Selective-area growth and optical properties of GaN nanowires on patterned SiO\textsubscript{x}/Si substrates

V O Gridchin\textsuperscript{1,2}, R R Reznik\textsuperscript{3}, K P Kotlyar\textsuperscript{1,2}, A S Dragunova\textsuperscript{4}, L N Dvoretckaia\textsuperscript{1}, A V Parfeneva\textsuperscript{5}, D S Shevchuk\textsuperscript{1}, N V Kryzhanovskaya\textsuperscript{4}, I S Mukhin\textsuperscript{1,2} and G E Cirlin\textsuperscript{1,6}

\textsuperscript{1}Alferov University, Saint-Petersburg, 194021, Russia
\textsuperscript{2}Saint-Petersburg State University, Saint-Petersburg, 198504, Russia
\textsuperscript{3}ITMO University, Saint-Petersburg, 197101, Russia
\textsuperscript{4}HSE University, Saint-Petersburg, 194100, Russia
\textsuperscript{5}Ioffe Institute, Saint-Petersburg, 194021, Russia
\textsuperscript{6}Institute for Analytical Instrumentation RAS, Saint-Petersburg, 190103, Russia

E-mail: gridchinvoyandex.ru

Abstract. We present the results of selective-area growth of GaN nanowires by molecular beam epitaxy on patterned SiO\textsubscript{x}/Si substrates without using seed layers. The morphological and optical properties of selectively grown GaN nanowires are compared to the properties of GaN nanowires grown on the amorphous SiO\textsubscript{x} layer. The experimental results show that the selectivity of GaN nanowires is achieved at a substrate temperature of 825 °C which is the lower limit for the selective-area growth of GaN nanowires on SiO\textsubscript{x}/Si substrates. The study of the photoluminescence spectra of the grown nanowires, measured at 77 K show an emission line at 3.47 eV, which corresponds to strain-free GaN.

1. Introduction

Nowadays, the research in nano-optoelectronics is partially focused on integrating III-N semiconductor materials with silicon technology. III-N semiconductor materials (InN, GaN, AlN) and their alloys are of great interest for optoelectronics due to their ability to cover a wide bandgap range from 0.7 to 6.2 eV [1]. One of the approaches to integrate III-N semiconductor materials with silicon is the synthesis of nanowires (NWs). Due to their ability to grow in a practically defect-free crystal structure on lattice-mismatched substrates, NWs can be used as a perfect platform for embedding 0D, 1D, and 2D quantum structures and creating new functional nano-optoelectronic devices on silicon substrates [2, 3]. For example, a single-photon source based on a quantum-dot-in-nanowire structure was demonstrated recently [4]. However, III-N NWs often grow with a very high density up to the formation of a planar structure [5], which significantly limits their application and makes it difficult to control the nanowire sizes. To overcome these challenges, various approaches such as selective-area growth (SAG) of nanowires are used, in which the substrate surface is generally patterned using electron-beam lithography. Usually, at the preliminary stage of the SAG of GaN NWs, an AlN seed layer is formed on the substrate surface, and then the surface is covered with an inhibitor layer (for example, SiO\textsubscript{x}, or Ti) [6, 7]. However, the AlN seed layer can negatively affect the transport properties of a GaN/Si heterojunction [8]. The classical approach to substrate surface patterning has the disadvantage that resistive nanostructures are obtained only in a small area, which is associated with line-by-line exposure.

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.

Published under licence by IOP Publishing Ltd
One of the promising approaches to substrate surface patterning by electron-beam lithography is the microsphere photolithography that provides a relatively inexpensive and fast technique to create ordered arrays of holes in the inhibitor layer over a larger area [9].

This work presents the results of experimental studies of the selective-area growth of GaN NWs without any seed layers on SiOx/Si substrates patterned by microsphere photolithography. The morphological and optical properties of selectively grown GaN NWs are compared with GaN NWs grown on an amorphous SiOx layer.

2. Experimental
GaN NWs were grown using Riber Compact 12 MBE system equipped with indium (In) and gallium (Ga) effusion cells and a nitrogen source. The morphological properties of the samples were investigated using SUPRA 25 C. Zeiss scanning electron microscope (SEM). The substrates used were n-type silicon wafers with the (111) orientation. Initially, for the formation of a SiOx layer on the substrate surface, the substrate was treated under a temperature of 850-950 °C in the environmental conditions. Then, the SiOx/Si substrates were patterned by photolithography through microsphere lenses. Figure 1 shows an SEM image of the patterned SiOx/Si substrates. The typical diameter of holes was about 0.3 μm, the average distance between them was 1 μm, and the surface density was 108 cm$^{-2}$.

![Figure 1](image)

**Figure 1.** SEM image of the patterned SiOx/Si substrate surface with an array of holes in SiOx [10]. The scale bar corresponds to 1000 nm.

In order to grow GaN NWs, the substrates were transferred into the growth chamber and the substrate temperature ($T_s$) was set at 870 °C for the thermal treatment. The growth chamber pressure was 7·10$^{-8}$ Torr. Next, the $T_s$ was decreased to 825 °C for the SAG of GaN NWs on the patterned SiOx/Si substrate and 810 °C for the growth of GaN NWs on an amorphous SiOx layer. The nitrogen plasma source was ignited and the N flux was set at 0.4 sccm at 450 W. Finally, the Ga partial pressure was set at 2·10$^{-7}$ Torr and the Ga source was opened. The substrate preparation and the growth conditions were described in detail in [10, 11].

The optical properties of the GaN NWs were studied by PL measurements at room temperature and 77 K using a 6.5 mW He-Cd laser (325 nm).

3. Results and discussion
3.1 Morphological properties
Figure 2 shows SEM images of the grown GaN NWs on an amorphous SiOx layer (a) and the patterned SiOx/Si substrate (b). As can be seen from figure 2(a), GaN NWs grown on a SiOx layer have a high density (2.5·10$^9$ cm$^{-2}$) and partially coalescent faces. The average height and diameter of nanowires are about 3-3.4 μm and 100-200 nm, respectively. In its turn, the selectivity of GaN on the patterned SiOx/Si substrate was achieved at $T_s$ = 825 °C, which represents the lower limit for successful SAG of GaN NWs on SiOx/Si substrates without seed layers (figure 2(b)). The surface density of the selectively grown
NWs is equal to the density of the holes and is about $10^8 \text{ cm}^{-2}$.

![Figure 2](image1.png)

**Figure 2.** SEM images of GaN NWs grown on the amorphous SiO$_x$ layer (a) and the patterned SiO$_x$/Si substrate (b) [10]. The scale bars correspond to 250 nm.

The average height and diameter of nanowires are about 700 nm and 200 nm, respectively. It is worth noting that selectively grown GaN NWs are formed mainly in the $<111>$ crystallographic direction, which indicates their epitaxial bond with the Si substrate.

3.2 Optical properties

Figure 3(a) shows room temperature PL spectra of selectively grown GaN NWs on a patterned SiO$_x$/Si substrate and GaN NWs grown on an amorphous SiO$_x$ layer.

![Figure 3](image2.png)

**Figure 3.** PL spectra of the samples, measured at room temperature (a) and 77 K (b).

As can be seen from figure 3(a), both PL spectra reveal a clear near-band-edge (NBE) emission line at 3.41 eV and the yellow band luminescence (YL) with two orders of magnitude lower intensity, which indicates the high optical quality of the GaN NWs with minimal structural defects. The full width at the half maximum (FWHM) of the NBE peak of selectively grown GaN NWs is about 80 meV, which is
comparable to the FWHM of a single-crystal GaN layer [12]. The FWHM of GaN NWs grown on SiO$_x$ is about 130 meV. It should be noted that the PL intensity of GaN NWs grown on a SiO$_x$ layer is higher than that of selectively grown GaN NWs, since their surface density is approximately an order of magnitude higher.

Figure 3(b) demonstrates the low-temperature PL spectra of the grown GaN samples in the near-bandgap region. As can be seen from the figure, the PL spectrum of GaN NWs grown on a SiO$_x$ layer exhibits the main NBE line at about 3.47 eV and broad emission with a peak position at about 3.43 eV, which can be attributed to stacking faults, predominantly formed between coalescent nanowires [13-15]. In their turn, selectively grown GaN NWs exhibit only the main photoluminescence peak at a maximum of 3.47 eV, which is attributed to a donor-bound exciton D$^0$X [16]. According to the reference [17], this peak position corresponds to the D$^0$X of strain-free GaN samples.

Based on the high NBE/YL ratio, the NBE peak position, and the absence of emission lines attributed to stacking faults, we suppose that selectively grown GaN NWs have high crystalline quality.

4. Conclusion

To summarize, it was shown that selectivity of GaN NWs is achieved on a patterned SiO$_x$/Si substrate without seed layers at $T_s = 825$ °C. The GaN NWs grown on a patterned SiO$_x$/Si substrate as well as on an amorphous SiO$_x$ layer have a typical room temperature photoluminescence of high-quality single-crystal GaN. The D$^0$X peak position in the low-temperature PL spectrum of selectively grown GaN NWs indicates an unstrained crystal structure of the nanowires. The results can be used to synthesize radial heterostructures on III-N NWs and integrate III-N materials with silicon technology.

Acknowledgements

The samples were grown with the support of RFBR under the research project № 20-32-90189. The SEM study of the synthesized samples was supported by the Ministry of Science and Higher Education of the Russian Federation (state task № 0791-2020-0003). The PL study was implemented in the framework of the Basic Research Program at the National Research University Higher School of Economics (HSE University).

References

[1] Bi W 2017 Handbook of GaN Semiconductor Materials and Devices (Florida: CRC Press)
[2] Dubrovskii V G, Cirlin G E and Ustinov V M 2009 Semiconductors 43 1539
[3] De La Mata M, Zhou Xi, Furtmayr F, Teubert J, Gradečak S, Eickhoff M, Fontcuberta A and Arbiol J 2013 J. Mater. Chem. C 1 4300–12
[4] Deshpande S and Bhattacharya P 2013 Appl. Phys. Lett. 103 241117
[5] Consonni V 2013 Phys Status Solidi Rapid Res Lett 7 699–712
[6] Schuster F, Hetzl M, Weiszer S, Garrido J A, de la Mata M, Magen C, Arbiol J and Stutzmann M 2015 Nano Lett. 15 1773–79
[7] Roshko A, Brubaker M, Blanchard P, Harvey T and Bertness K A 2018 Crystals 8 366
[8] Bolshakov A D, Fedorov V V, Shugurov K Yu, Mozharov A M, Sapunov G A, Shtrom I V, Mukhin M S, Uvarov A V, Cirlin G E and Mukhin I S 2019 Nanotechnology 30 395602
[9] Zhang Z, Geng C, Hao Z, Wei T and Yan Q 2016 Adv. Colloid Interface Sci. 228 105–22
[10] Gridchin V O, Kotlyar K P, Reznik R R, Dvoretskaia L N, Parfeneva A V, Mukhin I S and Cirlin G E 2020 Tech. Phys. Lett. 46 1080–83
[11] Bolshakov A D, Dvoretskaia L N, Fedorov V V, Sapunov G A, Mozharov A M, Shugurov K Y, Shkoldin V A, Mukhin M S, Cirlin G E and Mukhin I S 2018 Semiconductors 52 2088–91
[12] Asif Khan M, Kuznia J N, Van Hove J M and Olson D T 1991 Appl. Phys. Lett. 58 526–7
[13] Corfdir P, Hauswald C, Zettler J K, Flissikowski T, Lähnemann J, Fernández-Garrido S, Geelhaar L, Grah H T and Brandt O 2014 Phys. Rev. B 90 195309
[14] Korona K P, Reszka A, Sobanska M, Perkowsa P S, Wysmolek A, Klosek K and Zytkiewicz Z R 2014 Journal of luminescence 155 293–7
[15] Khromov S, Monemar B, Avrutin V, Morkoc H, Hultman L and Pozina G 2013 Appl. Phys. Lett. 103 192101
[16] Robins L H, Bertness K A, Barker J M, Sanford N A and Schlager J B 2007 J. Appl. Phys. 101 113506

[17] Reschikov M A, Morkoç H 2005 J. Appl. Phys. 97 5-19