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Analysis of crystal orientation in AlN layers grown on m-plane sapphire

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ABSTRACT

Our study reports on the microstructure of AlN layers grown on m-plane sapphire by metal organic vapor phase epitaxy. We have found that AlN can nucleate with three different orientations on the m-plane sapphire surface: semipolar $(11\bar{2}2)$ and $(1\bar{1}03)$ as well as m-plane $(1\bar{1}00)$. Depending on the growth conditions, i.e. V/III ratio, the differently oriented crystallites exhibit different lateral and vertical growth rates. At a low V/III ratio of 626 the vertical growth rate of semipolar $(11\bar{2}2)$ AlN regions is much lower than that of the $(1\bar{1}03)$ and $(1\bar{1}00)$ oriented grains, which results in an almost complete lateral overgrowth of the $(11\bar{2}2)$ AlN oriented regions. In contrast, a high V/III ratio of 1043 leads to the formation of uniform semipolar $(11\bar{2}2)$ AlN layers. Nevertheless, the formation of differently oriented AlN crystallites could not be suppressed completely. These randomly appearing crystallites still show a high vertical growth rate and lead to a deterioration of the surface morphology.

1. Introduction

III-nitride based materials are currently one of the most important families of semiconducting materials for light emitting diodes and laser diodes operating at wavelengths ranging from the deep ultraviolet to the green region. Among them, AlN-based heterostructures are especially important for light emitters operating in deep ultra–violet (UV) spectral range (below 280 nm), which can be used for water purification, gas sensing and medical diagnostics [1].

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 polarization fields which appear in the non-centrosymmetric wurtzite structure along the polar *c*-axis [2]. Thus, growth of so-called non-polar (i.e. m-plane $(1\bar{1}00)$ or a-plane $(11\bar{2}0)$) as well as semipolar (i.e. $(11\bar{2}2)$, $(1\bar{1}03)$, etc.) nitride layers has been intensively investigated in the last decade [2,3]. In particular, GaN growth on m-plane sapphire can result in a number of different orientations, e.g. semipolar $(11\bar{2}2)$ [4–9], $\{1\bar{1}03\}$ [7–10] or m-plane $(1\bar{1}00)$ [8,11]. Whereas most of the recent studies focus on the growth of nonpolar and semipolar GaN layers, less attention has been given to AlN. A detailed analysis of the structure of semipolar $(11\bar{2}2)$ AlN layers grown by plasma-assisted molecular beam epitaxy (PAMBE) on m-plane sapphire has been reported [12,13]. Simultaneous formation of crystallites with two orientation relations has been observed: semipolar $(11\bar{2}2)$ and non-polar m-plane $(1\bar{1}00)$. It has been noticed that depending on the applied growth conditions m-plane AlN nanocrystals can be restricted to the interfacial zone, and overgrowth by semipolar $(11\bar{2}2)$ AlN can be achieved. Recently, Stellmach et al. reported on a systematic study of the growth of semipolar AlN [14] as well as $Al_xGa_{1-x}N$ [15] by metal-organic vapor phase epitaxy in the whole compositional range $(0<x<1)$. Particularly for binary AlN layers a formation of morphological disruptions by dot-like structures was observed on the surface of semipolar $(11\bar{2}2)$ layers by atomic force microscopy (AFM) [14]. Obviously, these structures deteriorate the morphology of the semipolar AlN layer. It has been shown that lateral size and height of these dot-like structures strongly depend on the reactor pressure [14]. However, the origin of formation of these morphological features has not been clarified. It was assumed that the dot-like features might be attributed either to regions with the same orientation, but different morphologies

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(e.g. due to growth facet instabilities), or to crystallites with different orientations. In the present study we report on the influence of V/III ratio on the microstructure of AlN layers on m-plane sapphire and reveal the nature of the dot-like features observed on the semipolar AlN surface. Using transmission electron microscopy (TEM) we will show that variation of V/III ratio during the nitride deposition allows tuning of the crystallite growth and thereby the layer orientation: from a layer consisting of differently oriented crystallites to a preferentially oriented semipolar $(11\bar{2}2)$ AlN layer.

2. Experimental

AlN layers were grown on m-plane sapphire by metal-organic vapor phase epitaxy (MOVPE). The growth was performed in an Aixtron 3×2 inch close-coupled showerhead MOVPE reactor. Trimethylaluminium (TMAl) and ammonia (NH_3) were used as precursors. After heating up the substrates up to 1100 °C in H_2 ambient and nitridation with ammonia, TMAl was supplied with 107 μmol/min. During AlN growth the ammonia supply was 67 mmol/min or 111.6 mmol/min resulting in V/III ratios of 626 or 1043, respectively. Two samples were grown at a V/III ratio of 626:one at a reactor pressure of 100 hPa and one at 200 hPa. Additionally, one sample was grown at a V/III ratio of 1043 and a reactor pressure of 150 hPa [14].

High resolution TEM (HRTEM), diffraction contrast imaging as well as scanning transmission electron microscopy (STEM) including high-angle annular dark-field (HAADF) imaging 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. The crystallographic orientation maps were obtained by comparison of experimental electron diffraction patterns to a set of patterns systematically calculated for all AlN crystal orientations with an angular step width of 1°. Three TEM specimens were prepared for each growth condition: two cross-sections parallel to the [0001] and $[11\bar{2}0]$ Al_2O_3 directions and one plan-view specimen.

3. Results and discussion

Cross-sectional TEM analysis reveals that AlN growth on m-plane sapphire at a V/III ratio of 626 results in simultaneous formation of AlN crystallites with three different growth planes: semipolar $(11\bar{2}2)$ AlN and $(1\bar{1}03)$ AlN as well as non-polar m-plane $(1\bar{1}00)$ AlN. Fig. 1 shows a schematic representation of these orientations, which are well known from GaN epitaxy on m-plane sapphire [8]. Using TEM analysis we have not observed any significant structural differences between the samples grown at reactor pressures of 100 and 200 hPa. Thus, in the following we will concentrate on results obtained at 200 hPa. Nevertheless, it should be kept in mind that TEM analysis yields only local results, so that differences visible only on a larger scale cannot be excluded.

An exemplary HRTEM image in Fig. 2a proves the simultaneous presence of crystallites with the growth planes $(11\bar{2}2)$ and $(1\bar{1}03)$. As schematically illustrated in Fig. 1c, the formation of twins is observed in the case of the $(1\bar{1}03)$ AlN growth plane (Fig. 2a). This observation is similar to that published by Vennegues et al. for GaN layers grown on m-plane sapphire [8]. The reasons why the twinning occurs for the $(1\bar{1}03)$ and not for the $(11\bar{2}2)$ orientation have been previously discussed elsewhere [8,9]. The twin formation can be explained considering substrate symmetry with respect to the epilayer crystal planes, which exhibit a low lattice mismatch, as well as an extension of the atomic arrangement of sapphire into the epilayer at the common interface. In the case of the $(1\bar{1}03)$ grains, the orientation is predetermined by the epitaxial relation between the (0001) facet of the epilayer and one of the sapphire $\{11\bar{2}0\}$ planes, inclined by 30° to the m-plane sapphire surface (as indicated in Fig. 3a). Sapphire contains two crystallographically equivalent $\{11\bar{2}0\}$ planes inclined to the m-plane surface. Each of them can lead to formation of a $(1\bar{1}03)$ domain. These two domains are the observed $(1\bar{1}03)$ twins, which can be described by a 180° rotation around the surface normal. Similar considerations can be made for the sapphire $\{10\bar{1}0\}$ planes, as has been previously described by Frentrup et al. [9]. Notice, that in general both polarities, i.e. $(1\bar{1}03)$ and $(1\bar{1}0\bar{3})$ with the corresponding twins, are possible. In the following, we will use the $(1\bar{1}03)$ notation to describe this type of crystal orientation, since polarity determination of these AlN crystallites was not the scope of the present study.

In contrast, the $(11\bar{2}2)$ orientation is predetermined by the epitaxial relation between the sapphire r-plane and the a-plane of the epilayer [5]. The r-plane nanofacets on m-plane sapphire surface incline only to one direction (Fig. 3b). Furthermore, the atomic arrangement in the sapphire r-plane does not show any inversion symmetry [16]. Thus, the $(11\bar{2}2)$ orientation becomes unique. Consequently, there is no twin formation [9] and unique polarity [16].

In addition to the semipolar $(11\bar{2}2)$ and $(1\bar{1}03)$ crystallite orientations non-polar m-plane crystallites are formed as shown in Fig. 2b for a different sample region. However, these crystallites are not visible in Fig. 2a due to the small analyzed specimen area. Depending on crystallite orientation, basal plane stacking faults (BSFs) with different inclination angles with respect to the growth plane, i.e. to the AlN/Al_2O_3 interface, were frequently observed in all types of crystallites.

SNBD analysis of larger specimen regions was carried out in order to reveal the orientation distribution in the AlN layers. Fig. 4 (b, c and d) show exemplary crystallographic orientation maps obtained by this method using a probe size of 1 nm. The orientation of the AlN film is shown in color, whereas the substrate

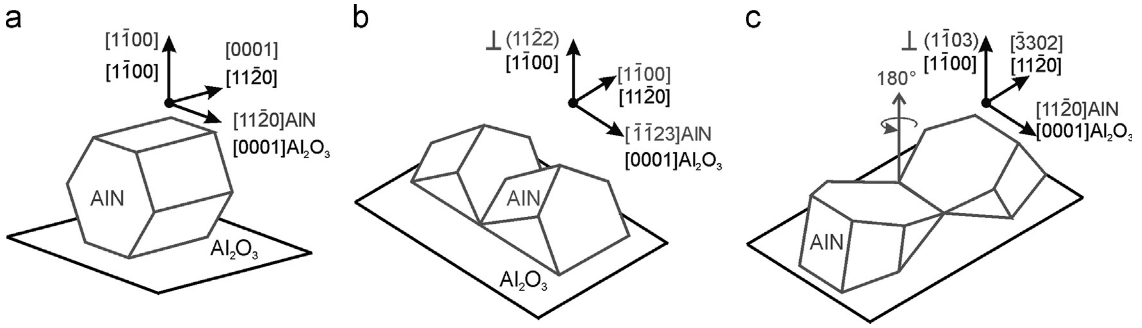

Fig. 1. Sketch of different epitaxial orientation relations observed in AlN layers grown on m-plane sapphire: a) non-polar m-plane $(1\bar{1}00)$ orientation; b) semipolar $(11\bar{2}2)$ orientation; c) semipolar $(1\bar{1}03)$ orientation.