700–850 °C. For all samples, as expected, the reflectance signal corresponding to steps (i-iii) does not exhibit any difference but differs in step (iv). In this step, sample $^{\#}S_2$, for which the GaN layer is grown at 750 °C, is characterized by regular oscillations of the reflectance signal with a nearly constant (or slightly decreasing) amplitude. In contrary, for the other samples the amplitude of the oscillations in the reflectance signal clearly decreases with time. The largest oscillation damping is observed in sample $^{\#}S_4$ for which the GaN layer is grown at the highest temperature (850 °C). The damping of the oscillation in the reflectance signal is attributed to changes in the surface morphology of GaN films and can be estimated by calculating the damping rate r_d using the following equation [18]:

$$r_d = \text{Log}\left(\frac{R_{max(i-1)}}{R_{max(i)}}\right) \quad (1)$$

where (i) is the order of the reflectance maximum (R_{max}). The inset

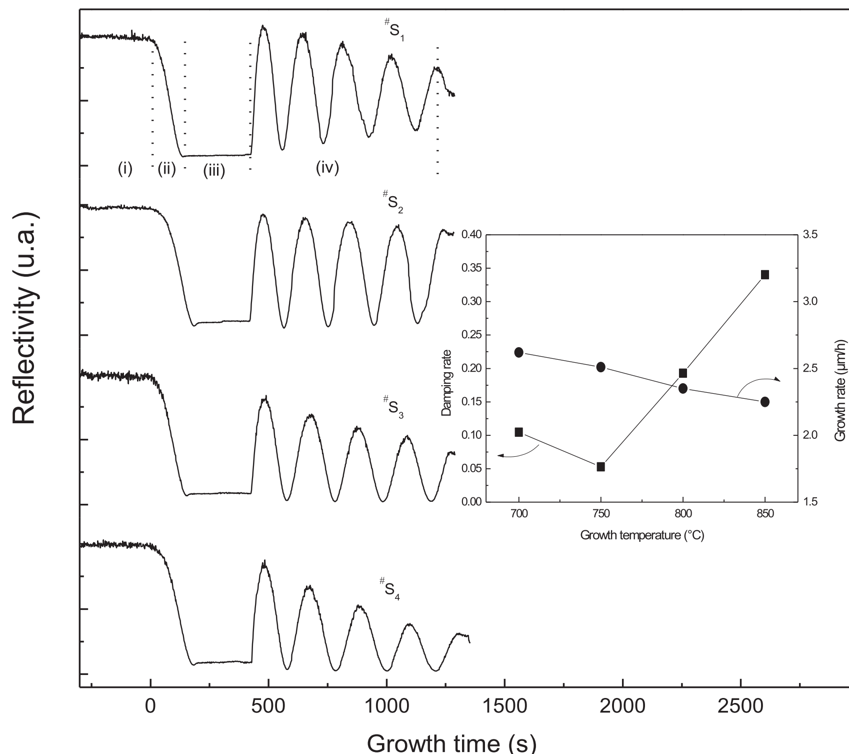

Fig. 1. In-situ real time laser reflectometry recorded during the growth of GaN/GaAs (001) samples. From run to run, the growth temperature of subsequent GaN layers was varied from 700 °C ($^{\#}S_1$) to 850 °C ($^{\#}S_4$). Inset shows the plot of damping rate and growth rate versus growth temperature.

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