Control of stress and threading dislocation density in the thick GaN/AlN buffer layers grown on Si (111) substrates by low-temperature MBE

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Abstract. We report on successful growth by plasma-assisted molecular beam epitaxy on a Si(111) substrate crack-free GaN/AlN buffer layers with a thickness more than 1 μm. The layers fabricated at relatively low growth temperature of 780°C have at room temperature the residual compressive stress of -97 MPa. Intrinsic stress evolution during the GaN growth was monitored in situ with a multi-beam optical system. Strong dependence of a stress relaxation ratio in the growing layer vs growth temperature was observed. The best-quality crack-free layers with TDs density of ~10^9 cm^-2 and roughly zero bowing were obtained in the sample with sharp 2D-GaN/2D-AlN interface.

1. Introduction

III-Nitrides on Si(111) templates are of great importance for fabrication of high electron mobility transistors (HEMT) [1] and power electronics due to low cost, large available sizes of silicon substrates, and the possibility of integration with the existing Si technologies. However, heteroepitaxy of III-Nitrides on Si(111) suffers from large lattice and thermal expansion coefficients (CTE) mismatches (17% and 54% for GaN on Si(111), respectively [2]) that leads to tensile strain generation during growth and cooling to room temperature (RT) of heterostructures. Conventionally, III-N layers grown on Si substrates by vapour phase epitaxy (VPE) [3] and metal-organic chemical vapour deposition (MOCVD) [4] have better structural quality than layers grown by molecular-beam epitaxy (MBE) [5]. Despite the lower TDs density obtained by MOVPE, the performance of optimized devices grown by MBE is not inferior to that of devices grown by MOVPE [6]. Moreover, VPE and MOCVD require a higher temperature for epitaxial growth of III-N layers than PA MBE. Hence, much more tensile stress generation is observed in samples grown by VPE and MOCVD during cooling down to RT. This tensile stress leads to film cracking [7] and bowing of the heterostructure, which can destroy the device performance or significantly complicate the post growth processing. The solutions for this problem can be reducing the growth temperature and using less lattice-mismatched (~2.4%) [8] AlN buffers. The interface between AlN and growing thick GaN layer is an effective source of compressive stress. Therefore, low growth temperature and in situ control of the stress level could make PA MBE suitable for growth of compressively strained GaN/AlN/Si(111) templates for further device fabrication.
The paper reports on fabrication of compressively strained at room temperature GaN/AlN buffer layers on Si(111) substrates by PA MBE. In addition, in situ analysis of stress evolution and morphology in the growing heterostructure has been presented, which has been done by home-made, multi-beam optical system (MOS), and Reflection High-Energy Electron Diffraction (RHEED).

2. Experimental

The Si substrates with thickness of 275 μm were annealed at 820°C for an hour until the 7×7 surface reconstruction was observed by RHEED. Before AlN growth, 6 MLs of Al were deposited to prevent Si-N interaction. Then, 30-nm-thick AlN:Ga/Si(111) nucleation layer (NL) was grown by migration enhanced epitaxy (MEE) at Al-rich conditions with $F_{Al}/F_{N}=1.02$ [9], while substrate temperature simultaneously was increased from $T_{S}=700°C$ to $T_{S}=780°C$, followed by an intermediate 170-nm-thick 2D-AlN buffer layer grown by metal-modulated epitaxy (MME) [10] technique at the flux ratio $F_{Al}/F_{N}=1.6$ and the $T_{S}=780°C$. Both NL and intermediate AlN layers were grown with the additional Ga-surfactant flux $F_{Ga}=0.3$ ML/s. The top 1-μm-thick GaN layer was grown at the Ga-rich conditions with the flux ratio $F_{Ga}/F_{N}=1.9$ and lower $T_{S}=700°C$.

RHEED and laser reflectometry were used for in situ control of the surface morphology and growth rate of the layers. In addition, a home-made multi-beam optical system (MOS) was installed at the standard ellipsometry ports of the setup to measure in situ substrate curvature. The surface morphology was investigated ex situ by an atomic force microscope (AFM). Crystalline quality of the heterostructures was evaluated by high-angle annular dark-field scanning TEM (HAADF STEM) (FEI Titan 80-300 keV), and X-ray diffraction (XRD) analysis of symmetric (0001) and skew-symmetric (10-15) reflexes. The latter were used to evaluate densities of TDs with Burgers vectors $b=c$ or $b=a+c$ (N_screw) and $b=a$ or $b=a+c$ (N_edge) in approximation of random TDs distribution [11].

2.1. Stress – thickness versus thickness plots

Film stresses can be evaluated by measuring the change in curvature of substrate during stressed film growth by using the modified Stoney equation [12]

$$\langle \sigma_f \rangle_{h_f} = \frac{M_f h_f^3}{6} \int_0^{h_f} \sigma(z) \, dz = \frac{M_f h_f^3}{6} \Delta \kappa,$$

where subscripts $f$ and $s$ refer to the film and substrate, respectively, $<\sigma_f>$ is the mean film stress, $\sigma(z)$ is the stress at particular thickness $z$, $\kappa$ is the curvature, $M_f$ is the substrate biaxial modulus. Thus, by measuring in situ the change in heterostructure curvature, stress-thickness product $<\sigma_f>_{h_f}$ can be determined in real time and converted to film stress if the film thickness is known. For film grown at constant stress, the plot of stress-thickness vs thickness is linear dependence with a slope determining the film stress. For films with varying stress with thickness, stress-thickness vs thickness plot is a non-linear function, where the incremental slope $d(<\sigma_f>_{h_f})/dh_f$ corresponds to microstructure changes in the layer.

3. Results

3.1. Film characteristics

HAADF STEM images at different magnifications (Fig. 1) illustrate rather planar morphology for all the interfaces in this heterostructure. One can observe also inclination of the TDs above the GaN/AlN interface, followed by their fusion or annihilation during propagation toward the film surface.
In addition, HAADF STEM revealed a flat interface between the MEE AlN NL and Si(111) substrate without pronounced interaction between Al and Si during first MEE cycles, contrary to step-like interface observed by D. Litvinov et. al. [13] in AlN/Si(111) layers grown by PA MBE without special NLs.

The AFM image (figure 2. left) of the GaN(1μm)/AlN(170nm)/NL-AlN:Ga(30nm)/Si(111) template showed relatively smooth surface morphology with RMS roughness of 1.7 nm and rather large grain sizes about 2 μm in diameter. The grain boundaries are clearly visible indicating incomplete coalescence, but peak-to-valley height of 14 nm with respect to heterostructure thickness of 1.2 μm corresponds to multi-step coalescence process.

The image of GaN surfaces obtained by optical microscopy (Fig 2. right) revealed crack-free surface of the sample, corresponding to absence of the high level of tensile strain at RT.

XRD θ-2θ scans of the template demonstrated full widths at half maximum (FWHM) of 782 and 1182 for symmetric (0002) and skew-symmetric (10- Ḹ5) reflexes, respectively, that correspond to screw and edge TD densities of \( N_{\text{screw}} = 1.2 \times 10^9 \text{ cm}^{-2} \) and \( N_{\text{edge}} = 7.4 \times 10^9 \text{ cm}^{-2} \).

3.2. Intrinsic stress evolution
The stress-thickness versus thickness and stress versus thickness curves for GaN(1 μm)/AlN(200 nm)/Si(111) heterostructure are plotted in figure 3. The stress versus thickness curve that characterize incremental stress in the layer was obtained from polynomial fit and further differentiation, \( d(<\sigma_1>h_j)/dh_j \), of stress-thickness vs thickness curve. As seen in figure 3, incremental stress during the growth of AlN layer was tensile and varied from 0.5 to 0.8 GPa. The stress behavior in GaN demonstrates continuous gradual relaxation from the highest observed compressive stress of -3.9 GPa to -0.3 GPa.
4. Discussion

Theoretically, the stress arising during growth of lattice-mismatched layers should be constant and can be evaluated as:

\[ \sigma_f = M_f \varepsilon, \]  

where \( M_f \) is the biaxial modulus of growing layer and \( \varepsilon \) is the strain. Using \( M_f = 470 \) GPa for AlN [14] and lattice mismatch between Si(111) and AlN \( \varepsilon = 0.19 \) [8], the tensile stress in AlN layer can be evaluated as \( \sigma_f = 89 \) GPa. As shown in figure 3, stress arising during growth AlN layer is varying from initial value of 482 MPa to 800 MPa with the minimum value of 181 MPa and average value of 491 MPa. We speculate that instantaneous relaxation of \( \sim 99.5\% \) of tensile stress is related with generation of the misfit dislocation with an edge component and transition to Volmer-Weber (VW) growth mode [15]. Further changes in stress vs thickness behavior are possibly related with multi-step nucleation of grains, their overgrowth, and coalescence processes, which were observed in films.

Fig. 3. Stress-thickness vs. thickness (top) and stress vs. thickness (bottom) plots during GaN/AlN/Si(111) heterostructure growth. The digits shows: 1- AlN growth, 2- GaN growth, 3-cooling to RT;
grown at VW growth mode [16]. AFM image of overlaying GaN indicates VW mechanism of growth that allows assuming similar mechanism of growth for AlN buffer.

Using biaxial modulus of GaN of 478 GPa [14] and lattice mismatch between GaN and AlN of -2.4% [8], the mismatch stress arising during GaN growth on AlN can be estimated as -11.5 GPa. However, the highest stress observed during GaN on AlN buffer layer deposition is about -3.9 GPa corresponding to 66% relaxation of theoretically calculated stress. The important factor affecting compressive stress formation is the stress level in the AlN buffer layer. As seen in figure 3, at GaN/AlN interface AlN is under a tensile stress of 0.8 GPa reducing the lattice mismatch with GaN to 2.2% and decreasing the theoretical value to -10.7 GPa. Further reduction is expected from generation of the misfit dislocation with an edge component at the interface between two lattice-mismatched layers. As shown in figure 3, incremental stress in GaN layer exhibits two different ratio of relaxation. Starting first monolayer up to thickness of about 100 nm incremental stress in GaN evidences high ratio of stress relaxation, changing from -3.9 to -1.5 GPa. We assume that this stress behavior is similar to behavior observed by N. Baron et al. [17], who reported that the main source of stress relaxation in compressively strained layers is reactions between TD’s with appropriate burgers vectors. This fact can be confirmed by HAADF STEM image of heterostructure (figure1) which shows that the most intensive interaction between TD’s with their further annihilation and TD loop formation was occurred at thickness of up to about 100 nm. We suspect that further gradual stress relaxation is associated with TD’s inclination in compressively strained layers, as described in [18]. It should be noted that reactions between TD’s at thickness more than 100 nm are also occurred but less intensively due to increasing in distances between TD’s. Contrary to S. Raghavan et al. [19], who observed transition of incremental stress from compressive to tensile at thickness about 88 nm in samples grown by high-temperature MOCVD, we did not observe in the present study this transition up to thickness of 1 μm in similar heterostructures grown by low-temperature PA MBE. Tensile growth stress along with tensile stresses arising during cooling to RT may lead to film cracking. Since we have not observed the transition of the compressive stress to tensile, we believe that this difference in stress behavior during MOCVD and MBE growth is related to lower growth temperature and metal-rich growth conditions, which imposes certain kinetic barrier for TD’s propagation and reactions at early growth stages.

Experimentally measured in this study tensile stress, generated at heterostructure cooling from $T_s=700 ^\circ C$ to RT is $\sigma_{CTE}=+0.55$ GPa, has been compensated by mean compressive epitaxial stress in the film of $\sigma_r=-0.65$ GPa that results in compressively strained at RT GaN(1 μm)/AlN(230 nm)/Si(111) template.

Summarizing, by using low-temperature PA MBE at metal-rich conditions and in situ control of substrate curvature, we have obtained compressively strained GaN(1 μm)/AlN(230 nm)/Si(111) template at RT with the smooth crack-free surface with RMS roughness of 1.7 nm. Optimization of NL growth conditions allows us to prevent Al-Si and Ga-Si interaction and achieve flat interfaces between substrate and AlN layer. A comparison of in situ measurements of substrate curvature and HAADF STEM of template has confirmed the crucial influence of interplay between TD’s and stress behavior in the investigated heterostructures. In addition, a comparison between the high-temperature MOCVD and low-temperature PA MBE growth processes has revealed that the usage of the low growth temperature and the metal-rich conditions is an effective way to suppress relaxation of compressive epitaxial stress arising from the lattice-mismatched AlN/GaN interface.

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