

## Morphology of the surface of semipolar GaN layers during epitaxy on a nano-patterned Si substrate

© V.N. Bessolov,<sup>1</sup> E.V. Konenkova,<sup>1</sup> T.A. Orlova,<sup>1</sup> S.N. Rodin,<sup>1</sup> A.V. Solomnikova<sup>2</sup>

<sup>1</sup> Ioffe Institute,  
194021 St. Petersburg, Russia

<sup>2</sup> St. Petersburg Electrotechnical University LETI,  
197022 St. Petersburg, Russia  
e-mail: lena@triat.ioffe.ru

Received January 12, 2022

Revised February 21, 2022

Accepted February 21, 2022

It has been studied the morphology of the surface of the semipolar gallium nitride layers synthesized on nano-patterned Si(100) or Si(113) substrates with a V-shaped or U-shaped surface profile, respectively. The morphology of the surface of the semipolar layers indicates that the different height-to-width ratio of the GaN(11-22) and GaN(10-11) blocks is associated with a higher growth rate of the GaN(11-22) face than GaN(10-11) and with different growth rates of the semipolar and polar crystal faces during the nucleation of the layer on a nano-patterned substrate.

**Keywords:** surface morphology, semipolar gallium nitride, nano-patterned silicon substrate.

DOI: 10.21883/TP.2022.05.53677.12-22

### Introduction

Semipolar and nonpolar GaN layers attract attention of researchers, starting from the paper, where authors reported increased intensity of photoluminescence in light diode InGaN/GaN-structures free of piezoelectric polarization [1]. Use of microstructured allogenic substrates to synthesize semipolar GaN layers is a promising method of heteroepitaxy [2,3]. Thus, for example, a structured sapphire substrate with a plane (22-43) made it possible to produce light diodes InGaN(20-21)/GaN with radiation wavelength 490 nm [4], and structured substrate *m*-Al<sub>2</sub>O<sub>3</sub> made it possible to synthesize light diodes with green emission color InGaN(11-22) [5].

For the last two decades, considerable efforts were directed at producing III-nitride semiconductors based on silicon (Si) for the purposes of optical communication and integration of nitride-gallium and silicon electronics. The first light diode based on GaN, grown on Si [6], was demonstrated already in 1998. Currently optoelectronic devices based on GaN/Si are mostly accommodated on polar InGaN/GaN-structures, where active areas contain undesirable strong polarization field. Growth of semipolar III-nitride emitters is one of the possible solutions to this problem, since it is expected that semipolar and nonpolar structures have high potential in increasing internal quantum efficiency of light diodes [7], and in most efficient introduction of indium atoms, especially in GaN(11-22) layers [8].

Currently attempts are undertaken to synthesize semipolar gallium and aluminum nitrides on micro- [3] and nano-patterned Si(100) [9,10] substrates, where it is proposed to use for synthesis an inclined Si(111) face.

Semipolar GaN(11-22) on silicon may be produced mainly from growth on structured silicon Si(113)-substrates with striped grooves [11].

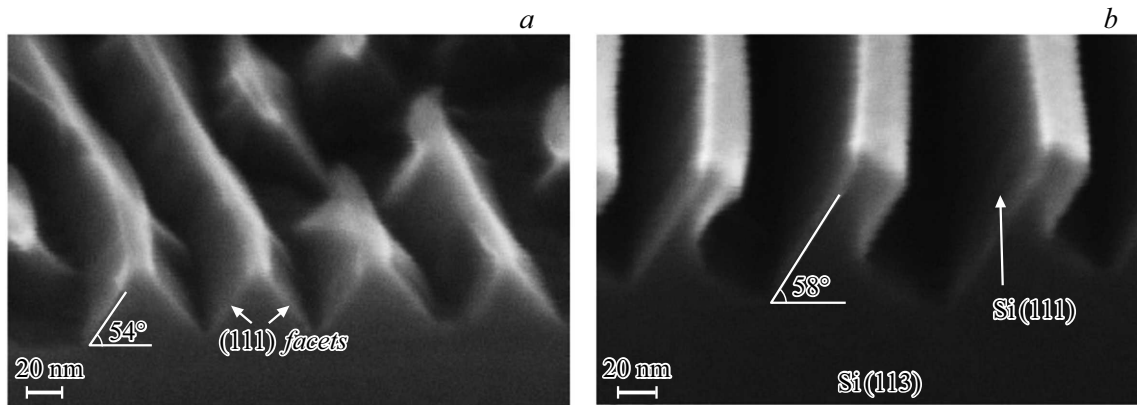
This paper is dedicated to study of morphology of the surface of semipolar GaN(10-11) and GaN(11-22) layers synthesized on nano-patterned — NP-Si(100) and NP-Si(113) substrates. To meet the set objective, a surface V-shaped (fig. 1, *a*) or U-shaped (fig. 1, *b*) structure was formed with a period of 75 nm, with height of inclined nanoridges 75 nm.

### 1. Experiment

A nanomask was formed in a two-stage process specified in [12]. AlN and GaN layers on NP-Si(100)- and NP-Si(113)-substrates were grown by the method of gas phase epitaxy from metal organic compounds (MOCVD) on a modified unit EpiQuip with a horizontal reactor similarly to [10]. Hydrogen was used as a carrier gas, and ammonia, trimethylgallium and trimethylaluminium as precursors. Structures consisted of a buffer AlN layer with thickness of ~ 35 nm and unalloyed GaN layer with thickness of ~ 1 μm.

Measurements of specimen surface morphology were carried out using atomic force microscopy in semicontact mode of measurements on a scanning probe microscope SolverNEXT.

In process of a technological experiment, semipolar GaN(11-22) layers on NP-Si(113) and GaN(10-11) layers on NP-Si(100) were grown under the same temperature and time conditions, and polar GaN(0001) layer on a flat Si(111) substrate for comparison.



**Figure 1.** REM image of nano-patterned substrates: NP-Si(100) — *a*, NP-Si(113) — *b*.

## 2. Results

X-ray diffraction demonstrated that GaN layers have half-width of X-ray diffraction curve  $\omega_\theta \sim 30'$  for structures GaN(11-22)/NP-Si(113) and  $\omega_\theta \sim 30'$  for GaN(10-11)/NP-Si(100), and  $\omega_\theta \sim 22'$  for GaN(0002)/Si(111).

Atomic force microscopy (AFM) of layer surface demonstrated significant difference in morphology of GaN(0001) layer synthesized on a flat surface of Si(111) substrate, and GaN(10-11) and GaN(11-22) layers synthesized on nano-patterned NP-Si(100) and NP-Si(113) substrates, accordingly. Surface of GaN(0001) layer contained defects with depth of around 50 nm, but heterogeneity of the layer on a site  $40 \times 40 \mu\text{m}$  made 20 nm max. (fig. 2, *a, b*). Surface of a semipolar GaN(10-11) layer had a marked asymmetric nature inherent in blocks of semipolar gallium nitride, which arise because of asymmetric properties of NP-Si(100) substrate after bombardment by ions  $\text{N}_2$ , as specified [10]. Value of surface heterogeneity in such structures in the surface area  $30 \times 30 \mu\text{m}$  was around 150–250 nm (fig. 2, *c, d*). It should be noted that when scanned in a direction perpendicular to the grooves, the distance between the peaks of humps in GaN(10-11) layer was equal to 3–5  $\mu\text{m}$  and considerably differs from similar distance set by NP-Si(100) substrate— 75 nm (fig. 1, *a*, fig. 2, *c, d*). Surface of semipolar GaN(11-22) layer under similar scanning in surface area  $50 \times 50 \mu\text{m}$  had the nature of rectangular blocks with size of  $4 \times 10 \mu\text{m}$ , where minima are observed of up to 1.3  $\mu\text{m}$ . Distance between humps in GaN(11-22) layer according to AFM data was around 10  $\mu\text{m}$  (fig. 2, *e, f*). Therefore, aspect ratio (height-to-width ratio) of semipolar gallium nitride blocks is 0.04 for GaN(10-11) and 0.13 for Ga(11-22) (fig. 2).

## 3. Discussion

Formation of GaN layers on NP-Si(100)- and NP-Si(113)-substrates takes place according to island mechanism on an open face of Si(111) nanogrooves, besides, orientation

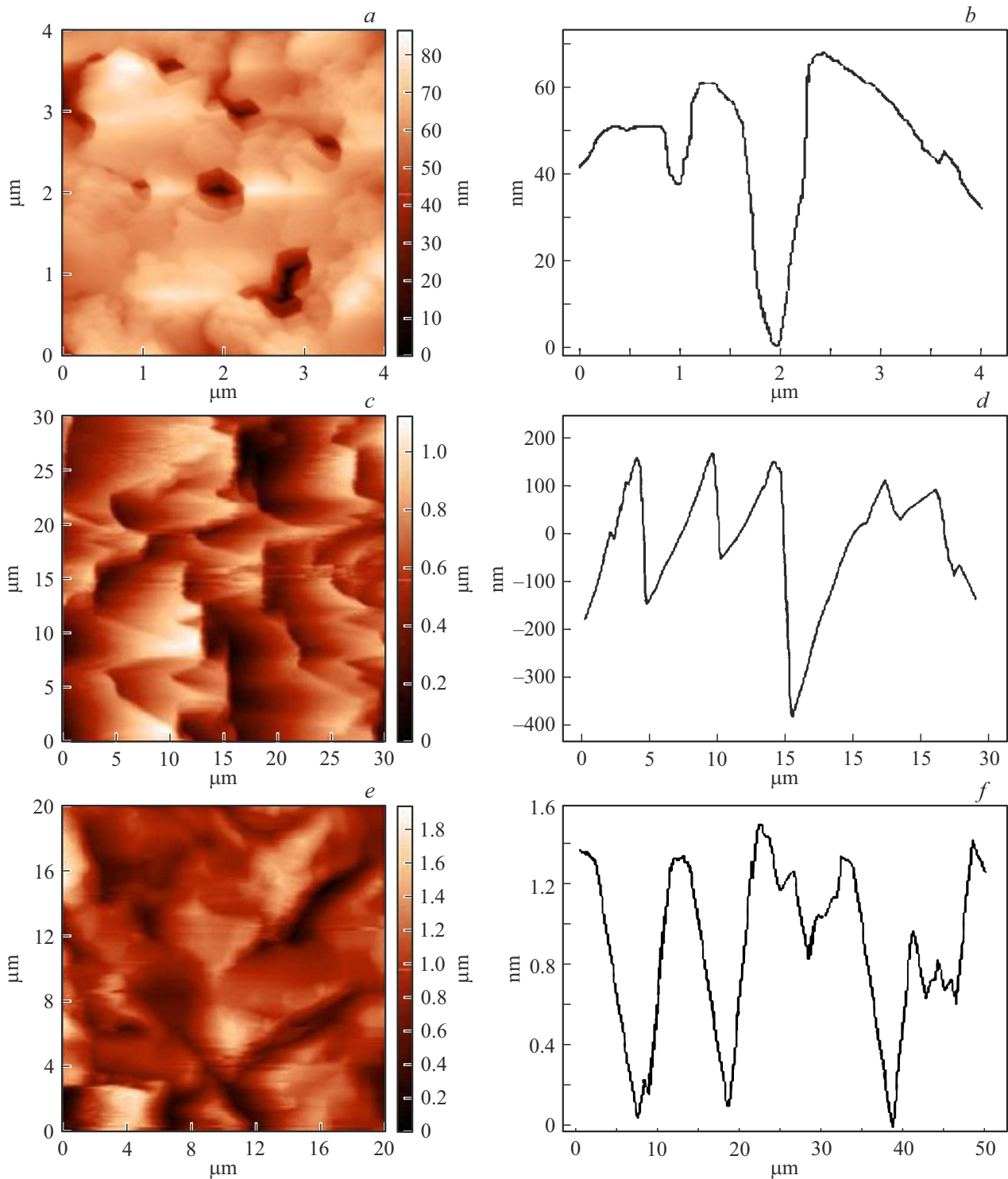
of nanocrystallites in an array is set by direction of plane Si(111) in NP-Si(113) and NP-Si(100).

In synthesis of gallium nitride, evolution of morphology will depend on the speed of layer growth depending on orientation of crystal surface faces. As is commonly known, for face (111) any atom adsorbed on this surface will be irreversibly built into the crystal structure. For face (111) the surface growth takes place according to random addition mechanism, and none of the adsorbed atoms is released back into vapor phase [13]. Surfaces with high growth speeds often disappear fully, i.e. only surfaces with least surface energies will remain in a thermodynamically balanced crystal. And this should result in formation of semipolar GaN(10-11) and GaN(11-22) planes on the surface, if grooves with Si(111) planes are used.

Previously it was noted that in selective gas phase epitaxy growth of GaN(0001) from metal-organic compounds on faces of GaN mesastripes previously formed by local epitaxy in windows  $\text{Si}_3\text{N}_4$ , if hydrogen is used as carrier gas, occurs mostly in lateral direction. According to experimental results [14], increase in growth temperature results in increased height and decreased diameter in epitaxy in the hydrogen atmosphere, i.e. aspect ratio increases. On the contrary, aspect ratio decreases with temperature growth under conditions of nitrogen as carrier gas [15].

In process of MOCVD you can identify two mechanisms for mass transfer on the surface of the nano-patterned substrate: diffusion in a vapor phase and surface diffusion. Molecules are included into epitaxial structure with the same probability in the entire surface area of inclined Si(111) face of nano-patterned NP-Si(100) and NP-Si(113) substrates, if effective length of diffusion exceeds half of the face length. Otherwise, uneven accumulation of epitaxial material will be observed on face tops.

Effective length of diffusion first of all depends on concentration of precursors of group III, since group V provides minor effect at growth speed. It is known that free path of Al adatom on AlN surface is short and is about 40 nm [16]. Free path of Ga adatom on GaN surface is different for polar (0001) and semipolar (10-11) faces of



**Figure 2.** AFM image and profile of structure surface: *a, b* — GaN(0001)/Si(111); *c, d* — GaN(10-11)/NP-Si(100), *e, f* — GaN(11-22)/NP-Si(113).

GaN during MOCVD epitaxy and is equal to 535 and 1430 nm, respectively [17]. However, there is data that free path of Ga on GaN surface at 1040°C is considerably longer and may be 15 μm in epitaxy in hydrogen atmosphere [18]. Therefore, epitaxial growth of AlN buffer layer on NP-Si(100) and NP-Si(113) occurs under the conditions, when

diffusion length of Al adatom is comparable to size of groove Si(111) face, and this provides for even growth along Si(111) faces. In hydrogen environment diffusion length of Ga adatom is much longer than distance between grooves of nano-patterned substrates, and this results in formation of gallium nitride blocks with large sizes on the substrate.

Indeed, from AFM data (fig. 2, *d,f*) one can assume that sizes of blocks formed on NP-Si(100) and NP-Si(113) surface, are close, which may be related to approximately the same length of Ga atoms diffusion in a near-surface layer equal to 4–10  $\mu\text{m}$ . Since GaN(10-11) growth speed is substantially slower than GaN(11-22) growth speed, this results in a different value of aspect ratio for semipolar layers.

Therefore, morphology of semipolar layer surface indicates that higher aspect ratio of the GaN(11-22) and GaN(10-11) blocks is associated with a higher growth rate of the semipolar GAN(11-22) face than GaN(10-11) and with different growth rates of the semipolar and polar faces. In general these results demonstrate that morphology of semipolar layers in epitaxy on a nano-patterned surface is formed under the conditions of competition between polar and semipolar planes of the crystal and differs significantly from the conditions of growth on flat polar surfaces.

### 3.1. Acknowledgments

Authors would like to thank V.K. Smirnov for provision of nano-patterned Si(100) substrates.

### 3.2. Funding

The studies were partially funded by RFBR within the framework of scientific project № 20-08-00096.

### Conflict of interest

The authors declare that they have no conflict of interest.

## References

- [1] P. Waltereit, O. Brandt, A. Trampert, H.T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche, K.H. Ploog. *Nature*, **406**, 865 (2000). DOI: 10.1038/35022529
- [2] A. Hirai, Z. Jia, M. Schmidt, R. Farrell, S. DenBaars, S. Nakamura, J. Speck, K. Fujito. *Appl. Phys. Lett.*, **91**, 191906 (2007). DOI: 10.1063/1.2802570
- [3] Y. Honda, N. Kameshiro, M. Yamaguchi, N. Sawaki. *J. Cryst. Growth*, **242**, 82 (2002). DOI: 10.1016/S0022-0248(02)01353-2
- [4] J. Song, J. Choi, K. Xiong, Y. Xie, J.J. Cha, J. Han. *ACS Appl. Mater. Interfaces*, **9** (16), 14088 (2017). DOI: 10.1021/acsami.7b01336
- [5] J. Hagggar, Y. Cai, S.S. Ghataora, R.M. Smith, J. Bai, T. Wang. *ACS Appl. Electron. Mater.*, **2**, 2363 (2020). DOI: 10.1021/acsaem.0c00399
- [6] S. Guha, N.A. Bojarczuk. *Appl. Phys. Lett.*, **73**, 1487 (1998). DOI: 10.1063/1.122181
- [7] Y. Zhao, S.-H. Oh, F. Wu, Y. Kawaguchi, S. Tanaka, K. Fujito, J.S. Speck, S.P. DenBaars, S. Nakamura. *Appl. Phys. Express*, **6**, 062102 (2013). DOI: 10.7567/APEX.6.062102
- [8] Y. Zhao, Q. Yan, C.-Y. Huang, S.-C. Huang, P.S. Hsu, S. Tanaka, C.-C. Pan, Y. Kawaguchi, K. Fujito, C.G. Van de Walle, J.S. Speck, S.P. DenBaars, S. Nakamura, D. Feezell. *Appl. Phys. Lett.*, **100**, 201108 (2012). DOI: 10.1063/1.4719100
- [9] V. Bessolov, A. Zubkova, E. Konenkova, S. Konenkov, S. Kukushkin, T. Orlova, S. Rodin, V. Rubets, D. Kibalov, V. Smirnov. *Phys. Stat. Sol. B*, **256**, 1800268 (2019). DOI: 10.1002/PSSB.201800268
- [10] V.N. Bessolov, E.V. Konenkova, S.N. Rodin, D.S. Kibalov, V.K. Smirnov. *Semiconductors*, **55** (4), 471 (2021). DOI: 10.1134/S1063782621040035
- [11] H. Li, H. Zhang, J. Song, P. Li, Sh. Nakamura, S.P. DenBaars. *Appl. Phys. Rev.*, **7**, 041318 (2020). DOI: 10.1063/5.0024236
- [12] V.K. Smirnov, D.S. Kibalov, O.M. Orlov, V.V. Graboshnikov. *Nanotechnology*, **14**, 709 (2003). DOI: 10.1088/0957-4484/14/7/304
- [13] P. Hartman, W.G. Perdok. *Acta Cryst.*, **8** (9), 521 (1955). DOI: 10.1107/S0365110X55001679
- [14] B.-O. Jung, S.-Y. Bae, Y. Kato, M. Imura, D.-S. Lee, Y. Honda, H. Amano. *Cryst. Eng. Comm.*, **16**, 2273 (2014). DOI: 10.1039/C3CE42266F
- [15] M. Nami, R. Eller, S. Okur, A. Rishinaramangalam, S. Liu, I. Brener, D. Feezell. *Nanotechnology*, **28**, 025202 (2017). DOI: 10.1088/0957-4484/28/2/025202
- [16] C. Bayram, J.A. Ott, K.-T. Shiu, Ch.-W. Cheng, Y. Zhu, J. Kim, M. Razeghi, D.K. Sadana. *Adv. Funct. Mater.*, **24** (28), 4492 (2014). DOI: 10.1002/adfm.201304062
- [17] T. Narita, T. Hikosaka, Y. Honda, M. Yamaguchi, N. Sawaki. *Phys. Stat. Sol. C*, **0** (7), 2154 (2003). DOI: 10.1002/pssc.200303511
- [18] M. Stepniak, M. Wóska, J. Prązmowska-Czajka, A. Stafiniak, D. Przybylski, R. Paszkiewicz. *Electronics*, **9**, 2129 (2020). DOI: 10.3390/electronics9122129