

Growth of long core-shell InGaN nanowires by plasma-assisted molecular beam epitaxy with gradually increasing substrate temperature

V. O. Gridchin^{1-3✉}, **T. Shugabaev**¹⁻³, **V.V. Lendyashova**¹⁻², **K. P. Kotlyar**¹⁻³, **A.I. Khrebtov**¹, **A. S. Dragunova**⁴, **N. V. Kryzhanovskaya**⁴, **R. R. Reznik**¹, **G. E. Cirlin**^{1-3,5}

¹ St Petersburg University, Saint Petersburg, Russia;

² Alferov University, Saint Petersburg, Russia;

³ IAI RAS, Saint Petersburg, Russia;

⁴ HSE University, Saint Petersburg, Russia;

⁵ ITMO University, St. Petersburg, Russia;

✉gridchin@spbau.ru

Abstract. This work presents the results of a study on the morphological and optical properties of InGaN nanowires grown using two different substrate temperature regimes. It is obtained that a gradual increase in the substrate temperature during the growth process makes it possible to obtain long morphologically homogeneous nanowires with a core-shell structure. The photoluminescence of nanowires is in the green range and is three times higher than that of similar structures grown in a stationary temperature regime.

Keywords: InGaN, nanowires, core-shell, MBE, photoluminescence, gradually increasing substrate temperature

Funding: The growth experiments and optical measurements were carried out under support of the Russian Science Foundation (project no. 23-79-00012). Optical measurements were performed, in part, in a large-scale research facility Complex Optoelectronic Stand operated by HSE University. For the morphological properties studies of grown samples the authors acknowledge Saint-Petersburg State University for a research project 95440344. Carrying out chemical etching procedures were supported by the Ministry of Science and Higher Education of the Russian Federation, research project no. 2019-1442 (project reference number FSER-2020-0013).

Citation: Gridchin V. O. et al. Modulated-temperature growth for long core-shell InGaN nanowires by plasma-assisted molecular beam epitaxy, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 16 () (2024) ...–.... DOI: <https://doi.org/10.18721/JPM>.

This is an open access article under the CC BY-NC 4.0 license (<https://creativecommons.org/licenses/by-nc/4.0/>)

© Author A. A., Author B. B., 2023. Published by Peter the Great St. Petersburg Polytechnic University.

Выращивание длинных нитевидных нанокристаллов InGaN со структурой ядро-оболочка методом плазменно-активированной молекулярно-пучковой эпитаксии при постепенном повышении температуры подложки

В. О. Гридчин^{1-3✉}, **Т. Шугабаев**¹⁻³, **В. В. Лендяшова**¹⁻², **К. П. Котляр**¹⁻³, **А. И. Хребтов**¹, **А. С. Драгунова**⁴, **Н. В. Крыжановская**⁴, **Р. Р. Резник**¹, **Г. Э. Цырлин**^{1-3,5}

¹ Санкт-Петербургский государственный университет, Санкт-Петербург, Россия, Россия;

² СПбАУ РАН им. Ж.И. Алферова, Санкт-Петербург;

³ ИАП РАН, Санкт-Петербург, Россия;

⁴ ВШЭ, Санкт-Петербург, Россия;

⁵ Университет ИТМО, Санкт-Петербург, Россия;

✉ gridchin@spbau.ru

Аннотация. В данной работе представлены результаты исследования морфологических и оптических свойств нитевидных нанокристаллов InGaN, выращенных с использованием двух различных температурных режимов. Установлено, что постепенное повышение температуры подложки в процессе роста позволяет получать длинные морфологически однородные нитевидные нанокристаллы со структурой ядро-оболочка. Фотолюминесценция таких нитевидных нанокристаллов находится в зеленом диапазоне и в три раза выше, чем у аналогичных структур, выращенных в стационарном температурном режиме.

Ключевые слова: InGaN, нитевидные нанокристаллы, ядро-оболочка, МПЭ, фотолюминесценция, постепенное повышение температуры

Финансирование: Ростовые эксперименты и исследования оптических свойств выполнены при поддержке Российского Научного Фонда (РНФ) (проект номер 23-79-00012). Оптические измерения проводились на исследовательской установке «Комплексный оптоэлектронный стенд» НИУ ВШЭ. Исследования морфологических свойств выращенных образцов были выполнены при поддержке СПбГУ, шифр проекта 95440344. Процедуры химического травления были проведены при поддержке Министерства науки и высшего образования Российской Федерации, проект тематики научных исследований № 2019-1442 (код научной темы FSER-2020-0013).

Ссылка при цитировании: Гридчин В.О. Выращивание длинных нитевидных нанокристаллов InGaN со структурой ядро-оболочка методом плазменно-активированной молекулярно-пучковой эпитаксии при постепенном повышении температуры подложки // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2024. Т. 16. № . С. ...–... . DOI: <https://doi.org/10.18721/JPM>.

Статья открытого доступа, распространяемая по лицензии CC BY-NC 4.0 (<https://creativecommons.org/licenses/by-nc/4.0/>)

© Автор А.А., Автор Б.Б., 2023. Издатель: Санкт-Петербургский политехнический университет Петра Великого.

Introduction

InGaN binary and ternary compounds are important solids for new-generation optoelectronic devices due to their direct band gap and the possibility to tune a band gap energy from ~ 0.7 to 3.4 eV [1]. However, InGaN ternary compounds with a high In content (In > 30 %) tend to phase decomposition because of the notable difference in bond lengths between In-N and Ga-N (miscibility gap) [2]. This feature complicates the growth of InGaN thin films on the commercially available substrates (c-Al₂O₃, SiC, Si) and results in poor crystal quality of the InGaN thin films with high In content.

A promising method for the fabrication of InGaN ternary compounds with high In content and high crystal quality is the growth of nanowires (NWs) [3]. Recently, we have revealed a novel type of InGaN NWs with spontaneously formed core-shell structure directly grown on Si substrate by plasma-assisted molecular beam epitaxy (PA-MBE) [4,5]. As it was shown, this type of structures is highly sensitive to the local III/N flux ratio and growth temperature which complicates the fabrication of homogeneous nanowires with needful properties [6]. In the present work, we study an approach to grow morphologically homogeneous long InGaN NWs with core-shell structure by gradually increasing substrate temperature during the growth.

Materials and Methods

The growth experiments were carried out on a Riber Compact 12 MBE system equipped with an Addon RF-N 600 plasma source and effusion cells of Ga and In. The (111)-oriented silicon substrates with n-type conductivity were used. Before the growth the substrates were etched in HF:H₂O solution and thermally annealed at 850 °C for 20 min in the growth chamber for silicon oxide removing. The growth experiments were carried out under slightly nitrogen-rich growth conditions. The beam equivalent pressures of the In and Ga sources were equal to each other and corresponded to $1 \cdot 10^{-7}$ Torr. In the first case, we carried out growth with stationary substrate temperature which consisted of 650 °C. The growth time corresponds to 51h. In the second case, we gradually increased substrate temperature after 21h of the growth process. The rate of the increase of substrate temperature was 1 °C per hour.

A study of the morphology properties was conducted via a Carl Zeiss Supra 25 scanning electron microscopy (SEM) system. The optical properties of the samples are investigated using photoluminescence (PL) spectroscopy at room temperature with a He-Cd laser (325 nm) and pump power of 6.5 mW and silicon photodetector. The polarity of long InGaN NWs was determined by wet chemical etching of NWs in a KOH:H₂O solution according to the method described in [7]. The etching time was 40 seconds.

Results and Discussion

Figure 1(a,b) demonstrates typical SEM images in cross-section and plan view of InGaN nanowires grown at a stationary substrate temperature (a) and gradually increasing substrate temperature (b). On the left side, the substrate temperature change graphs are presented. Three-dimensional nanostructures consisting of nanowires near the substrate surface and partly-coalesced nanostructures above are obtained in the stationary growth regime (Fig. 1a). These nanostructures correspond to similar ones grown at lower substrate temperatures [6]. The formation mechanism of them can be explained by the decreasing effective temperature on the growth surface with increasing the height of nanostructures. To eliminate the influence of the effective growth temperature, the substrate temperature was gradually increased starting from 21 h of growth. In this growth regime, the formation of morphologically homogeneous NWs with height of about 7.5 μm was achieved (Fig. 1b). From the plan-view SEM image “cracks” are visible that definitely indicate the spontaneously formed core-shell structure within the NWs [5].

To identify the polarity of long core-shell InGaN NWs they were treated in the KOH solution [7]. For the first time, it was obtained that the cores and shells of NWs exhibit a Me-polarity and N-polarity, respectively.

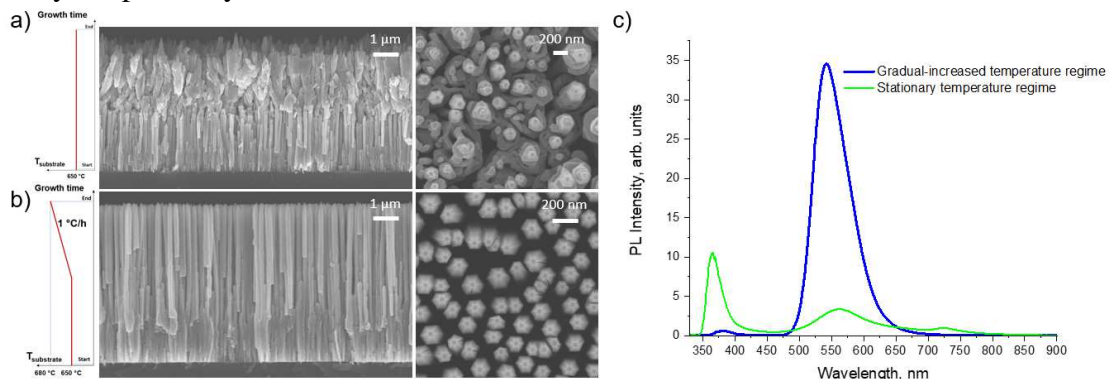


Fig. 1. Typical SEM images in cross-section view and plan-view of InGaN nanowires grown at stationary substrate temperature (a) and gradual-increased substrate temperature (b). Room-temperature PL spectra of grown samples (c). On the left side, the substrate temperature change graphs are presented.

Figure 1c demonstrates room-temperature PL spectra of grown samples. The PL spectrum consisting of relatively low-intensive areas in the range from 350 to 750 nm was obtained for the sample grown at the stationary temperature regime (green line in Fig. 1c) that explained by the formation of InGaN nanostructures with different indium content. The relatively homogeneous PL spectrum with a maximum at 540 nm and corresponding full-width at half-maximum about 64 nm

was obtained in the gradual-increased temperature regime (blue line in Fig. 1c). Moreover, the integral PL intensity of these NWs is 3 times higher than that of nanostructures grown at stationary temperature regime. A relatively low-intensity PL area near 380 nm corresponds to the emission from the shell of NWs.

Conclusion

In this work, the results of studies on the morphological and optical properties of InGaN nanowires grown at two different substrate temperature modes were presented. It was obtained that using gradual-increased temperature growth allows one to fabricate morphological homogeneous long NWs with core-shell structure. The PL spectrum of these NWs corresponds to the green range and 3 times higher than similar ones for nanostructures grown at steady-state temperature growth mode. The results obtained could provide valuable data in the field of nanomaterials engineering.

REFERENCES

1. **Pandey A., Reddeppa M., Mi Z.**, Recent progress on micro-LEDs, *Light: Advanced Manufacturing*. 4 (2024) 519–542.
2. **Ho I., Stringfellow G.**, Solid phase immiscibility in GaInN, *Applied Physics Letters*. 69(18) (1996) 2701–2703.
3. **Dubrovskii V., Cirlin G., Ustinov V.**, Semiconductor nanowhiskers: synthesis, properties, and applications, *Semiconductors*. 43 (2009) 1539–1584.
4. **Gridchin V.O., Kotlyar K.P., Reznik R.R., Dragunova A.S., Kryzhanovskaya N.V., Lendyashova V.V., Kirilenko D.A., Soshnikov I.P., Shevchuk D.S., Cirlin G.E.**, Multi-colour light emission from InGaN nanowires monolithically grown on Si substrate by MBE, *Nanotechnology*. 32(33) (2021) 335604.
5. **Soshnikov I.P., Kotlyar K.P., Reznik R.R., Gridchin V.O., Lendyashova V.V., Vershinin A.V., Lysak V.V., Kirilenko D.A., Bert N.A., Cirlin G.E.**, Specific Features of Structural Stresses in InGaN/GaN Nanowires, *Semiconductors*. 55(10) (2021) 795–798.
6. **Reznik R.R., Gridchin V.O., Kotlyar K.P., Kryzhanovskaya N.V., Morozov S.V., Cirlin G.E.** Synthesis of morphologically developed InGaN nanostructures on silicon: influence of the substrate temperature on the morphological and optical properties, *Semiconductors*. 54 (2020) 1075–1077.
7. **Lendyashova V.V., Kotlyar K.P., Reznik R.R., Berezovskaya T.N., Nikitina E.V., Soshnikov I.P., Cirlin G.E.** Wet chemical etching of GaN or InGaN nanowires on Si substrate for micro and nano-devices fabrication. *1695(1)* (2020) 012047.

THE AUTHORS

GRIDCHIN Vladislav O.
gridchinv@yandex.ru
ORCID: 0000-0002-6522-3673
SHUGABAEV Talgat
talgashugabaev@mail.ru
ORCID: 0000-0002-4110-1647
LENDYASHOVA Vera V.
erilerican@gmail.com
ORCID: 0000-0001-8192-7614
KOTLYAR Konstantin P.
konstantin21kt@gmail.com
ORCID: 0000-0002-0305-0156
Khrebtov Artem I.
khrebtovart@mail.ru
ORCID: 0000-0001-5515-323X

DRAGUNOVA Anna S.
anndra@list.ru
ORCID: 0000-0002-0181-0262
KRYZHANOVSKAYA Natalia V.
nataliakryzh@gmail.com
ORCID: 0000-0002-4945-9803
REZNIK Rodion R.
moment92@mail.ru
ORCID: 0000-0003-1420-7515
CIRLIN GeorgeE.
cirlin.beam@mail.ioffe.ru
ORCID: 0000-0003-0476-3630