Stress in (Al, Ga)N heterostructures grown on 6H-SiC and Si substrates by plasma-assisted molecular beam epitaxy

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Abstract. The paper describes experimental results on low temperature plasma-assisted molecular beam epitaxy of GaN/AlN heterostructures on both 6H-SiC and Si(111) substrates. We demonstrate that application of migration enhanced epitaxy and metal-modulated epitaxy for growth of AlN nucleation and buffer layers lowers the screw and edge(total)threading dislocation (TD) densities down to 1.7·10⁸ and 2·10⁹ cm⁻², respectively, in a 2.8-μm-thick GaN buffer layer grown atop of AlN/6H-SiC. The screw and total TD densities of 1.2·10⁸ and 7.4·10⁹ cm⁻², respectively, were achieved in a 1-μm-thick GaN/AlN heterostructure on Si(111). Stress generation and relaxation in GaN/AlN heterostructures were investigated by using multi-beam optical stress sensor (MOSS) to achieve zero substrate curvature at room temperature. It is demonstrated that a 1-μm-thick GaN/AlN buffer layer grown by PA MBE provides planar substrate morphology in the case of growth on Si substrates whereas 5-μm-thick GaN buffer layers have to be used to achieve the same when growing on 6H-SiC substrates.

1. Introduction

Development of high energy/frequency electronics is based largely on progress in the epitaxial technologies of wide bandgap (Al,Ga)N-based heterostructures. Among different technologies used to fabricate these heterostructures a plasma-assisted molecular beam epitaxy (PA MBE) has a set of unique features, namely, low-temperature epitaxial growth (T<800°C), precise growth control of quantum-sized heterostructures with monolayer accuracy etc.

Until now, the absence of commercially available homoepitaxial substrates causes employing different lattice-mismatched substrates. Among them, two play a crucial role in manufacturing (Al,Ga)N HEMT transistors. First, 6H-SiC substrates have a relatively small lattice mismatch (~1%) and the same coefficient of thermal expansion (TEC) as compared to AlN buffer layers. The highest thermal conductivity of SiC provides the most efficient heat dissipation in high-power devices. Despite the small crystallographic mismatch, of emerging of threading dislocations (TD) is an important issue for these substrates. In addition, GaN layers usually involved in device heterostructures have a larger TEC mismatch with the 6H-SiC substrates, which can generate a tensile residual stress resulting in cracking the device heterostructures. Finally, the high cost of these substrates still limits their wide applications. Second type of substrates is Si(111) which is attractive for mass-production of (Al,Ga)N devices due to their low cost, large size, and well-developed technology. However, these substrates have rather high crystallographic and TEC mismatches with AlN epitaxial layers (~±19% ~±13.5% respectively), that inevitably leads to the generation of strains in the heterostructures, which relax through the formation of TDs with a density up to ~10¹⁰ cm⁻². These TDs lead to degradation of carrier transport characteristics in the device structures, whereas the high level of residual stress in the heterostructures followed by their significant bending makes it difficult to
perform the post-growth device fabrication and even can result in device cracking in the case of positive (tensile) strain. Thus, the growth of thick buffer layers for reduction of TD density through their interactions and re-orientation in the basal plane as well as strain engineering to provide a small residual stress (preferably compressive) are the principal issues for the epitaxial technology of high-performance device heterostructures.

This paper describes an experimental study of the stress generation and relaxation in GaN/AlN heterostructures grown on both Si and 6H-SiC substrates by using low-temperature (<800°C) PA MBE. Special attention is paid to the development of migration enhanced epitaxy (MEE) for growth of nucleation layers with a minimum TD density and metal-modulated epitaxy (MME) for the growth of subsequent AlN and GaN buffer layers to achieve high compressive incremental stresses in the heterostructures, compensating the tensile stress arising during heterostructure cooling, and in this way to reduce the residual stress therein.

2. Experiment and estimations

Two structures A and B were grown by using a PA MBE set up Compact21T on commercial 2 inch 6H-SiC and Si(111) substrates, respectively. These structures are schematically shown in Figure 1. The same nitrogen flux of ~0.5 ML/s was used for growth of all layers in both structures. The AlN nucleation layers (NLs) in both structures were grown by MEE with alternating Al and N fluxes having an intensity ratio of 1.2, as described in [1]. The NLs with thickness of 65(30) nm were grown at substrate temperatures of $T_S=700(740)^\circ$C in samples A(B), respectively. Then, thick AlN buffer layers of 455 and 170 nm in thickness were grown by MME with the same flux ratio III/N ~1.7 in samples A and B, respectively, as described in [2]. Finally, top GaN layers with thickness of 2.8 and 1 µm in structures A and B were grown by MME and standard PA MBE at $T_S=700^\circ$C, respectively.

![Figure 1. Sketches of structures A (a) and B (b) under study.](image)

A home-made multi-beam optical stress sensor (MOSS) was used for in situ evaluation of incremental stresses $\sigma(h_l)$ in the heterostructures from the experimental data on the substrate curvature change through Stoney’s differential equation

$$\sigma(h_l) = \frac{M_S h_s^2}{6} \frac{\partial k}{\partial h_l},$$

where $M_S$ is the biaxial module of substrate, $k$ is the substrate curvature; $h_l$ and $h_s$ are the thicknesses of substrate and epilayer, respectively. The values of biaxial modules, $a$-lattice constants and TECs are represented in Table 1. Residual stress in the heterostructures $\langle \sigma_l \rangle$ was calculated through an integration of incremental stresses

$$\langle \sigma_l \rangle = \frac{1}{h_l} \int_0^{h_l} \sigma(z) \, dz.$$
Table 1. The reference data of III/N compounds and heteroepitaxial substrates.

| Compound | \(a\)-lattice constant (Å) | Thermalexpansion coefficient \((10^{-6} \text{K}^{-1})\) | Biaxialmodulus (GPa) | Latticemismatch\(^6\) toAlN (%) | Latticemismatch\(^6\) toGaN (%) |
|----------|-----------------------------|---------------------------------|----------------------|-------------------------------|-------------------------------|
| AlN      | 3.111\(^3\)                | 4.20\(^3\)                      | 470\(^4\)            | 0                             | -2.5                          |
| GaN      | 3.189\(^3\)                | 5.59\(^3\)                      | 449\(^4\)            | +2.4                          | 0                             |
| 6H-SiC   | 3.081\(^3\)                | 4.20\(^3\)                      | 602\(^4\)            | -1                            | -3.5                          |
| Si(111)  | 5.431\(^3\)                | 3.7\(^5\)                       | 229\(^5\)            | +19                           | +17                           |

The crystallographic quality of the heterostructures was evaluated by high angle annular dark field scanning Transmission Electron Microscopy (HAADF STEM) and conventional TEM as well as X-ray diffraction (XRD) analysis of symmetric (0002) and antisymmetric (10\(\bar{5}\)) reflexes.

3. Results and discussion

Figure 1 demonstrates different behaviors of the substrate curvature during PA MBE growth of heterostructures A and B. For the former, the constant compressive incremental stresses of \(-3\)\(-2.2\)GPa were measured during growth of AlN nucleation (buffer) layer, respectively, that corresponds to the stress relaxation degree as large as 36\(\pm3\)% in these layers (Figure 2a). The low degree of stress relaxation in these layers is related to a two-dimensional (2D) growth mechanism due to MEE and MME techniques used for their growth. Moreover, XRD data presented in Table 2 and TEM images (see below) indicate that the MEE technique effectively suppresses the TD occurrence in this layer. It is likely due to large size of 2D grains formed during MEE, which results in significant reduction of total length of the grain boundaries serving as preferential sites for the TD generation.

In contrast, in the case of growth of structure B the constant tensile stress of 0.2GPa was measured in AlN nucleation layer, whereas the stresses with the same sign but gradually raised from 0.2 up to 0.8 GPa were found in the AlN buffer layer (Figure 2b).

![Figure 2. Changes of the substrate curvature during PA MBE growth of structure A (on 6H-SiC) (a) and B (on Si (111)) (b).](image-url)

These results appear to be related to different lattice mismatch between AlN layers and substrates in structures A and B, as listed in Table 1. Indeed, the relatively small lattice mismatch in structure A leads to a gradual relaxation of the initial moderate compressive stress (~1%) during the entire AlN growth run, while a huge mismatch in structure B (~19%) results in the practically instant stress relaxation.
relaxation through generation of TDs in AlN layers. This is confirmed by XRD data summarized in Table 2, which indicate a much higher TD density in AlN layers grown on Si as compared to the layer grown at the same modes on SiC.

**Table 2. The results of X-ray diffraction analysis.**

| Reflex | Sample 1 | Sample 2 |
|--------|----------|----------|
|        | FWHM, arcsec | TD density, cm\(^{-2}\) | FWHM, arcsec | TD density, cm\(^{-2}\) |
| (0002) | 290 | 1.7·10\(^8\) | 780 | 1.2·10\(^9\) |
| (10\(\overline{5}\)) | 620 | 2·10\(^9\) | 1180 | 7.4·10\(^9\) |

During the next stage of growing thick GaN layers on AlN buffer the compressive stresses with maximum values of -6.1 and -3.8 GPa were observed in structures A and B, respectively. Figure 2 shows that these stresses are almost completely relaxed at the GaN thickness of about 1µm in the both structures. One should note also the generation of constant tensile stress of +0.4 GPa in sample A after the complete stress relaxation at around 2µm. This stress can be induced by different processes occurring in the growing GaN layer with the large TD density, namely TD inclination and annihilation [7].

Figure 1 indicates also a tensile stress generation during post-growth cooling in both structures, that results in moderate residual compressive stresses in sample A with a convex bending, and practically zero curvature (planar substrate) for the sample B. In addition, using the data from Table 1 and assuming the constant incremental stress, we estimate GaN thickness as large as 5 µm to achieve the planar substrate morphology in GaN/AlN heterostructures on 6H-SiC substrate typically used for HEMTs manufacturing.

**Figure 3.** TEM images of sample A with reflexes g(0002) (a) and g(01\(\overline{1}\)0) (b) as well as HAADF STEM image of sample B(c).

The observed thickness of 520 nm for highly stressed AlN layer on 6H-SiC substrate exceeds the critical thickness calculated from the Matthews and Blakeslee model [8] and corresponds to the data obtained by Okumura et al. [9] who used extra-low PA MBE growth temperatures (~650°C). Thus, our results obtained at higher temperatures of 740°C indicate a high potential of MEE technique for growing highly stressed III-N films at the moderate substrate temperatures. It should be noted also, that in the case of MOCVD of analogous heterostructures at the much high substrate temperature (~1100°C) the instant stress relaxation has been observed [10]. Furthermore, in accordance with our data the compressive stress of 6.1 GPa induced during PA MBE growth of subsequent GaN/AlN heterostructure are much higher in comparison with the stress of 1.04 GPa measured for such heterostructures grown by MOCVD [10].
4. Conclusion
The efficiency of a MEE mode for PA MBE growth of AlN nucleation layers on both 6H-SiC and Si substrates was confirmed by the complex study of GaN/AlN heterostructures by using MOSS (in situ) as well as XRD and TEM (ex situ) techniques. The TD densities of $1.7 \cdot 10^8 (2.10^9) \text{cm}^{-2}$ were measured for screw and edge dislocations in the 3µm-thick GaN/AlN heterostructure grown on 6H-SiC substrate, whereas analogous densities of $1.2 \cdot 10^8 (7.4 \cdot 10^9) \text{cm}^{-2}$ were revealed in the 1.2-µm-thick GaN/AlN heterostructure grown on Si(111) substrate at close growth parameters. Also, we demonstrate that PA MBE can provide the relatively high compressive stress in GaN/AlN heterostructures grown on both SiC and Si substrates. As a result, these stresses can completely compensate the harmful tensile stress inevitably generating during post-growth cooling of HEMT device heterostructures.

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References
[1] Nechaev D V, Aseev P A, Jmerik V N, Brunkov P N, Kuznetsova Y V, Sitnikova A A, Ratniov V V, and Ivanov S V 2013 J. CrystalGrowth 378, 319.
[2] Jmerik V N, Mizerov A M, Nechaev D V, Aseev P A, Sitnikova A A, Troshkov S I, Kop'ev P S, and Ivanov S V 2012 J. CrystalGrowth 354, 188.
[3] Ponce F A, Krusor B S, Major J S, Plano W E and Welch D F 1995 Appl. Phys. Lett. 67 410.
[4] Wright A F 1997 J. Appl. Phys. 82 2833.
[5] Ayers J E 2007Heteroepitaxy of semiconductors: theory, growth, and characterization (Storrs-CRC Press).
[6] Zhu D, Wallis D J, and Humphreys C J 2013 J. Rep. Prog. Phys. 76 106501.
[7] Romanov A E and Speck J S 2003 J. Appl. Phys. 83 2569.
[8] Matthews J W and Blakeslee A E 1974 J. Crystal Growth 27 118.
[9] Okumura H, Kimoto T, and Suda J 2012 Appl. Phys. Express 5 105502.
[10] Redwing J M and Al Balushi Z Y 2015 J. Mater. Res. 30 2900.