Investigations of HVPE grown Nonpolar a-plane GaN on Slightly Misoriented r-plane Sapphire Substrates

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We have investigated the growth of nonpolar GaN templates by hydride vapor phase epitaxy (HVPE). This includes a systematic study of misoriented r-plane sapphire wafers with a miscut angle up to $\pm 1°$ *towards the c-axis of the crystal as starting substrates. Starting with an AlN nucleation layer approximately 3.3µm of nonpolar a-plane GaN are grown by metalorganic vapor phase epitaxy (MOVPE). The subsequent growth by HVPE was optimized to obtain flat and homogeneous layers with very good crystal quality. This was illustrated by very small full widths at half maximum (FWHM) of x-ray rocking curve (XRC) measurements of less than 500 arcsec. The surface quality was evaluated by scanning electron microscopy (SEM) and optical phase contrast microscopy showing a high number of surface defects (pits) for the samples with a high miscut towards the positive direction whereas a slight miscut of* $+0.5°$ *reduces the surface roughness. Additionally, smallest XRC FWHM values have been obtained for this particular miscut. A miscut towards negative directions results in lower crystalline quality and the surfaces shows hillock-like features. All GaN layers on misoriented substrates are tilted with respect to the substrate. Their tilt angle increases with the miscut angle. Its direction is opposed to the miscut of the sapphire.*

1. Introduction

Currently, commercially available devices based on the nitrides, like light emmitting diodes, are typically grown along the crystallographic c-direction. Unfortunately, huge piezoelectric fields are present within respective heterostructures, because of the lattice geometry and the induced strain. The resulting effects are mostly summarized as the quantum confined Stark effect [1]. One negative consequence is the reduced carrier recombination probability in quantum wells (QWs) [2]. To get rid of these negative effects, growth in nonpolar or semipolar direction is investigated by many groups. Typical nonpolar directions are a $[11\bar{2}0]$ and m $[10\bar{1}0]$. Due to the lack of real bulk GaN, the structures have to be grown either on foreign substrates like e.g. r-plane sapphire leading to a-plane GaN [3], or m-plane SiC [4] or $LiAlO_2$ [5] which results in m-plane GaN. However, the resulting material grown in those unusual directions typically contains a high number of defects. Other groups use sliced pieces from hydride vapor phase epitaxially (HVPE) grown GaN as starting substrates [6] resulting in a drastic reduction of the defect density. However, only small sized wafers of a few square millimeters are nowadays available for extremely high prices. In short the perfect substrate is still missing.

Therefore we are currently investigating the optimized growth of nonpolar quasi-substrates by HVPE which can be used for subsequent device epitaxy, as this method provides large enough growth rates. However, the nucleation on foreign substrates usually requires a wide range of optimization parameters and is quite challenging. Therefore, we are studying the optimization of the template layers by metal organic vapor phase epitaxy (MOVPE) for the subsequent hydride vapor phase epitaxial (HVPE) process. In this manner, we ensure to use the advantages of both systems. Misoriented sapphire wafers are known to improve the surface quality of c-plane GaN layers [7]. Here, we present a systematic study of the influence of slightly misoriented r-plane sapphire substrates on the growth of non-polar a-plane GaN layers via the combination of MOVPE and HVPE.

2. Experimental

All samples investigated within this study are grown on quarters of 2 inch epi-ready r-plane sapphire wafers resulting in a GaN growth in $[11\bar{2}0]$ direction. We used a commercial horizontal flow Aixtron AIX-200/4 RF-S reactor with the standard precursors trimethylgallium (TMGa), trimethylaluminum (TMAl) and ammonia (NH_3) for the MOVPE growth. As carrier gas, we used Pd diffused hydrogen. The process temperature was controlled by a pyrometer at the backside of the rotation tray. Before starting growth the substrates were exposed to an in-situ desorption step at 1200 °C for 10 min in hydrogen atmosphere. For the HVPE growth, a commercial horizontal flow Aixtron single-wafer system with five different heating zones was used. The carrier gas usually is a mixture of nitrogen and Pd diffused hydrogen. The precursors for the GaN growth are high purity NH_3 and GaCl which can be produced by streaming HCl gas over liquid gallium at a temperature of approximately 850°C.

As substrates we used r-plane sapphire wafers with a slight miscut ranging from $-1.0°$ to $1.0°$ towards the [0001]-axis of the sapphire in steps of $0.5°$. The miscut is defined positive when the angle between surface and c-direction is smaller than the angle between the r-direction and the c-direction of the sapphire. The different samples were labeled as samples A ($-1.0°$), B ($-0.5°$), C (reference sample, $\pm0°$), D ($+0.5°$) and E ($+1.0°$), were lower case letters denote the respective MOVPE templates and capital letters the final HVPE grown samples. The growth process was identical for all samples. The MOVPE growth was carried out on half wafers, whereas the HVPE growth was performed on quarters in order to have exactly the same growth conditions for several samples.

First, a high temperature AlN nucleation layer (NL) with a thickness of about 30 nm was deposited in MOVPE. Then an initial GaN layer with a total thickness of approx. 3.3 µm was grown in a two step growth process, interrupted by two in-situ SiN defect reduction layers. The first GaN layer was grown at a temperature of 1120 °C, a pressure of 150 hPa, with a V/III-ratio of ≈ 2180 and a growth rate of ≈ 1.2 µm/h. After 0.3 µm, SiN was deposited by applying a constant flow of silane (SiH_4) and ammonia (NH_3). The surface was fractionally covered with SiN acting as a nanomask and influencing the morphology of the overgrown layer resulting in a defect reduction [8,9]. At a nominal thickness of 1.0 µm a second SiN layer was deposited. Now, the growth parameters were changed to the second GaN step conditions: mainly the V/III-ratio was decreased to about 540, resulting in an increased growth rate of $\approx 2.5\,\mu$m/h. The growth parameters of the following single-step HVPE growth were as follows: A temperature of approximately 900 °C, a pressure of 900 hPa, with a V/III-ratio of ≈ 18 and a total thickness of approximately 12 µm.

For investigating the crystal quality, all samples were analysed by x-ray diffraction (XRD) rocking curve measurements (XRC) as well as low temperature (14K) photoluminescence (PL) spectra. The latter enables the qualification of typical defects in non-polar layers like basal plane stacking faults (BSFs) or prismatic stacking faults (PSFs) [10,11]. The surface quality was evaluated by scanning electron microscopy (SEM), optical phase contrast microscopy (OM) and atomic force microscopy (AFM).

3. Results and Discussion

Nonpolar a-plane GaN grown by MOVPE usually shows a typical morphology: A stripe like feature along the c direction and some surface pits [12]. We observed a reduced and more homogenous surface roughness for negative miscuts (samples a, b) whereas for positive miscuts the stripe like pattern becomes more pronounced and surface defects start to develop (d, e). This is consistent with the data reported by Araki et al. [13]. Even earlier, Imura et al. [14] reported a specular surface for miscut orientations of −0.5°

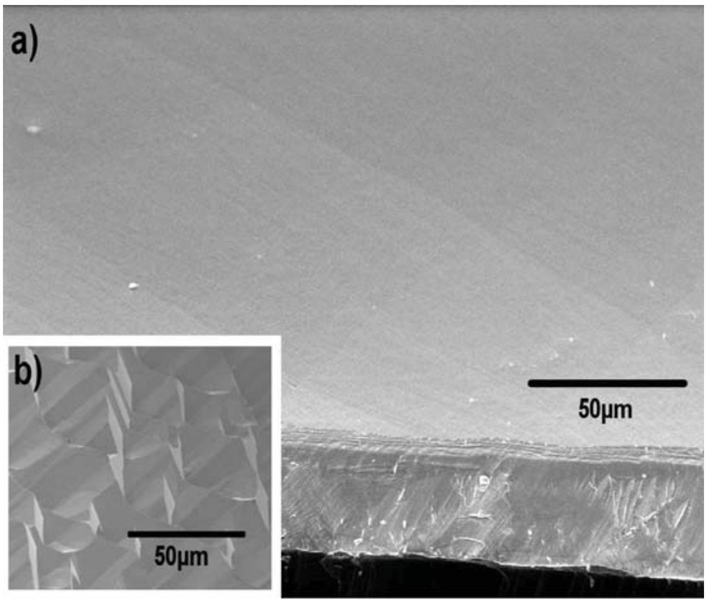

Fig. 1: a) SEM micrograph of the typical HVPE grown layers after optimization. The surface looks smooth without any facets, pits or stripe like features. b) The inset shows unoptimized HVPE GaN grown under high temperature conditions (color online).

(the definition of miscut angles is exactly opposite in this paper), which is also in line with our data. Investigations on the crystal quality via XRD and PL (not shown) are in contrast to these findings of the surface quality. The samples with higher miscut towards the positive direction (d, e) show reduced XRD linewidths and less stacking fault related luminescence (typically at 3.42 eV and 3.30–3.35 eV) compared to the PL signal from the near band edge region.

3.1 GaN grown by HVPE

Before growing on these miscut templates, we have optimized the growth of nonpolar GaN on exactly oriented r-plane wafers. The growth parameters like temperature, pressure, V/III-ratio, H_2/N_2-ratio, growth rate etc. have been varied to get layers with optimized surface and crystal quality. Layers grown before this optimization usually had a very rough and facetted surface (Fig. 1 b).

3.2 MOVPE template

The typical surface of HVPE grown GaN after optimization (Fig. 1 a) is very smooth and does exhibit neither the often observed surface defects ("pits") nor the stripe like features otherwise typical for a-plane GaN on r-plane sapphire grown by MOVPE.

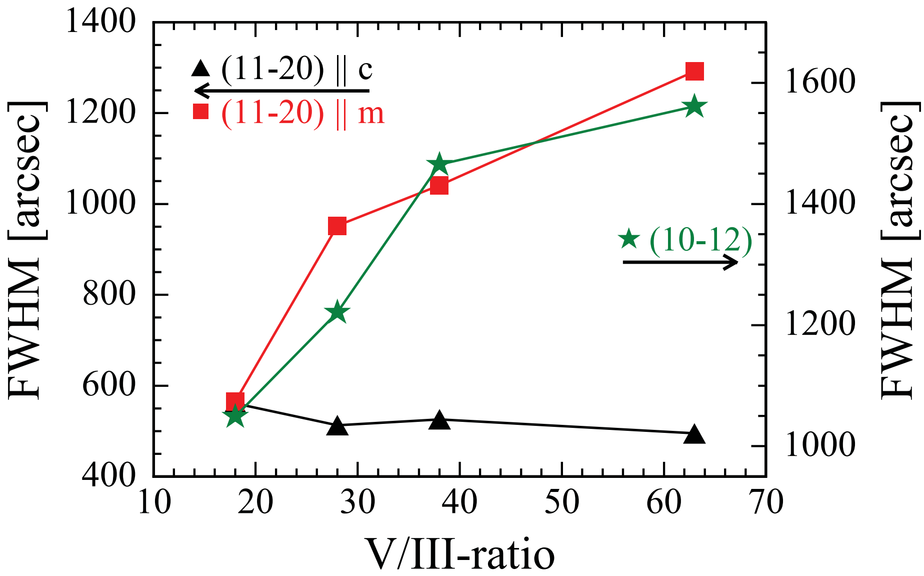

Fig. 2: XRC FWHM values for HVPE grown GaN layers with different V/III-ratio during growth. The improvement for lower values can clearly be seen (color online).

The optimized overgrowth in HVPE does not only improve the surface morphology but also the crystal quality of the layers. X-ray rocking curve linewidths of the symmetrical reflections could be reduced from around 750 arcsec for the MOVPE template to less than 500 arcsec for the HVPE grown layer regardless of the direction of the incident beam. Besides the temperature which is the key parameter for a flat surface ($T \approx 900\,°\mathrm{C}$, cf. Fig. 1), the ammonia flow rate is the growth parameter that shows the strongest influence. Only for low V/III-ratios (≤ 20) it is possible to grow a-plane GaN layers in HVPE with nearly no anisotropy in XRD measurements (Fig. 2). Smaller asymmetric reflection linewidths like $(10\bar{1}2)$ are also indicating a better crystal quality.

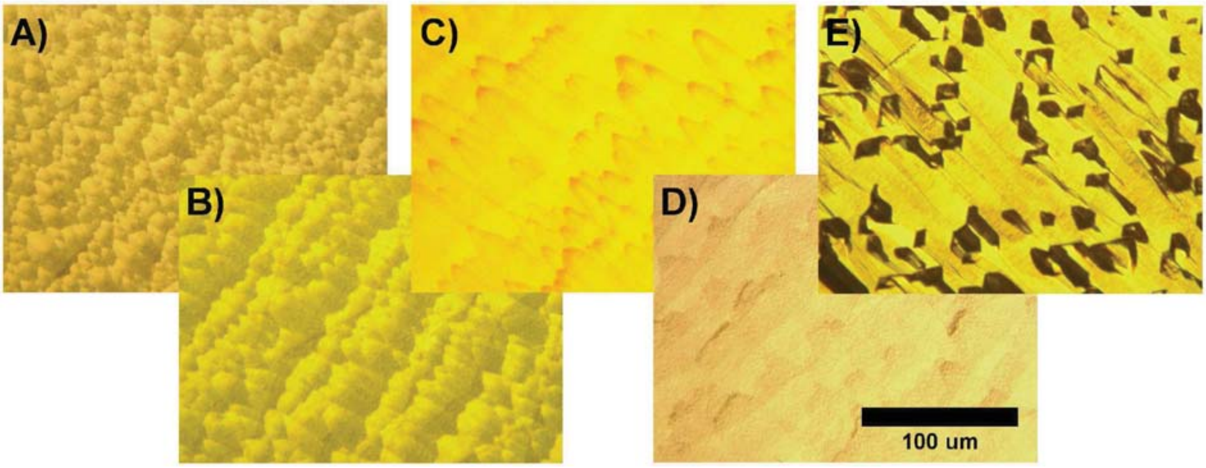

Fig. 3: Optical microscope images of the surfaces of the HVPE grown samples with different miscut A) $-1°$, B) $-0.5°$, C) $0°$, D) $+0.5°$, E) $+1°$ (color online).

The final HVPE GaN layers show a different surface morphology for the different miscut angles (Fig. 3). It seems that a negative miscut (samples A, B) leads to a rough surface with a high density of microscopic hillocks with an average diameter of approximately 20 µm. Both, no miscut and a miscut towards positive angles leads to drastically improved surfaces. The morphology gets more homogenous and the roughness is decreased. This also could be proven by AFM measurements (not shown). However, if the miscut is

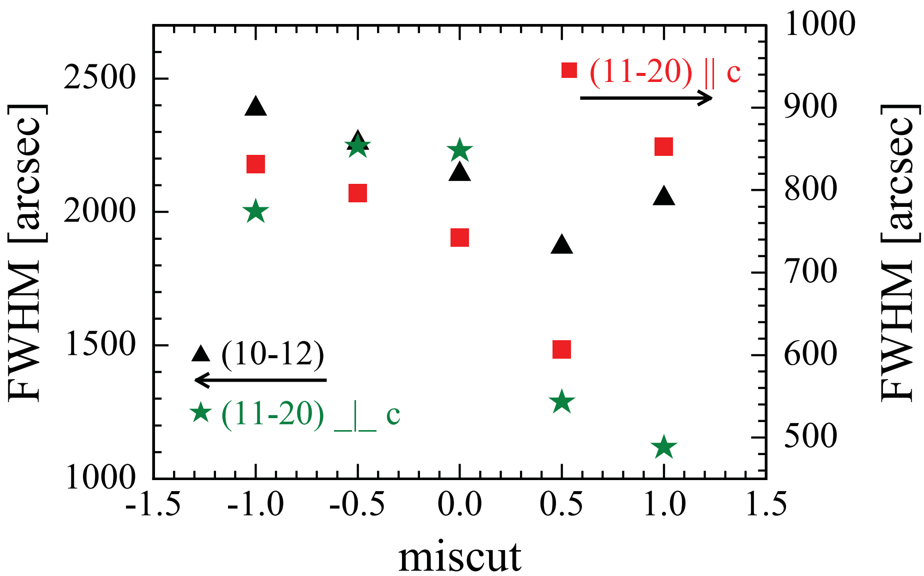

Fig. 4: FWHM of XRD rocking curves. The asymmetrical $(10\bar{1}2)$ reflection and the symmetrical $(11\bar{2}0)$ reflection with the incident beam perpendicular (parallel) to the c direction is plotted to the left (right). (color online).

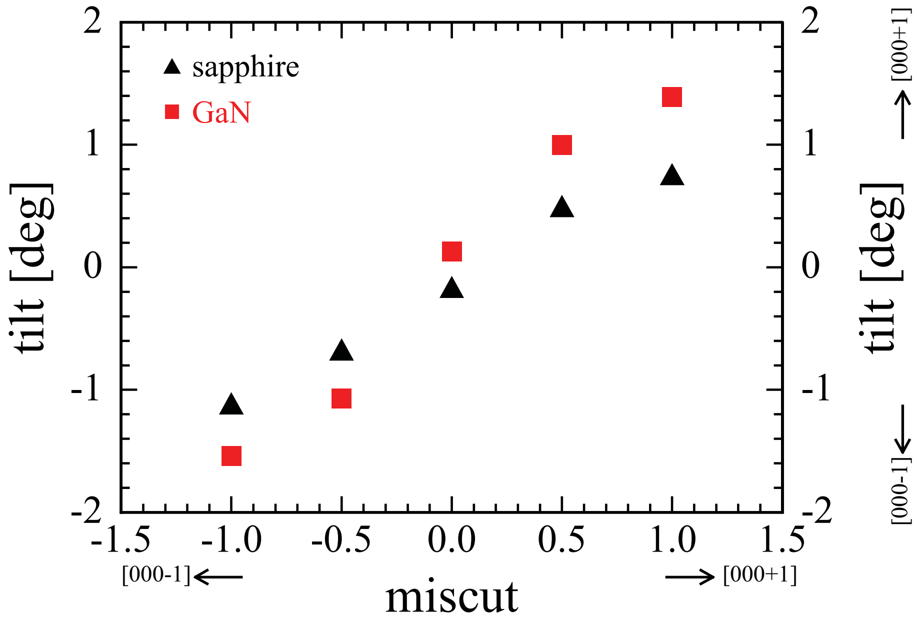

Fig. 5: Inclination of the HVPE grown GaN layers and the sapphire wafers, measured by XRD. The tilt of the sapphire (black triangles) and the GaN (red squares) is plotted against the (nominal) miscut of the sapphire substrate (color online).

too large, the — from MOVPE growth — well known surface defects start to develop so that the resulting surface is even rougher than for negative miscut angles (E). The pits theirselves are of increased size when compared to the MOVPE templates because of the increased thickness of the sample.

In order to judge the crystal quality of the samples, XRC measurements were performed (Fig. 4). Somehow as expected from the results of the MOVPE grown templates, the crystal quality for the samples with negative miscut is worse compared to layers grown on positive miscut substrates. But in contrast to those findings, the smallest FWHM values were not measured for the sample with the highest positive miscut (E) but for sample D with a slight miscut of +0.5°. Nevertheless this fits quite well to the results of the optical investigations of the HVPE grown samples. As the $(10\bar{1}2)$ reflection is known to be sensitive to edge and screw dislocations [15], it is often used to judge the crystal quality. The FWHM of this reflection is essentially smaller for the samples with a positive miscut. In particular sample D shows the strongest improvement and smallest values.

One question of course will rise if one grows on miscut substrates. How is the inclination between substrate and layer, between sapphire and GaN? To analyze this, again XRC measurements were recorded (Fig. 5). The measured miscuts of the sapphire wafers agree to the nominal values. For all miscuts, the overgrown GaN layer shows an inclination angle with respect to the surface that is larger than the miscut of the sapphire. The data points shown in figure 5 represent the direction of the normal of the reflection plane. This means that the tilt is measured with respect to the surface. So the inclination of the a-direction of GaN is even higher than and in the same direction as the tilt of the sapphire r-plane. Hence the tilting of the GaN layer is totally opposed to the miscut of the sapphire (with respect to the 'normal' r-plane surface).

4. Summary

We studied the influence of slightly misoriented r-plane sapphire substrates on the crystal quality and surface morphology of a-plane GaN grown by HVPE on MOVPE templates. We found strongly different miscuts for the best values of either of these two parameters. The situation got better after HVPE growth: Now, both, surface quality and crystal quality can be improved by a slight miscut of $+0.5°$ towards the [0001] direction. It was also shown that the GaN layers are grown tilted with respect to the sapphire wafers with an inclination totally opposed to that of the miscut sapphire substrate.

5. Acknowledgments

This work was financially supported by the Deutsche Forschungsgemeinschaft within the research group "Polarization Field Control in Nitride Light Emitters (PolarCoN)" under contract no. Scho 393/22-1.

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