Journal of Crystal Growth 364 (2013) 149–154

Contents lists available at SciVerse ScienceDirect

Journal of Crystal Growth

journal homepage: www.elsevier.com/locate/jcrysgro

Dependence of N-polar GaN rod morphology on growth parameters during selective area growth by MOVPE

Shunfeng Li [a,*], Xue Wang [a], Matin Sadat Mohajerani [a], Sönke Fündling [a], Milena Erenburg [a], Jiandong Wei [a], Hergo-Heinrich Wehmann [a], Andreas Waag [a], Martin Mandl [a,b], Werner Bergbauer [b], Martin Strassburg [b]

[a] Institut für Halbleitertechnik, TU Braunschweig, Hans-Sommer-Straße 66, 38106 Braunschweig, Germany
[b] Osram Opto Semiconductors GmbH, Leibnizstr. 4, 93055 Regensburg, Germany

ARTICLE INFO

Article history:
Received 21 May 2012
Received in revised form
19 October 2012
Accepted 12 November 2012
Communicated by J.M. Redwing
Available online 19 November 2012

Keywords:
A1. Crystal morphology
A1. Growth models
A3. Metalorganic chemical vapor deposition
B1. Nitrides
B1. Microcolumn
B2. Semiconductor gallium compounds

ABSTRACT

Selective area growth of GaN rods by metalorganic vapor phase epitaxy has attracted great interest due to its novel applications in optoelectronic and photonics. In this work, we will present the dependence of GaN rod morphology on various growth parameters i.e. growth temperature, H_2/N_2 carrier gas concentration, V/III ratio, total carrier gas flow and reactor pressure. It is found that higher growth temperature helps to increase the aspect ratio of the rods, but reduces the height homogeneity. Furthermore, H_2/N_2 carrier gas concentration is found to be a critical factor to obtain vertical rod growth. Pure nitrogen carrier gas leads to irregular growth of GaN structure, while an increase of hydrogen carrier gas results in vertical GaN rod growth. Higher hydrogen carrier gas concentration also reduces the diameter and enhances the aspect of the GaN rods. Besides, increase of V/III ratio causes reduction of the aspect ratio of N-polar GaN rods, which could be explained by the relatively lower growth rate on (000-1) N-polar top surface when supplying more ammonia. In addition, an increase of the total carrier gas flow leads to a decrease in the diameter and the average volume of GaN rods. These phenomena are tentatively explained by the change of partial pressure of the source materials and boundary layer thickness in the reactor. Finally, it is shown that the average volume of the N-polar GaN rods keeps a similar value for a reactor pressure P_R of 66 and 125 mbar, while an incomplete filling of the pattern opening is observed with P_R of 250 mbar. Room temperature photoluminescence spectrum of the rods is also briefly discussed.

1. Introduction

The growth of GaN nano (or sub-μ) rods is a rapidly expanding field driven in part by the unique functionality of nanorods and their high crystalline quality [1]. The small diameter of the nanorod can induce additional relaxation of a high degree of strain in lateral dimension, which is often generated by the large lattice and/or thermal mismatch between the GaN and substrates. This leads to a reduction of dislocation density and allows GaN nanorod growth on large area substrates without inducing wafer bowing or cracks, which are always difficult issues for large area GaN growth [2]. Furthermore, the dislocations generated at the nanorod/substrate interface can bend to the sidewall of the GaN nanorods, especially when the aspect ratio of nanorods is high, which effectively reduces the dislocation density in the top part of the GaN nanorods [2–4]. Besides, the 3D GaN nanorod geometry might help to enhance the light extraction in a nanorod based LEDs. It has been demonstrated that by varying the nanorod diameter multi-color emission can be obtained, which is promising to realize low-cost monolithic white light emission by RGB color mixing [5]. Recently, core-shell nanoLED structure is suggested to be able to increase the area of the active region grown on the sidewall of the nanorods via increasing the aspect ratio of nanorods [6]. In this case, the InGaN/GaN MQWs active region is grown on non-polar m-planes, in which the polarization fields are absent. In addition, optical polarization effects have to be taken into account, which have recently been discussed by Scheibenzuber et al. [7]. Overall, both the emission wavelength stability and the efficiency will be improved [8].

Up to now, the self-assembled growth and characterization of GaN nanorods has been extensively explored. Light emitting devices have been realized based on the self-assembled GaN nanorods [9]. However, the statistical distribution of the rod geometry during self-assembled GaN nanorod growth leads to a broad distribution of GaN nanorod properties, such as QW

* Correspondence to: Institute of Semiconductor Technology, Braunschweig University of Technology, Hans-Sommer-Strasse 66, 38106 Braunschweig, Germany. Tel.: +49 531 3913806; fax: +49 531 3915844.
E-mail address: sf.li@tu-bs.de (S. Li).

0022-0248/$ - see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jcrysgro.2012.11.027

thickness, strain and alloy composition of the QWs within the nanorods. For a better control of those properties, which then determine the emission properties, well controlled selective area growth (SAG) of GaN nanorods on patterned substrates, e.g. patterned SiO_2/GaN [10] or Ti/GaN [11,12], was proposed. However, the development of selective area growth of GaN nanorods by metalorganic vapor phase epitaxy (MOVPE), which is a favorable tool for industrial application, has been hampered in the past due to the difficulties in achieving rod growth with vertical sidewalls by MOVPE [10,13], in contrast to MBE. Pulsed growth mode was reported to be able to realize the GaN nanorod growth by selective area MOVPE growth [10].

Polarity is an inherent property of the wurtzite GaN lattice, which is known to determine a large number of properties such as surface reactivity and thermal stability [14,15]. Recently, we found that the polarity is also critical in determining the morphology of the GaN rods, i.e. N-polar GaN rod growth is 'easier' than the growth of Ga-polar GaN rods [16,17]. The growth mechanism of N-polar GaN rods in MOVPE growth was also discussed [16]. Similar experimental results are also reported by other research group [18]. Based on this work, we reported N-polar GaN core-shell LED structure in recent publication [19].

In this work, we will present the results of a thorough investigation of the selective area growth of N-polar GaN rods in a sub-micrometer size by MOVPE. The dependence of the N-polar GaN rods on growth parameters, including growth temperature, H_2/N_2 carrier gas concentration, V/III ratio, the reactor pressure and total carrier gas flow will be presented and discussed in detail. The photoluminescence properties of our N-polar GaN rod ensemble are also briefly introduced.

2. Experimental details

The growth templates used in this work were patterned SiO_2/sapphire substrates. As the first step, a SiO_2 passivation layer was deposited on sapphire substrates by plasma-enhanced chemical vapor deposition with 30 nm thickness. Subsequently, the pattern was transferred onto the SiO_2 covered substrates by photolithography, followed by inductively-coupled plasma etching to open the holes in the SiO_2 mask layer. The patterns were composed by hexagonally shaped openings with different diameters and inter-hole distances. The schematic drawing of the complete template fabrication and growth processes is shown in Fig. 1.

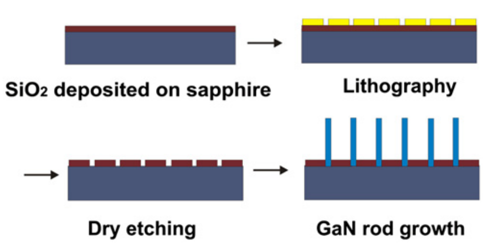

Fig. 1. Schematics of the fabrication process of GaN rod sample by selective area growth.

For the subsequent GaN rod growth, we use a vertical 3×2 Thomas Swan MOVPE system. After chemical cleaning, the growth templates were loaded into the reactor for growth. Prior to the growth a nitridation step was employed. After that, TMGa flow was introduced into the reactor to start the GaN rod growth. The growth temperatures were kept at about 1080 °C. The V/III ratio was 100 during growth, if not otherwise mentioned. Other detailed information can be found in our previous publication [16]. The GaN rods were doped during growth with the same SiH_4 flow rate used for growth of a GaN layer with Si concentration of about 1×10^{18} cm^{-3}. This low SiH_4 flow did not have a distinguishable influence the GaN morphology, which is different from the Koester et al. work [20]. The growth time was kept at 1200 s in all samples.

The morphology of GaN rod samples was characterized by a Zeiss supra 35 field emission scanning electron microscope (FESEM) with an acceleration voltage of 2 kV. The room temperature photoluminescence (PL) spectra were measured by a 325 nm He-Cd laser with excitation power density of $\sim 2 \times 10^4$ W cm^{-2}.

3. Results and discussion

3.1. Growth temperature influence

The growth temperature is considered to be a critical factor in the patterned GaN rod growth. In order to check the effect of growth temperature we performed GaN rod growth at 1030 °C, 1070 °C and 1100 °C, respectively. The FESEM images of these three samples are presented in Fig. 2. These images are taken from a region with opening diameter of about 1 μm and pitch of about 5.9 μm.

As seen in Fig. 2a, the low temperature (1030 °C) GaN rods show six {1–100} vertical facets, with an average aspect ratio of about 1. A clear inhomogeneity in diameter can be observed. Higher temperature (1070 °C) rod growth leads to a higher aspect ratio of the GaN rods, i.e. the diameter decreased and their height increased (Fig. 2b). The average aspect ratio is about 2. When the growth temperature is further increased to 1100 °C, a further increase of the aspect ratio is observed. However, in this case the height homogeneity of the GaN rod array gets worse, i.e. some rods are much higher than the others. Based on our recent results [16], the high temperature may enhance the surface diffusion and reduce the rod diameter by increased hydrogen etching, especially at lateral direction, causing the increase of the height and aspect ratio of the rods. On the other hand, the reduced height homogeneity indicates that the material which is available for growth could be transported and contributed more to the growth

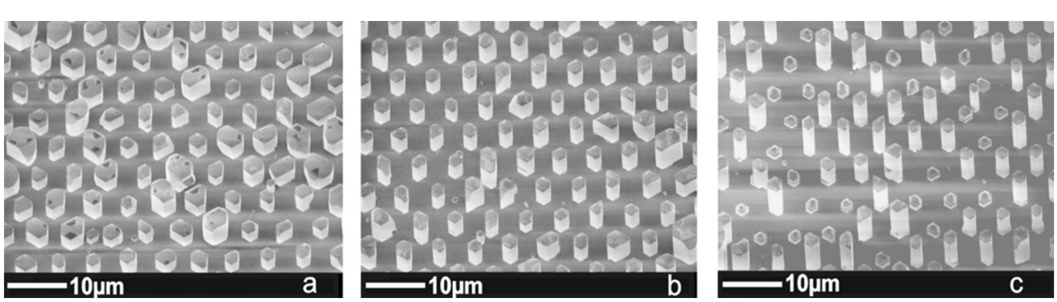

Fig. 2. FE-SEM images (bird view, 30°) of GaN rods grown at different temperatures: (a) 1030 °C, (b) 1070 °C, and (c) 1100 °C. The diameter of the opening is about 1 μm and the distance between the openings is 5 μm.