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Thick Hydride Vapour Phase Epitaxial GaN Layers Grown on Sapphire with Different Buffers

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We report a comparative study of the crystalline quality of thick GaN layers grown by hydride vapour phase epitaxy, using a nitridation and a GaCl pretreatment of the sapphire as well as a reactive sputtered AlN buffer and metalorganic chemical vapour deposition grown GaN 'template' layers. The structure quality was investigated using X-ray diffraction measurement and cathodoluminescence spectroscopy and imaging of cross-section of the films. The morphology of the layers was revealed by optical and atomic force microscopy. A distinct reduction of both the columnar near-interface region and the domain formation were observed in layers grown on AlN and GaN 'template' buffers resulting in improved bulk quality and significant smoother film surfaces.

1. Introduction

Despite large differences in lattice parameters and thermal expansion coefficients, technologically promising GaN layers have been grown on sapphire substrates. Nevertheless, the growth of GaN layers on sapphire is plagued by high densities of extended defects such as dislocations, domain boundaries and cracks. Most of these problems have been significantly reduced or completely solved in thin metalorganic chemical vapour deposition (MOCVD) grown layers by using low temperature grown AlN [1,2] or GaN [3] buffer layers. A great promise for the combination of MOCVD-grown GaN 'templates' with subsequent homoepitaxially grown GaN by molecular beem epitaxy has been reported as well [4]. Another approach is to develop hydride vapour phase epitaxy (HVPE) grown quasi-bulk material suitable for subsequent homoepitaxial growth. Over the last years considerable progress has been made in the crystalline quality of thick GaN layers grown by HVPE [5,6], although many defect-related issues of thick films are still rather controversial and undesirable. One example is the origin and mechanism for reduction of large-scale nonuniformities close to the substrate–layer interface responsible for the residual highly n-type conductivity [7,8].

In this work, we study in a comparative way the effect of AlN and MOCVD-GaN 'template' buffers, and of nitridation and GaCl pretreatment of sapphire substrates on the near-interface columnar defects and on the crystalline quality of subsequently HVPE grown GaN layers.

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2. Experimental

GaN layers were grown in a conventional HVPE system on *a*-plane sapphire substrates at a growth temperature of 1080 °C. A nitridation pretreatment in 20% NH_3 for 15 min was used (sample #1). GaCl pretreatment in a 0.5% stream for 10 min was used after the nitridation of the sapphire for the second group of layers (sample #2). AlN buffer layers with a thickness of 500 Å were reactively sputtered from elemental targets in a UHV sputtering system using pure N_2 as the working gas at a substrate temperature of ≈1000 °C (sample #3). MOCVD-GaN:Si 'template' layers grown in the Aixtron application laboratory with a thickness of 2 μm were used as well (sample #4). The effect of buffer layers was studied for variable thickness of buffers, for different HVPE growth rate, and also as a function of substrate orientation (*a*-plane vs. *c*-plane), and the results are presented in detail elsewhere. In this work, we compare the results obtained for representative thick HVPE-GaN layers grown at the same growth conditions with thicknesses of about 45 μm.

Atomic force microscopy (AFM) measurements were performed with a Nanoscope IIIa instrument operated in tapping mode using Si tips. Cathodoluminescence (CL) spectroscopy and imaging were performed, using a Gemini 1550 LEO electron microscope equipped with a MONOCL Oxford Instrument. The CL spectra and images were taken at 7 K with an accelerating voltage of 15 kV. The spatial resolution of the images is determined by the interaction volume of one micrometer from the surface and a diffusion length considerably less than a micrometer. The X-ray diffraction (XRD) measurements were performed with a Philips MRD system with a Cu radiation source operated at 40 kV and 40 mA. The triple axis configuration was utilised in the HRXRD measurements with a resolution limit of ≈12 arcsec.

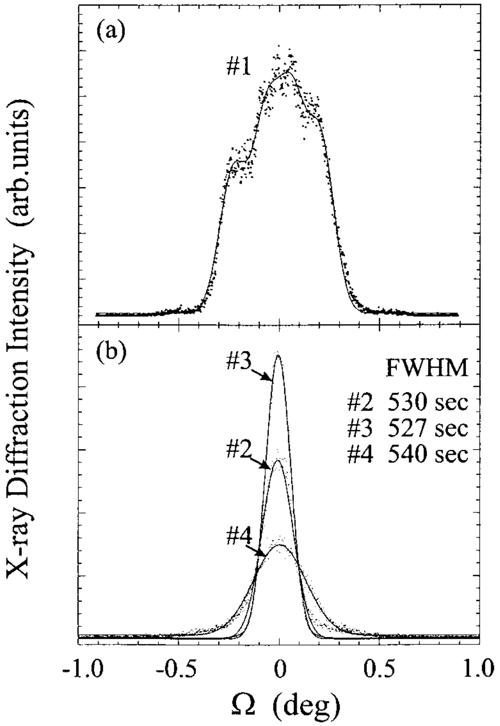

Fig. 1. X-ray ω-scans of GaN layers with a thickness of ≈45 μm grown on sapphire: a) with a nitridation pretreatment (sample #1); b) with a GaCl pretreatment (sample #2), with an AlN buffer (sample #3) and with a GaN 'template' layer (sample #4)

3. Results and Discussion

The HRXRD was used to determine the crystallinity and the orientation of both buffer and main layers. The θ–2θ and ϕ-scans indicate that buffer layers have a single crystalline character and a good symmetry. The ω measurements show full

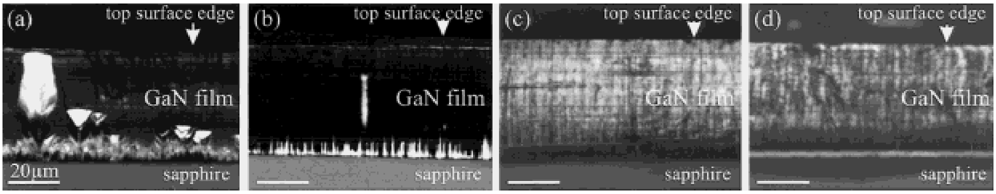

Fig. 2. Panchromatic CL images of cross-sections of the GaN layers grown at the same growth conditions on a) nitrided sapphire, b) GaCl pretreated sapphire, c) AlN buffer, and d) GaN 'template' layer

width at half maximum (FWHM) values in the ranges of 33 to 40 arcsec and 270 to 300 arcsec for AlN and GaN 'templates', respectively, indicative of good crystalline quality.

Fig. 1 shows the ω-scans of the four representative samples of thick HVPE-GaN layers. It can be seen that the spectrum of GaN film grown on a nitrided sapphire (Fig. 1a) exhibits multiple peaks indicative of a presence of several domains, while the spectra of the rest samples show single ω-peak (Fig. 1b) and single domain structure, respectively. The FWHM of the [0002] reflection in ω-scans show a comparative quality of the samples #2, 3 and 4 (Fig. 1b). While the ω-scan provides information on the mosaic structure of the films, the peak width of the ω–2θ scan provides more direct information on the crystal coherence which can be directly related to defect density in the material [9]. Values of 47, 40, 40 and 39 arcsec were obtained for samples #1 to 4, respectively. These results indicate an improved quality of GaN films grown on buffered substrates.

A CL study of cross-sections of the films reveals three different zones of thick GaN layers grown without buffers: a high defect density nonradiative nucleation layer with thickness of about or less than 1 μm; a near-interface columnar bright emission region; and a good quality main part of the layer. The spatially resolved CL spectra taken from the bright columnar region show a broad band typical of highly doped material [8]. In the GaN layers grown without buffers (Fig. 2a, b), the thickness of the defective columnar region varies from 5 to 20 μm depending on growth conditions and some of the columns protrude up to the film surface. Using buffer layers, the columnar region can be removed (Fig. 2c, d) resulting in improved electrical properties of the films. The near-interface region in sample #4 appears as a bright stripe with a thickness of about 2 μm (Fig. 2d) attributed to the GaN 'template' layer properties (n-type in the 10^{18} cm^{-3} range).

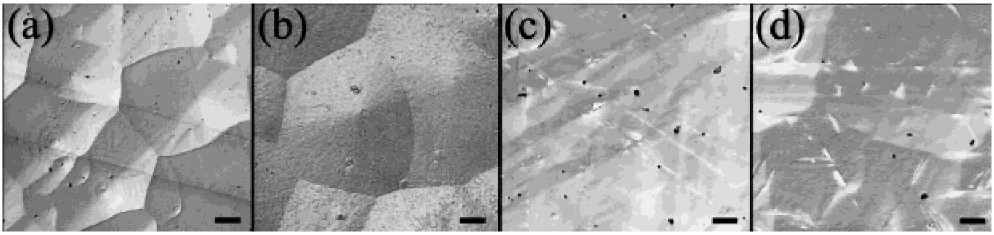

Fig. 3. Optical microphotographs of the growth surface of GaN layers grown on a) nitrided sapphire, b) GaCl pretreated sapphire, c) on AlN buffer, and d) GaN 'template' layer. The marker represents 100 μm

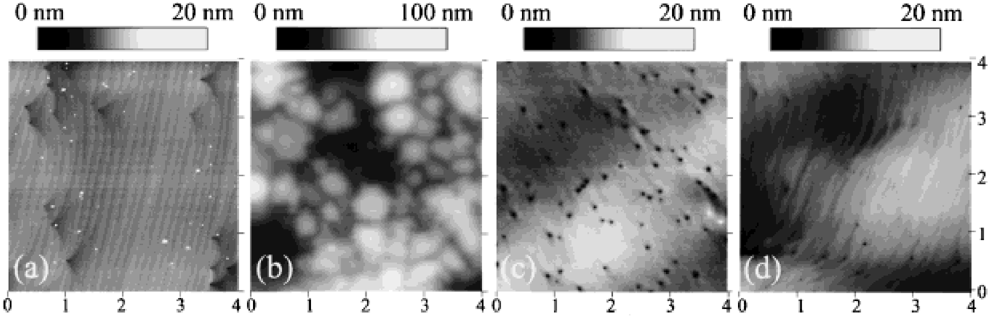

Fig. 4. AFM images of the top surface of the GaN layers grown on a) nitrided sapphire, b) GaCl pretreated sapphire, c) AlN buffer, and d) GaN 'template' layer

The surface topography images of GaN layers grown on sapphire without buffers show a hillock type morphology (Fig. 3a, b) which is a typical surface for thick GaN films. The surface of GaN layers grown on AlN and GaN 'template' layers exhibits a significantly smoother morphology (Fig. 3c, d), indicative of higher lateral/vertical growth ratio.

The AFM images (Fig. 4) of the layers reveal the microstructure of the surfaces and show a flat morphology of the layers, with an exception of sample #2 (Fig. 4b) grown on GaCl pretreated sapphire. Despite good crystalline parameters, the sample #2 shows a distinct rough topography with a hill and valley structure with a height difference of about 200 nm between the highest and the lowest points. The surface of the layers grown on nitrided sapphire (Fig. 4a) is quite smooth, consisting of an array of terraces with a width of 120 to 140 nm separated by steps of 5 to 6 Å height. Two-dimensional step-flow growth is dominant for this sample, as evidenced by well defined steps and terraces. In Fig. 4a one can also see that the steps are strongly affected by holes with a diameter of $\approx$50 nm. The steps bend into the holes and are pinned by them. These holes or depression pinning of the steps can be correlated to the surface terminations of threading dislocations [10]. The dislocations should be of either a pure screw or a mixed screw-edge character with a screw component with a Burgers vector equal to the *c*-axis lattice parameter of GaN. The layers grown on MOVPE-GaN 'templates' (Fig. 4d) exhibit similar terrace-type morphology. There are also some holes and depression pinning of the steps, however, some smaller holes can be seen which do not terminate the steps. They might be attributed to pure edge dislocations which do not have a component of Burgers vector perpendicular to the surface. The smoothest surface is shown for GaN grown on AlN buffer (Fig. 4c). All holes show pure edge threading dislocations. No terraces observed is indicative of a layer-by-layer growth mechanism.

4. Summary

Thick GaN films grown by HVPE on both AlN and GaN 'template' buffers have been demonstrated, with improved morphology and crystalline quality. A reduction in the domain structure formation has been shown by X-ray ω-curve spectra of films using both buffers and GaCl pretreated sapphire. The film–substrate interface shows an absence of columnar structure in layers grown on buffers, compared to the interface of thick GaN layers grown without buffers. The typical hillock-type surface morphology for thick GaN layers grown with both nitridation and GaCl pretreatment is observed, although the GaCl pretreatment results in some roughness on micro-morphology. The use of AlN or MOCVD-GaN 'template' buffers results in significant smoother film surfaces, both for macro-morphology and micro-morphology.

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