Mask-less MOVPE of arrayed n-GaN nanowires on site- and polarity-controlled AlN/Si templates"

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Supporting information

Reflected interferometry on SAE islands

In this publication reflected interferometry data has been used to analyze and interpret the growth of GaN islands on an AlN/Si surface. Here, we explain possible causes for signal oscillation and signal decreasing during the GaN island and NW growth on AlN/Si 2D-layers. To simplify the model, we do not consider the Si pillars in the following analyzation but chose a flat AlN- and Si-layer. Furthermore, absorption effects are neglected. The measured reflected interferometer intensity is a superposition of reflected light from the GaN islands and from light reflected at the AlN/Si surface, where no GaN should grow. Thus, we have calculated the light path and the intensity change during its propagation through the crystals at different positions. Figure 1 schematically illustrates how the light propagates, if it hits the *r*-facet of a GaN island (a) and how it is reflected without (b) and with increasing parasitic growth (c and d) on the flat AlN/Si-template.

First, the incident light on a GaN island is discussed (Figure 1 a). We have calculated the incident (a), the exit and the reflected angle based on the fixed angular dependence of the facets and the Fresnel equations. E.g. the angle between the *m*-direction ($[11\bar{2}0]$) and the *r*-direction [($11\bar{2}1$]) is 61.5°, and thus, the incident angle a at the air/*r*-facet interface is also 61.5°. Using the refractive indexes of air (1), GaN (2.387), AlN (2.17) and Si (3.87) at the interferometer wavelength of 633 nm, the exit angles at each interface and the total reflection (R) and transmission (T = 1 - R) can be calculated. The results are summarized in Table 1.

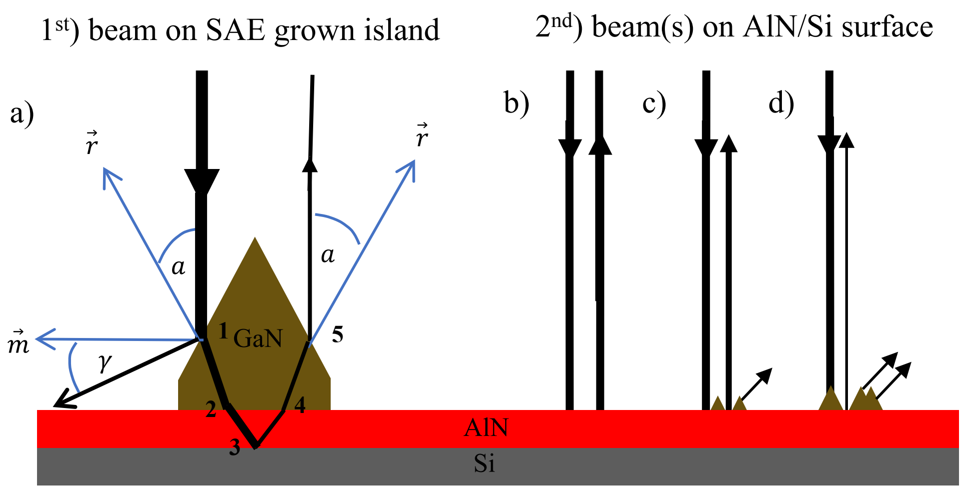

Figure 1. Path of the interferometer LASER light falling on a GaN island (a), on the AlN/Si surface without parasitic GaN growth (b) and with increasing parasitic growth (c and d). The numbers 1 to 5 are the interfaces, used for the calculation in Table 1. The thickness of the solid black lines qualitatively represents the light intensity of the beams.

Table *1:* exit angles and reflectance values at the interfaces

interface	#-interface	exit angle [°]	reflectance R[%]
air/GaN *r*-facet	1	21.6	21.4
GaN *r*-facet/AlN	2	44.87	0.4
AlN/Si	3	23.3	9.1
AlN/GaN	4	39.9	1.6
GaN *r*-facet/air	5	61.5	21

The calculation shows that the exit angle at the last interface (GaN r-facet/air) is again 61.5°, which leads to vertically leaving light. Due to the periodic pattern, light can also be reflected from one island to a neighboring island, depending on the pitch and the height of the islands or NWs. The reflection angle is fixed (γ = 33° in Figure 1 a). Our calculations show, that re-reflection on neighboring GaN NWs can occur by the in this work used pattern and island/NW-sizes (pitch = 2.5 µm, d_{NW} = 800 nm, h_{NW} = 820 nm). Interestingly, a part of the light intensity is also vertically reflected to the detector. All vertically back reflected light from the GaN islands is phase shifted compared to light, which is reflected from the AlN/Si-surface surrounding the GaN islands. This causes interference. By epitaxially growing the GaN islands, the optical path (and the phase) changes. The interferometer LASER spot is large compared to the pitch (2.5 µm) and the size of the growing GaN structures, thus, a high density of 123,000 islands/mm² are simultaneously illuminated. If the GaN growth is homogeneous the extension of the islands is similar, which leads to a superposition of similar reflected waves, causing an oscillating signal. At the same time, a large part of the signal intensity is not reflected to the reactor, as shown by the reflectance values R in Table 1. Thus, the total measured intensity is also decreasing during perfect homogeneous growth. Eventually, homogeneous growth leads to a simultaneously oscillating and decreasing signal, originating from the islands.

If additional parasitic GaN growth occurs on the AlN surface, as sketched in Figure 1 c and d, the total intensity of the reflected light strongly decreases. The effect is dominant, since the area, where no GaN growth is intended covers ≈ 95 % of the whole AlN/Si-surface. Consequently, the detection of homogeneous growth is prevented if strong parasitic growth occurs.

Additional SEM image of sample B5 (carrier gas) after KOH(aq.) etching

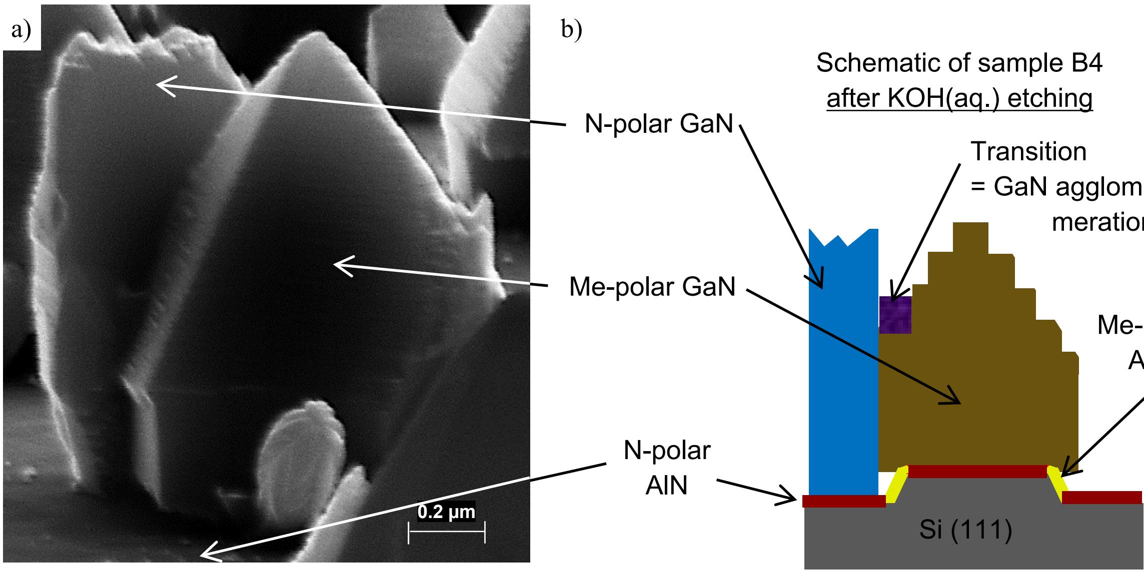

Figure 2. b) KOH (aq.) etched mixed-polar NW structure of sample B5: 80°-tilted SEM image and the corresponding sketch.

SEM images of samples B7, B8, B9 and B10, grown with a varied Si/Ga ratio

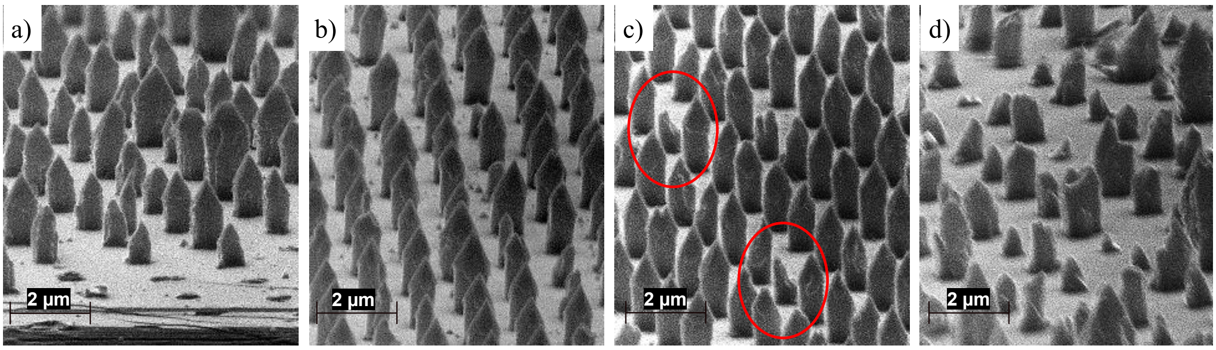

Figure 3. 80°-tilted SEM-images of samples grown at varied Si/Ga-ratios, increasing from left to right: a) B7 (Si/Ga-ratio = 2.6 ‰), b) B8 (Si/Ga-ratio = 4.1 ‰), c) B9 (Si/Ga-ratio = 5.2 ‰) and d) B10 (Si/Ga-ratio = 7.2 ‰). The Si/Ga-ratio variation was realized by adjusting Q_{TMGa} and keeping constant Q_{SiH4} (288 µmol/min) and Q_{NH3} (0.7 mmol/min).

Additional experimental results for Si/Ga ratio and time-dependent NH_3 flow

a) b)

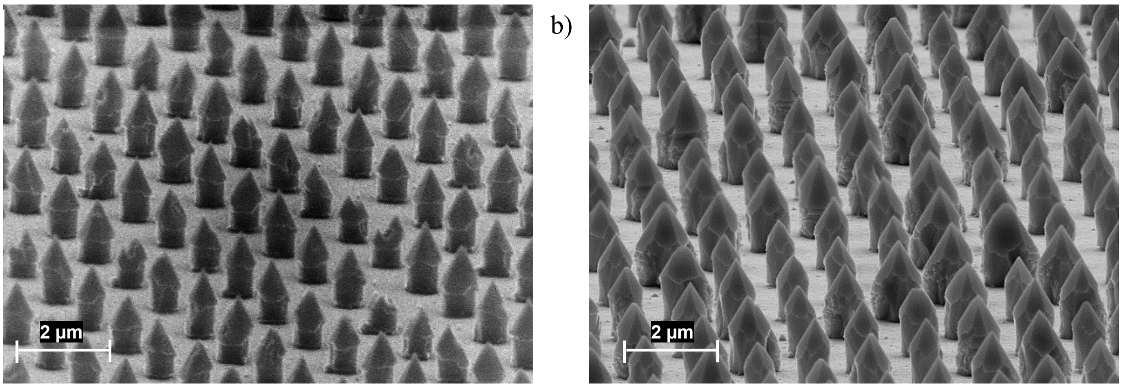

Figure 4. a) variation of Si/Ga-ratio by solely increasing SiH_4 flow to the system-maximum SiH_4 flow (372 nmol/min). The Q_{TMGa} (110 µmol/min) and Q_{NH3} (0.7 mmol/min) are constant, taking sample B7 as reference. b) NWs grown at two different Q_{NH3}: first 180s at 0.7 mmol/min followed by 360 s at 2.2 mmol/min. The Q_{TMGa} is constant. Both SEM-images are 80°-tilted.

Additional SEM images of sample B16 without and with outer shell

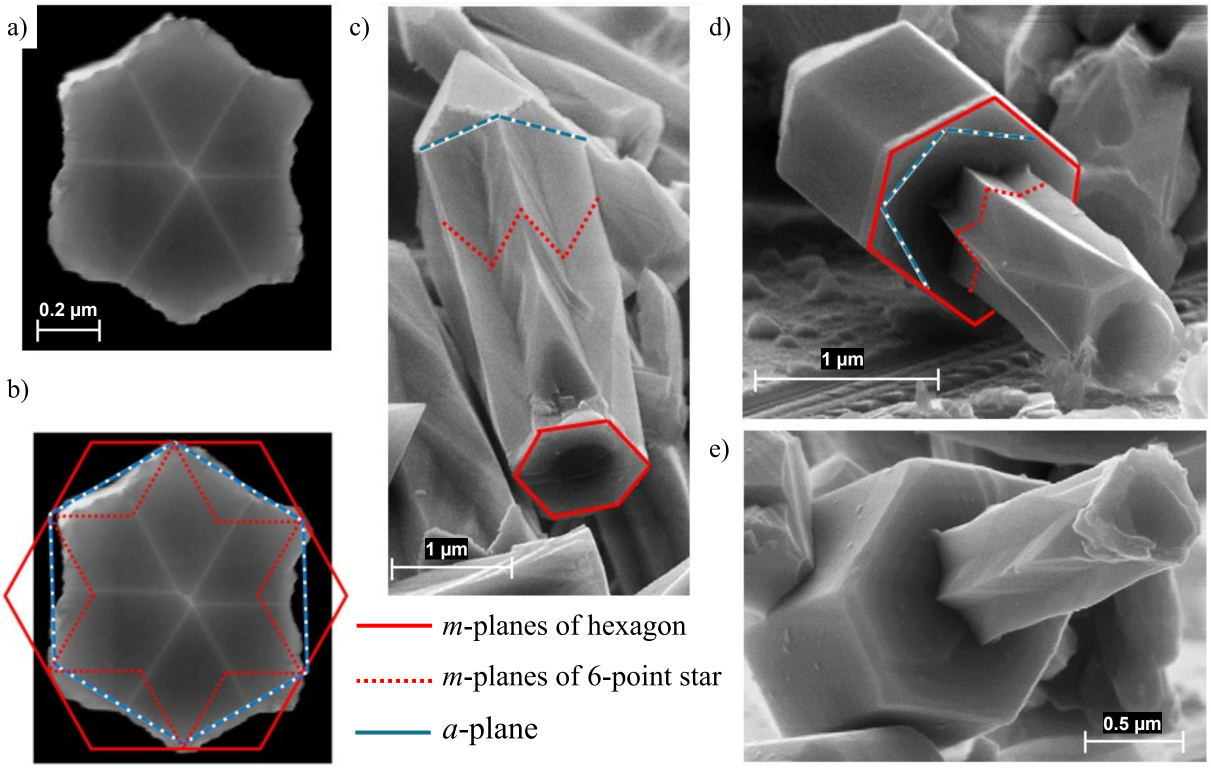

Figure 5. a) and b) top view image of a standing NW without and with superimposed marks to show the location of the different facets. c), d) and e) scratched NWs without and after GaN shell growth.

Figure 5 depicts SEM images of NWs from sample B16 from different perspectives with and without a shell. The images help to identify the outer core facets. Additionally, the *m*- and *a*-facets are illustrated on some NWs. By comparing the different possible facets, it is evident, that the core has no *a*-facets. In contrast, 12 *m*-planes are visible on the 6-point star cross section.