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Origin of a-plane (Al,Ga)N formation on patterned c-plane AlN/sapphire templates

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Abstract. a-plane (Al,Ga)N layers can be grown on patterned c-plane AlN/sapphire templates with a ridge direction along $[1\bar{1}00]Al_2O_3$. Scanning nanobeam diffraction reveals that the formation of a-plane layers can be explained by nucleation of c-plane (Al,Ga)N with $[11\bar{2}0]$(Al,Ga)N ‖ $[0001]Al_2O_3$ at the ridge sidewalls. Faster growth of the top $(11\bar{2}0)$(Al,Ga)N facet in the vertical direction leads to the overgrowth of c-plane (Al,Ga)N nucleated on the horizontal ridge and trench surfaces. Phase separation into binary GaN and AlN takes place during the first growth stages. However, this fades out and does not influence the composition of the final thick a-plane (Al,Ga)N layer.

1. Introduction

Most of today's III-nitride based device heterostructures are grown along the c-axis direction of the wurtzite structure (so called c-plane layers). However, optoelectronic devices obtained on the basis of c-plane nitrides suffer from spontaneous and piezoelectric polarisation fields which appear in the noncentrosymmetric wurtzite structure along the polar c-axis. To overcome this problem growth along non-polar or semipolar directions was proposed [1]. However, this growth usually requires application of a number of different substrates and/or substrate orientations. For example, growth of non-polar a-plane GaN and AlN layers is generally achieved on r-plane sapphire [2]. Recently, we have shown that predominant growth of thick a-plane Al-rich (Al,Ga)N layers is possible by epitaxial lateral overgrowth (ELOG) of patterned c-plane sapphire with ridges arranged along $[1\bar{1}00]Al_2O_3$ [3]. This ridge direction in ELOG of III-nitrides often leads to formation of strongly faceted non-planar surfaces as well as to problems with layer coalescence. This led to the common opinion that this ridge geometry is inappropriate for the fabrication of planar III-nitride layers of high quality. In this study we focus on the orientation and compositional homogeneity of (Al,Ga)N layers during initial growth stage on c-plane AlN/sapphire templates patterned along this $[1\bar{1}00]Al_2O_3$ direction.

2. Experimental details

In our experiment a 15 µm thick $Al_xGa_{1-x}N$ layer with a nominal composition of $x = 0.95$ was grown using hydride vapour phase epitaxy (HVPE) and ELOG on a patterned c-plane template consisting of a 600 nm thick AlN buffer layer on sapphire (see [3] for details). The ridges in the c-plane AlN/sapphire template were oriented along the $[1\bar{1}00]Al_2O_3$ direction, which results in a-plane (i.e.

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$(11\bar{2}0)Al_2O_3$) trench sidewalls. Secondary electron microscopy (SEM) and scanning transmission electron microscopy (STEM) including high-angle annular dark-field (HAADF) imaging, energy dispersive X-ray spectroscopy (EDXS) and scanning nanobeam diffraction (SNBD) have been applied to understand the layer structure. The ASTAR/DigiSTAR system of NanoMEGAS has been used for mapping of crystallite orientation and phase in STEM. The crystallographic orientation maps were obtained by comparison of experimental electron diffraction patterns to a set of patterns calculated for different phases.

3. Experimental results

Figure 1a shows a cross-sectional SEM image of an a-plane (Al,Ga)N layer grown in our experiment. The a-plane orientation was confirmed by electron backscatter diffraction as published elsewhere [3]. During the growth there is simultaneous (Al,Ga)N nucleation on the horizontal c-plane surfaces, i.e. on the AlN/sapphire ridges (region 1) and in the trenches (region 2), as well as on the vertical ridge sidewalls (regions 3, 4). In order to understand the formation of a-plane layers on patterned c-plane AlN/sapphire templates we have analysed the layer structure during the first growth stages. Figure 1b shows an exemplary HAADF STEM image of a $[1\bar{1}00]$-oriented Al_2O_3 ridge with AlN template on top, which was overgrown by Al-rich (Al,Ga)N using HVPE. The HAADF STEM micrograph shows a non-uniform intensity distribution within the (Al,Ga)N layer with bright regions of triangular shape appearing at the ridge sidewalls. EDXS mapping suggests a preferential Ga accumulation at these positions (figure 1c). As result, there is no Ga incorporation into the (Al,Ga)N layer at the beginning of the growth, and the ridges are covered with binary AlN. The formation of ternary (Al,Ga)N is delayed most probably until the ridge sidewalls are completely overgrown by AlN. Notice that EDXS suggests the formation of binary AlN on top of the AlN template as well as in the trenches. However, there is an inhomogeneous intensity in the HAADF image with regions growing on the horizontal surfaces (regions 1, 2) appearing darker than regions growing on ridge sidewalls (regions 3, 4). We attribute these differences in intensity to different channelling conditions, which can appear in sample regions with the same composition, but different orientation.

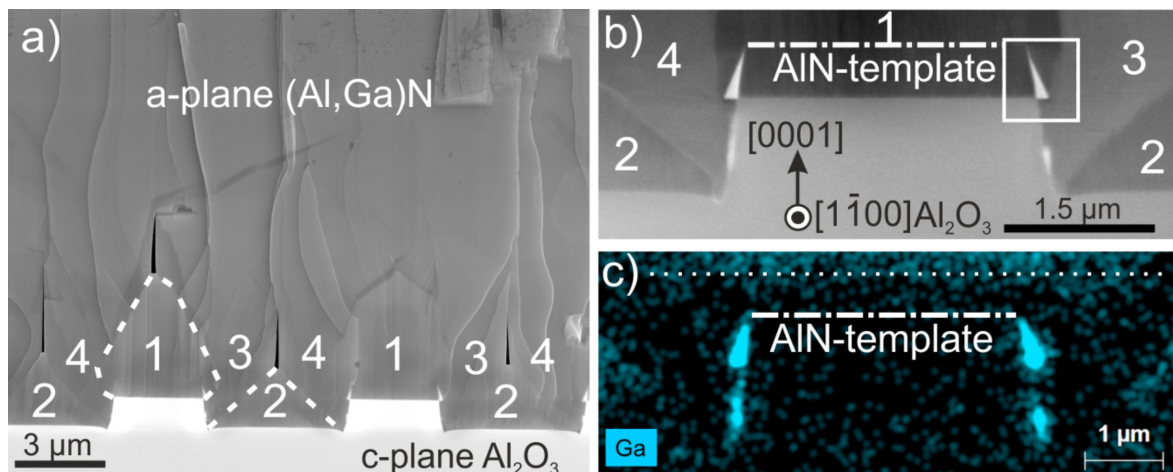

Figure 1. a) Cross-sectional SEM micrograph of (Al,Ga)N layer grown on patterned c-plane sapphire with ridge direction along $[1\bar{1}00]Al_2O_3$; b) HAADF STEM of an overgrown AlN/Al_2O_3 ridge; c) EDXS mapping of Ga-K_α peak intensity in the region shown in b). The white dash-dotted line indicates the interface between the AlN template and the (Al,Ga)N layer grown on top, whereas the white dotted line indicates the delayed onset of Ga incorporation into the growing (Al,Ga)N layer. The white rectangle in in b) marks the area for further SNBD analysis.

SNBD has been applied to prove this suggestion. Figures 2b and c show exemplary SNBD maps of a part of a $[1\bar{1}00]$-oriented Al_2O_3 ridge with AlN template on top (see the rectangle marked in figure

1b). The analysed sample region is shown in a virtual bright-field image in figure 2a, which was reconstructed by depicting the intensity of the none-diffracted beam for each pixel of the scan. According to the orientation maps the AlN template shows the classical orientation on c-plane sapphire: $(0001)[11\bar{2}0]$AlN ∥ $(0001)[1\bar{1}00]Al_2O_3$. During the HVPE layer deposition (Al,Ga)N grows with this orientation relation on top of the AlN template (region 1 in figure 1a) and in the trenches (region 2 in figure 1a). Simultaneously, competing growth takes place on the Al_2O_3 ridge sidewalls. The sidewall surface corresponds to the sapphire a-plane. As indicated by the colour zone selectors in the bottom line of figure 2 the layer orientation in this region corresponds to $(0001)[11\bar{2}0]$AlN ∥ $(11\bar{2}0)[0001]Al_2O_3$. This layer orientation is well known from growth experiments on planar a-plane sapphire [4]. Consequently, the formation of an a-plane (Al,Ga)N layer in our experiment can be explained by nucleation of c-plane (Al,Ga)N at the ridge sidewalls of the patterned sapphire substrate (figure 2e).

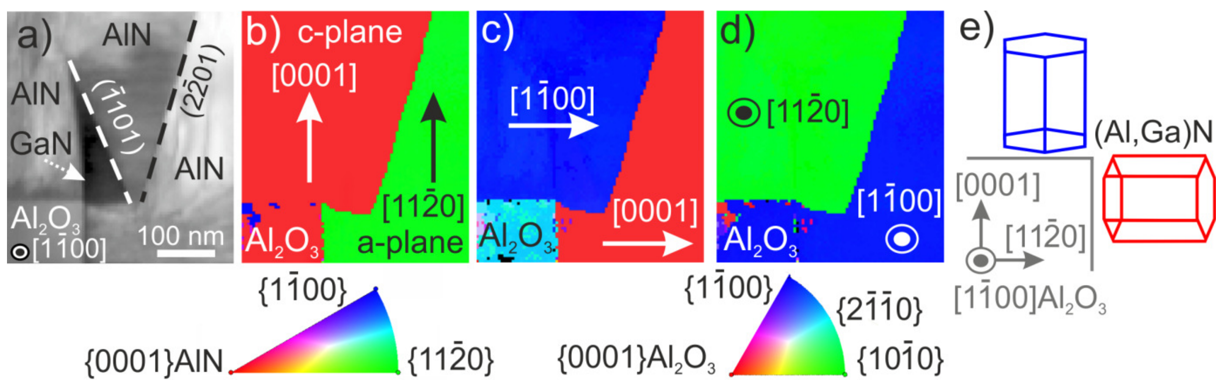

Figure 2. Bright field image (a) reconstructed from a set of electron diffraction patterns obtained by SNBD and crystallographic orientation maps (b), (c) and (d) for three different directions. The images are obtained at the sample region marked by a rectangle in figure 1b. Arrows mark the normal of the identified lattice planes, which can be extracted from colour zone selectors; e) sketch showing the wurtzite unit cell arrangement on a ridge and its sidewall.

Analysis of individual diffraction patterns shows that Ga-rich regions observed by EDXS correspond to wurtzite structure. Consequently, a phase separation into binary GaN and AlN compounds appears at the beginning of the growth. This phase separation can be explained by a preferential agglomeration of Ga atoms on non-polar surfaces. Notice that the a-plane material originates only from the a-plane sapphire sidewalls, whereas the m-plane sidewalls of the AlN template supply nucleation sites for c-plane GaN. These GaN regions contain a high density of basal plane stacking faults (BSFs), which can be explained by a high lattice mismatch between the (0002) lattice plane distances in the AlN template and GaN growing on its m-plane side facet. The presence of BSFs explains the appearance of vertical streaks in $[11\bar{2}0]$ electron diffraction patterns (DPs) of GaN (figure 3b), whereas no streaks were observed in DPs of the AlN template (figure 3c).

Analysis of crystal faceting can supply important information on the evolution of the growth fronts. As visible in figure 2a, the GaN region formed on the m-plane sidewall of the AlN template develops facets which are close to the $\{\bar{1}101\}$ facets generally observed during growth of c-plane III-nitride layers in ELOG experiments with this pattern geometry. However, GaN is subsequently overgrown by c-plane AlN growing on the top of the AlN template and propagating into the lateral direction with formation of $\{2\bar{2}01\}$ facets. The observed crystallite shape suggests that the overgrowth of c-plane GaN by c-plane AlN starts at the upright point of the triangular shaped GaN grain and propagates not only laterally, but also to the bottom (down to the base of the triangular-shaped GaN grain). The lateral propagation of the $\{2\bar{2}01\}$ facets in c-plane AlN might be slower compared to the vertical growth of a-plane (Al,Ga)N nucleating at ridge sidewalls, which would explain the fast overgrowth of

c-plane (Al,Ga)N growing at the horizontal surfaces (regions 1 and 2 in figure 1a) and the coalescence of the a-plane (Al,Ga)N regions.

Using SNBD analysis it is possible to determine crystallite orientation and distinguish between different phases. We have tried to distinguish between AlN and GaN using the ASTAR software of NanoMEGAS. The phase identification was carried out by cross-correlation of each experimental electron diffraction pattern with calculated patterns for AlN and GaN. The obtained phase map is shown in figure 3a. As visible, the regions (i) and (ii) were correctly assigned to AlN and GaN, whereas phase identification in region (iii) failed (compare figures 2a and 3a). In particular, AlN nucleating at the ridge sidewall and viewed in the $[1\bar{1}00]$ zone axis was mistakenly assigned to the GaN phase. A detailed analysis of DPs suggests that this technique allows distinction between AlN and GaN viewed in the $[11\bar{2}0]$ direction (figures 3b,c). However, any accurate phase identification fails for the $[1\bar{1}00]$ zone axis. The lower accuracy of the phase recognition observed for the $[1\bar{1}00]$ zone axis can be explained by a lower number of reflections present in these electron diffraction patterns (compare figures 3c and d).

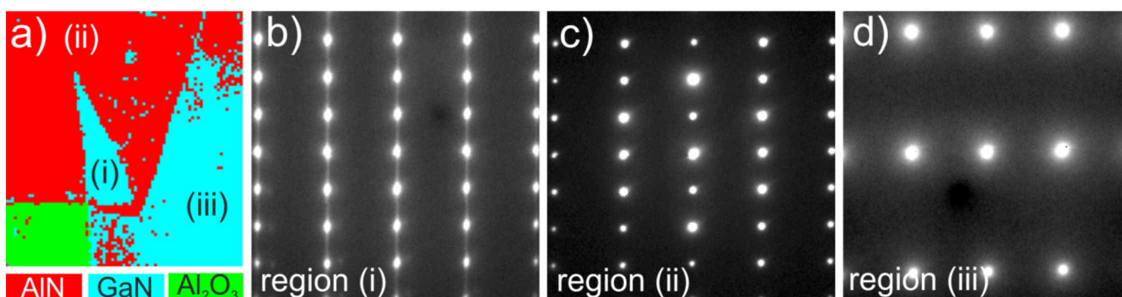

Figure 3. Calculated phase map (a) of the sample region shown in figure 2 and experimental diffraction patterns from (b) c-plane GaN (region (i)) viewed in $[11\bar{2}0]$GaN; (c) c-plane AlN (region (ii)) viewed in $[11\bar{2}0]$AlN; (d) a-plane AlN (region (iii)) viewed in $[1\bar{1}00]$AlN.

4. Conclusions

We have analysed composition and crystallographic orientation of (Al,Ga)N layers grown on c-plane AlN/sapphire substrates patterned in the $[1\bar{1}00]Al_2O_3$ direction. Nucleation of c-plane (Al,Ga)N on vertical a-plane ridge sidewalls of sapphire results in a-plane (Al,Ga)N orientation with respect to the horizontal growth front. Simultaneously, there is a formation of c-plane crystallites on the horizontal surfaces, i.e. on the top of the AlN/sapphire ridges and in the trenches. We suggest that a slow growth rate of $\{2\bar{2}01\}$ facets in c-plane (Al,Ga)N prevents coalescence of the c-plane (Al,Ga)N regions. In contrast, fast propagation of a-plane crystallites in the vertical $[11\bar{2}0]$ (Al,Ga)N direction is suggested to be responsible for the rapid overgrowth of the c-plane regions and formation of continuous a-plane layers. Phase separation into binary GaN and AlN is observed during the first growth stages, which leads to a delay in formation of ternary (Al,Ga)N.

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