Stress evolution during growth of AlN templates on c-Al2O3 substrates by plasma-assisted molecular beam epitaxy

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Abstract. We describe stress evolution during plasma-assisted molecular beam epitaxy (PAMBE) of AlN nucleation and buffer layers on c-Al2O3 substrates at varying growth temperatures of 780 and 850°C. Moreover, different mechanisms of stress generation in the growing AlN films, related to the processes of grain coalescence and impact of Me-excess during PA MBE by migration enhanced epitaxy and metal-modulated epitaxy, are considered.

Introduction

Modern manufacturing of optoelectronic and electronic high power/frequency devices based on III-Nitrides widely uses c-sapphire substrates since they have excellent transparency, radiation, chemical and thermal stability combined with a quite reasonable cost. However, a high lattice mismatch between these substrates and standard AlN buffer layers (~13%) [1] leads to the generation of high compressive stresses in the latter, which are relaxed and turned to tensile stresses due to appearance of various misfit defects (misfit dislocations et.al), as well as due to forming of pronounced grain (island) morphology and development of the related coalescence process in AlN nucleation layers (NLs). As a result, the various types of vertical threading dislocations (TDs) are generated at the grain boundaries with an typical initial density more than 10^{11} cm^{-2} [2]. Previously, we have described a decrease of initial TDs density in AlN NLs grown on c-Al2O3 due to increasing of grain size in the layers grown by migration enhanced epitaxy (MEE) [3]. Then, processes of fusion and annihilation of TDs in the thick AlN buffer layers decrease in TD concentration with the highest rate at the beginning of growth and less intensively during TD propagation in more thick AlN layers. For a typical AlN buffer layer thickness of 1-2µm the density of TDs can be reduced to a range of 10^8-10^{10} cm^{-2} [4].

However, thick-AlN BLs face a problem of high tensile stresses generation, resulting in cracking film issue. The generation of tensile stresses was theoretically described by Nix and Clemens [5], who demonstrated the relation of this process with the grain coalescence and found that the stress is inversely proportional to the grain diameter. Tensile stresses in the range of several GPa can be observed in standard AlN buffer layers grown by MOCVD, which limits the thickness of these buffer layers or leads to the development of special methods to reduce these stresses using superlattices, inserting additional stress compensator layers etc.

The described above processes of TD-generation and stress evolution are actively studied by different analytical techniques such as X-ray diffraction analysis, transmission electron microscopy (TEM) etc. For in situ measurements of the stress in the growing films the most MOVPE and MBE groups use multi-beam optical stress sensors (MOSS) based on the optical (laser) measurements of the substrate curvature [6,7]. Moreover, in situ technique of reflection high electron energy diffraction (RHEED) can be used in MBE technology for direct measurements of a-lattice constant [8].

The study of stress evolution during MOCVD growth of AlN buffer layers on c-Al2O3 substrate revealed a generation of initial compressive stresses, which transformed then to tensile ones after growth of several tens of nanometers [9, 10]. In contrast, the stress evolution in the AlN/c-Al2O3 templates during their growth by low-temperature MBE is scarcely studied and the most studies were devoted to the description of stress evolution in binary heterostructures GaN/AlN and GaN/InN [8]. There is only one
study describing the stresses in AlN films grown on Si(111) substrates using PA MBE, in which complex mechanisms for the generation of both tensile and compressive stresses were demonstrated [11].

This study focuses on the stress evolution and generation of TDs during PA MBE of AlN films on c-Al₂O₃ substrates at temperatures of 780 and 850°C. Complex interplay of various mechanisms of both tensile and compressive stresses generation in AlN films, related to the numerous processes at the grain boundary are discussed.

Experiment

AlN layers were grown by PA MBE setup Compact 21T (Riber) on c-Al₂O₃ substrates, which were annealed at substrate temperature of 850°C and nitridated using plasma-activated nitrogen flux (N₂*) of 0.5 ML/s during 10 min at 780°C. Figure 1 shows design of A and B samples.

First, the 65 nm-thick AlN nucleation layers (NLs) were grown by MEE mode [3] at both moderate temperature (MT) of 780°C (A sample) and high temperature (HT) of 850°C (B sample) with the same III/N ratio ~1. The following AlN buffer layers (BLs) were grown by metal-modulated epitaxy (MME) at the same temperatures used to grow AlN NLs (see Fig.1) [12]. During the growth of BLs, the III/N ratio changed between values 1.1 and 1 as well as the growth temperature of the sample B decreased from 850 to 780°C after the growth of layer with a thickness of 840 nm, as shown in Fig.5. Upper AlN parts with a thickness of 200 nm in both samples were grown using the continuous wave (CW) mode of PA MBE with a III/N ratio of 1.1 to smooth the AlN surface.

Figure 1. Schematic views of the design of samples A (a) and B (b).

A home-made multi-beam optical stress sensor (MOSS), based on a 10 mW solid-state laser (λ=532 nm), and a standard CCD-camera, was used to measure in situ the substrate curvature at a frequency of 15 Hz to evaluate the incremental and average stresses using the Stoney equations [6]. Thickness of AlN layers was controlled in situ by laser reflectometry (LR) (λ=532 nm). The surface morphology of the AlN layers was monitored in situ using RHEED and studied ex situ using both scanning electron (SEM) and atomic-force (AFM) microscopes. The structural quality of the AlN layers was evaluated by x-ray diffraction (XRD) analysis through measuring the full width at half maximum (FWHM) of ω-scans of symmetric AlN (0002) and skew-symmetric AlN (10-15) reflections.

Results and discussion

Figures 2a,b show the spotty and streaky RHEED patterns of MT-NL-A observed at thicknesses of 20 and 65 nm, respectively, which indicate a change in surface morphology from 3D to 2D during MEE growth. AFM image of 65nm-thick MT-NL (see Fig.2c) demonstrates an island surface morphology with a lateral size of the islands of 200-300 nm and a root-mean-square (RMS) roughness of about 1.2 nm over 1×1 µm². It should be added, that the pits with a density of about 7·10⁹ cm⁻² are observed on the NL surface. In contrast, the 65nm-thick HT-NL demonstrates during its growth the unchanged RHEED patterns corresponding to transient spotty-streaky mode (see Fig. 2d, e). Figure 2f shows AFM image of this NL with a somewhat coarser surface morphology compared to MT-NL. This NL consists of island with a lateral size of ~150-250nm and a RMS of about 5 nm over 1×1 µm². However, the large value of the latter is related with a formation of the small droplets with a surface density of ~8·10⁹ cm⁻² and a diameter of a few tens of nanometers. The surface morphology beneath these droplets is quite similar to the morphology observed in MT-NL.
Figure 2. RHEED patterns of MT-NL measured at thickness of 20 (a) and 65 (b) nm, as well as its AFM image at a thickness of 65nm (c). RHEED patterns of HT-NL measured at thickness of 20 (d) and 65 (e) nm as well its AFM image at a thickness of 65nm (f).

Figure 3 demonstrates a different character of temporal evolution of both growth rates and curvature (stress × thickness) in the AlN NLs described above. The growth of MT-NL starts immediately after the nominal initiation of the growth (Fig.3a) whereas the beginning of the growth of HT-ML has some delay in a few minutes (~7min) (Fig.3b).

Figure 3. LR signals vs time measured during growth of MT-NL (a) and HT-NL(b). MOSS results of measurements (stress×thickness) vs thickness for MT-NL (c) and HT-NL -B (d). The black continuous curves are experimental data, and red dashed lines show average stresses.

It should be noted oscillatory change in the curvature evolution graphs for both NLs. As we have demonstrated previously in [13] the upper levels of the oscillating signal are related with a periodical appearance of the excess metal on the flat surface of the growing films, whereas the lower signal levels correspond to the bare AlN surface without a metal excess. Therefore, one can suppose that increase of oscillation amplitude corresponds to increase of grain’s area during MEE growth of NLs with alternative nitrogen and aluminum fluxes. Therefore, we evaluated the stress using the lower levels in the oscillating curvatures. Thus, MT-NL demonstrates the generation of a compressive stress of -1.5GPa at the beginning of growth, which are fully relaxed at a layer thickness of about 50 nm, as shown in Fig.3c. In contrast, Fig.3d shows that the growth of HT-NL is almost fully relaxed.

There are several processes determining the stress generation and relaxation in the grain films growing on mismatched substrate. First, crystal mismatch leads to giant compressive stress which is relaxed through generation of misfit dislocations. This process is determined by quality of the interface...
between AlN and c-sapphire but in the most studies the almost full relaxation of this stress was demon-
strated. The second mechanism of stress generation during grain coalescence in the NLs, theoretically
described by Nix and Clemence [5]. This stress is templated in the upper AlN films, as commonly
observed in the AlN films grown by MOCVD, where its contribution is decisive in the magnitude of the
residual stress [10]. However, some authors described the third mechanism of stress generation, related
with the transport of metallic atoms in the grain boundaries with a lower chemical potential compared to
the film surface. This results in intrinsic compressive stress in the film. Thus, if there are some residual
mismatch, coalescence and excess metal on the surface of growing AlN NL, competition between several
mechanisms of the generation of stresses with opposite signs can be assumed.

The continuously observed streaky RHEED pattern of AlN-A sample indicates its 2D surface
morphology, which is also confirmed by the results of its post-growth SEM and AFM characterization,
shown in Fig.4. Sample B showed some transient streaky-spotty RHEED pattern during the initial growth
of AlN-BL by MME, which after the growth of the first 100nm became streaky and maintained the same
during the rest growth. The surface morphology of sample B (not shown) is the same as for sample A, and
both AlN layers exhibit RMS values of about 0.7 nm, as shown in Fig.4d.

Figure 4. SEM cross-(a) and plan-(b) view images of the surface of sample A, its RHEED pattern
during growth of AlN BL (c) and AFM image (d).

Figure 5 shows the generation of tensile stress in both samples of AlN BLs, grown by MME
under similar stoichiometric conditions, but at the different growth temperature. The MT-AlN (sample A)
exhibited very low magnitude of this stress (+0.05GPa), whereas much higher stress of about 0.85GPa
was found in the sample B grown at HT. Moreover, in the latter layer the transition from initial almost
stress-free growth to growth with the tensile stress observed after decrease of the stoichiometric ratio,
used for MME AlN BL, from 1.1 to 1. A further increase in stoichiometric conditions did not lead to a
relaxation of this tensile stress in the sample B, and only slight decrease in its value from 0.85 to
0.68 GPa was observed only when the growth temperature dropped from 850 to 780°C, as shown in
Fig.5b. Then, it is necessary to note the lack of oscillations in end of the growth of both samples under
cw-growth mode using Me-rich growth conditions that confirms hypothesis on relation of the oscillation
with a periodic appearance of the Me-adlayer on the growing surface.

Figure 5. The measured by MOSS values of (stress×thickness) vs thickness in AlN layers of A (a) and
B (b) samples. The curves with oscillations demonstrate experimental data, and red dashed lines show
average stress values.
The observed different values of the stress in the growing AlN BLs can be explained by the dominance of the grain coalescence processes in the HT AlN BL (sample B), whereas one can suppose the suppression of this mechanism in the MT AlN BL (sample A) grown at the lower growth temperature. In addition, stress evolution in this film can indicate an importance of intrinsic compressive stress due to metal adatom insertion into the top of the grain boundaries in the AlN epilayers grown by PA MBE at the Me-rich conditions. This type of stress evolution is drastically differed from the commonly observed high tensile stresses in AlN/c-Al2O3 grown by high-temperature MOCVD. However, further studies of the stress-related phenomena should be carried out to elucidate the optimal growth conditions of low defect and stress-free AlN BLs.

According to the ω–scans from the XRD (0002) reflections with FWHM of 600 and 167 arcsec the screw-type dislocation density of $7.8 \times 10^8$ and $6.1 \times 10^7$ cm$^{-2}$ can be estimated for the samples A and B, respectively [14]. In contrast, measuring the reflex (10-15) for these samples gives the opposite ratio. Its FWHM is 960 arcsec in sample A ($\approx 5.1 \times 10^9$ cm$^{-2}$), whereas it is equal to 1799 ($\approx 1.8 \times 10^{10}$ cm$^{-2}$) arcsec in sample B. Thus, a total density of TDs is lower in MT AlN sample, which also have almost negligible stress.

Conclusion

Thus, PA MBE growth of AlN NLs and BLs using MEE and MME growth modes at the slightly Me-rich conditions allows one to achieve an average stress in the films with tensile stress with a value varying from $\approx 0.6$-0.8 GPa to values close to zero. The former value was observed in the films grown mainly at the high growth temperature of 850°C, whereas the stress-free growth was realized at the lower growth temperature of 780°C. Moreover, the films grown at the moderate temperatures demonstrate lower TD density of $\approx 6 \times 10^9$ cm$^{-2}$. More studies of different mechanisms of generation of both tensile and compressive stresses in growing NL and BL are needed to clarify the obtained result.

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