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The influence of MOVPE growth conditions on the shell of core-shell GaN microrod structures

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ABSTRACT

A core-shell geometry is employed for most next-generation, three-dimensional opto-electric devices based on III–V semiconductors and grown by metal organic vapor phase epitaxy (MOVPE). Controlling the shape of the shell layers is fundamental for device optimization, however no detailed analysis of the influence of growth conditions has been published to date.

We study homogeneous arrays of gallium nitride core-shell microrods with height and diameter in the micrometer range and grown in a two-step selective area MOVPE process. Changes in shell shape and homogeneity effected by deliberately altered shell growth conditions were accurately assessed by digital analysis of high-resolution scanning electron microscope images.

Most notably, two temperature regimes could be established, which show a significantly different behavior with regard to material distribution. Above 900 °C of wafer carrier temperature, the shell thickness along the growth axis of the rods was very homogeneous, however variations between vicinal rods increase. In contrast, below 830 °C the shell thickness is higher close to the microrod tip than at the base of the rods, while the lateral homogeneity between neighboring microrods is very uniform. This temperature effect could be either amplified or attenuated by changing the remaining growth parameters such as reactor pressure, structure distance, gallium precursor, carrier gas composition and dopant materials. Possible reasons for these findings are discussed with respect to GaN decomposition as well as the surface and gas phase diffusion of growth species, leading to an improved control of the functional layers in next-generation 3D V–III devices.

1. Introduction

In recent years, many publications have been released concerning the growth of three-dimensional structures. The applications are manifold – future three-dimensional LED structures [1,2] are envisioned as well as three-dimensional (3D) GaN transistors [3], sensors [4] and solar-cells [5]. The structures take the form of either pyramids [6], nano- or microrods as well as GaN fins [7], but also hybrid forms [8,9] have been demonstrated.

In the majority of structures grown by metal organic vapor phase epitaxy (MOVPE), an epitaxial core-shell geometry is employed for gaining an advantage over planar state-of-the art devices. In core-shell configuration, controlling the shape of the shell is fundamental for device optimization, for example in the InGaN quantum well of 3D LEDs, where growth rate inhomogeneity leads to wavelength gradients along the structures [10–12] due to the indium incorporation dependency on the local growth rate [13] and due to confinement effects in narrow quantum wells. In this publication, we show the result of extensive investigations of the influence of the major MOVPE growth parameters on the morphology of the microrod (MR) shell.

First the growth method for the core-shell MRs is given, then the analytic procedure for the shell layers will be described. The main part shows the specific effects of the investigated growth parameters in subsequent individual sections. Finally, the findings are summarized and a physical hypothesis is established.

2. Experimental

The growth templates for the microrods consist of n-doped GaN on 4″ sapphire wafers on which a 100 nm thick SiO_x mask layer was deposited by PECVD. The mask layer was uniformly structured

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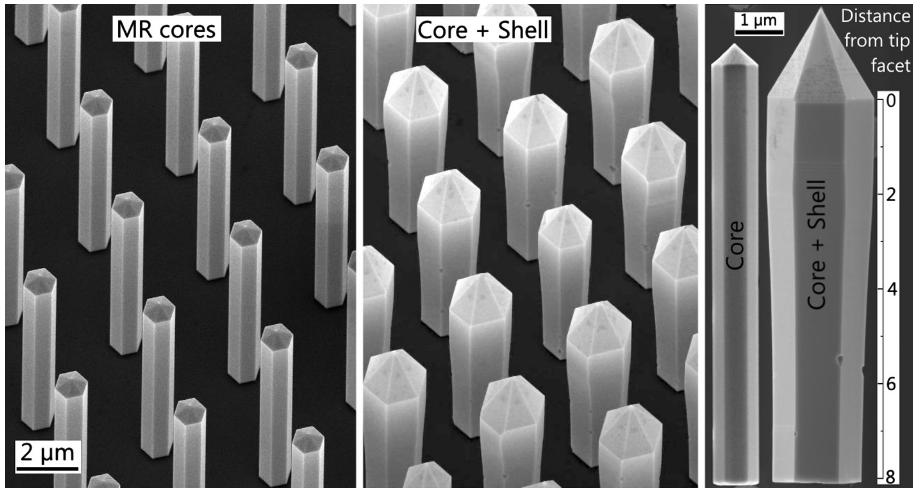

Fig. 1. Array of microrod cores (left) and core-shell structures (center). Right: Collage of lateral images of microrod core and core-shell structure.

with a hexagonal lattice of round 800 nm diameter openings with a 4 μm pitch using nanoimprint and dry etching. On these templates, the core-shell structures were grown in one step in a commercial large-volume MOVPE reactor equipped with in-situ temperature monitoring equipment. The MR cores were grown under conditions similar to those previously published [14], under very low V/III ratio and with a high silane flux. The cores form perfectly regular hexagonal columns, with high homogeneity in lateral direction and without any noticeable tapering along their height (Fig. 1, left). The pitch and large-axis diameter of the MR cores are 4 μm and 1050 nm, respectively, the height is approximately 10 μm. After core growth, conditions were switched to be beneficial for shell layer growth, namely high V/III ratio (≈20,000). An AlGaN layer with a thickness of a few nm was deposited to promote smooth shell layer growth on the silicon-rich surface of the highly doped cores [14], followed by a shell of ≈400 nm thickness (Fig. 1, center). Due to the lower growth rate of the semipolar $\{1\,\bar{1}\,0\,1\}$ tip facets of the shell, the resulting m-plane height decreases with the shell layer thickness in accordance to the Wulff-Curie law [15] (Fig. 1, right).

3. Method of analysis

Unless specified otherwise, the homogeneity of the shell layers along the growth direction was analyzed by the following procedure: Lateral images of several MRs were taken with an SEM at high resolution and the MR shape was digitized. The circumdiameter of the Core-Shell rod (r_{CS}) was evaluated along its height as the respective distance between the nonpolar sidewalls. Subtracting the previously measured circumdiameter of the MR core ($2r_{Core}$) and dividing by a factor of two yielded the layer thickness d on an edge of the MR along its entire length. However, the increase of the layer thickness on the side facet of the MR for a given amount of deposited material is proportional to the square of the total rod diameter.

Since the amount of deposited shell material needed to increase the layer thickness increases with the square of the rod radius, the layer thickness is not a good, linear indicator for the material deposition. We therefore calculate the cross-sectional area of the shell (A_{shell}) at each measured height, which scales linearly with the amount of deposited material.

$$A_{shell} = A_{CS} - A_{Core} = \tfrac{3}{2}\sqrt{3}(r_{CS}^2 - r_{Core}^2) = \tfrac{3}{2}\sqrt{3}\left((r_{Core}+d)^2 - r_{Core}^2\right) = \tfrac{3}{2}\sqrt{3}(d^2 + 2r_{Core}d)$$

As shown in Fig. 2, A_{CS} and A_{Core} denote the cross-sectional area of the core-shell microrod and of the core, respectively, r_{CS} and r_{Core} are the corresponding circumradii. For simplifying the formula it was used that $A_{CS} = A_{Core} + A_{Shell}$. Integrating over the cross-sectional area of the shell along the length of the rod yields the total volume of the shell deposited on the core. It is important to note that the cross-sectional area will be shown versus the distance from the tip facet, as shown in the scale in Fig. 1, right.

To account for different growth rates and precursor gases used in this study, A_{shell} is normalized with regard to the total amount of gallium precursor used, resulting in units given as nm^2/nmol. The pointwise averaged values for all investigated rods from one sample were then compared to those of samples obtained under different growth conditions.

For the analysis of the homogeneity of the MR shape cross section, high resolution SEM images were taken in top view geometry. These images contain several hundreds of individual rods, whose individual cross section areas and diameters were analyzed. To assess the homogeneity of the cross section area – corresponding to the uniformity of material distribution between individual MRs – the standard deviation of the cross section area σ_A is taken as the figure of merit. A large value of the standard deviation indicates large fluctuations in the material distribution and the rods are differing significantly in shell volume. Furthermore, the homogeneity of the core-shell rod shape was analyzed. While the MR

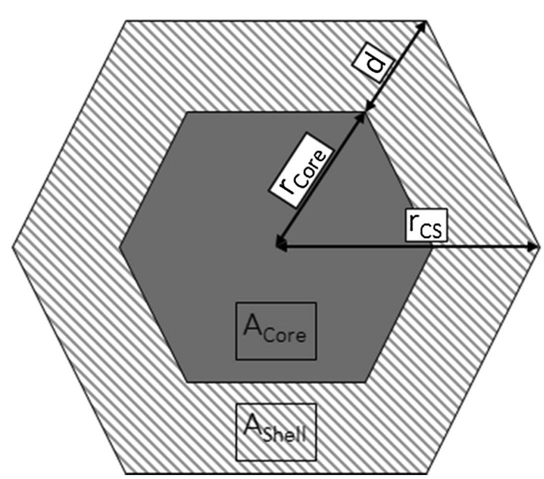

Fig. 2. Cross-sectional areas and diameters of a core-shell microrod.