

Conference materials

UDC 538.975

DOI: <https://doi.org/10.18721/JPM.153.361>

Plasma assisted molecular beam epitaxy growth of InGaN nanostructures on Si substrates

V. O. Gridchin^{1, 2, 3}✉, R. R. Reznik^{1, 2, 3}, K. P. Kotlyar³, T. M. Shugabaev¹,
A. S. Dragunova⁴, N. V. Kryzhanovskaya⁴, G. E. Cirlin^{1, 2, 3}

¹ Alferov University, St. Petersburg, Russia;

² IAI RAS, St. Petersburg, Russia;

³ St. Petersburg State University, St. Petersburg, Russia;

⁴ National Research University Higher School of Economics, St. Petersburg, Russia

✉ gridchinfo@gmail.com

Abstract. In this work, we study the influence of the substrate temperature on the structural and optical properties of InGaN nanostructures synthesized by plasma-assisted molecular beam epitaxy. We show that ternary InGaN alloys with a chemical composition within the miscibility gap can be synthesized under N-rich growth conditions at the substrate temperatures from 600 to 670 °C. The results can be used to create visible and white light-emitting diodes on Si substrates.

Keywords: InGaN, silicon, structural properties, optical properties, plasma-assisted molecular beam epitaxy

Funding: The samples were grown under financial support of RSF grant (project No.19-72-30010). The structural properties were studied under financial support of the research grant of St. Petersburg State University No. 93020138. The study of photoluminescence was implemented in the framework of the Basic Research Program at the National Research University Higher School of Economics (HSE University).

Citation: Gridchin V. O., Reznik R. R., Kotlyar K. P., Shugabaev T. M., Dragunova A. S., Kryzhanovskaya N. V., Cirlin G. E., Plasma assisted molecular beam epitaxy growth of InGaN nanostructures on Si substrates. St. Petersburg State Polytechnical University Journal. Physics and Mathematics, 15 (3.3) (2022) 311–314. DOI: <https://doi.org/10.18721/JPM.153.361>

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

Материалы конференции

УДК 538.975

DOI: <https://doi.org/10.18721/JPM.153.361>

Молекулярно-пучковая эпитаксия с плазменной активацией азота InGaN наноструктур на кремнии

В. О. Гридчин^{1, 2, 3}✉, Р. Р. Резник^{1, 2, 3}, К. П. Котляр³, Т. М. Шугабаев¹,
А. С. Драгунова⁴, Н. В. Крыжановская⁴, Г. Э. Цырлин^{1, 2, 3}

¹ Санкт-Петербургский академический университет им. Ж. И. Алферова РАН, Санкт-Петербург, Россия;

² Институт аналитического приборостроения РАН, Санкт-Петербург, Россия;

³ Санкт-Петербургский государственный университет, Санкт-Петербург, Россия;

⁴ Национальный исследовательский университет «Высшая школа экономики», Санкт-Петербург, Россия

✉ gridchinfo@gmail.com

Аннотация. В работе представлены результаты исследования влияние температуры подложки на структурные и оптические свойства наноструктур InGaN, синтезированных методом молекулярно-пучковой эпитаксии с плазменной активацией азота. Показано, что тройной раствор InGaN с химическим составом в зоне разрыва смешиваемости

может быть синтезирован в азот-обогащенных ростовых условиях при температурах подложки от 600 до 670 °С. Полученные результаты могут представлять интерес для создания оптоэлектронных устройств на кремниевых подложках в видимом диапазоне.

Ключевые слова: InGaN, кремний, структурные свойства, оптические свойства, молекулярно-пучковая эпитаксия с плазменной активацией азота

Финансирование: Ростовые эксперименты проведены при поддержке Российского научного фонда (проект № 19-72-30010). Исследования структурных свойств проведены при поддержке гранта СПбГУ № 93020138. Исследования фотолюминесценции проведены в рамках Программы фундаментальных исследований НИУ ВШЭ.

Ссылка при цитировании: Гридчин В. О., Резник Р. Р., Котляр К. П., Шугабаев Т. М., Драгунова А. С., Крыжановская Н. В., Цырлин Г. Э. Молекулярно-пучковая эпитаксия с плазменной активацией азота InGaN наноструктур на кремнии // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2022. Т. 15. № 3.3. С. 311–314. DOI: <https://doi.org/10.18721/JPM.153.361>

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

Introduction

Ternary InGaN alloys are promising semiconductor materials for visible solid-state lighting [1] and renewable energy sources [2, 3] due to the direct band gap with the energy from 3.37 (GaN) to 0.7 eV (InN). However, the growth of InGaN alloys with the high In content ($x_{\text{In}} > 0.3$) and high crystal quality is an extremely challenging task due to the large difference in interatomic spacing between InN and GaN [4]. Plasma-assisted molecular beam epitaxy (PA-MBE) technique possesses a great potential for growing high-quality InGaN epitaxial structures. In particular, relatively low growth pressures and consumptions of the high purity nitrogen and group-III materials (7N) allow one to reduce contamination of the epitaxial structures. The PA-MBE growth process may proceed far from thermodynamic equilibrium that can potentially contribute to grow of InGaN over the entire compositional range. One of the possible approaches to obtain InGaN alloys with the high In content and high crystal quality is the growth of InGaN nanowires (NWs) [5, 6]. It has recently been shown that InGaN NWs with the In content of about 35% can be grown on silicon substrates by the PA-MBE [7, 8]. However, the influence of the growth conditions in the PA-MBE on the physical properties of ternary InGaN alloys is still poorly studied. This work is devoted to studying the substrate temperature influence on the physical properties of ternary InGaN alloys grown by plasma-assisted molecular beam epitaxy on Si substrates.

Materials and Methods

The InGaN nanostructures were grown directly on p-type Si(111) substrates using Riber Compact 12 MBE setup, equipped with Ga, In effusion cells, and the nitrogen plasma source. Initially, the substrate was transferred to the growth chamber and heated to 950 °C for thermal treatment. Next, the substrate temperature was decreased to the desired value allowing the growth of InGaN nanostructures. We carried out several experiments at growth temperatures from 600 °C to 670 °C. An atomically clean silicon surface was observed by the reflection high-energy electron diffraction (RHEED) at all growth temperatures. After the stabilization of the growth temperature, the nitrogen plasma source was ignited at 450 W and the N flux was set to 0.4 sccm. At this moment, the growth chamber pressure was $7.4 \cdot 10^{-6}$ Torr. Finally, In and Ga effusion cells were simultaneously opened and the InGaN structure was grown during 20 hours. The growth was performed under N-rich conditions. The fluxes of In and Ga, measured by Bayard-Alpert vacuum gauge before the growth, were equal to $1 \cdot 10^{-7}$ Torr.

The morphology of the samples was studied by scanning electron microscopy (SEM Supra 25 Zeiss). The measurements of photoluminescence were performed at room temperature (RT PL) under a helium–cadmium (He–Cd) metal-vapor laser with a wavelength of 325 nm at 15.5 mW. The PL signal was detected using a DK480 Spectral products monochromator and a single-channel silicon detector using synchronous detection (SRS 510 “Stanford Research Systems”).

Results and Discussion

Fig. 1 shows the SEM images of the samples grown at: (a) 600 °C; (b) 650 °C; (c) 670 °C. As can be seen from Fig. 1, *a*, the ternary InGaN alloy is synthesized in the three-dimensional structure consisted of a nanocolumn layer near the substrate and “nanoflowers” formed above. The structural properties of this sample were studied in detail in [9]. The average height of nanostructures is about 4.8 μm. The InGaN NWs separated from each other are grown at the substrate temperature of about 650 °C (see Fig. 1, *b*). The average height of NWs is 2.9 μm and their average diameter is 100 nm. The InGaN compact layer with an average height of 1.6 μm is formed at 670 °C (see Fig. 1, *c*).

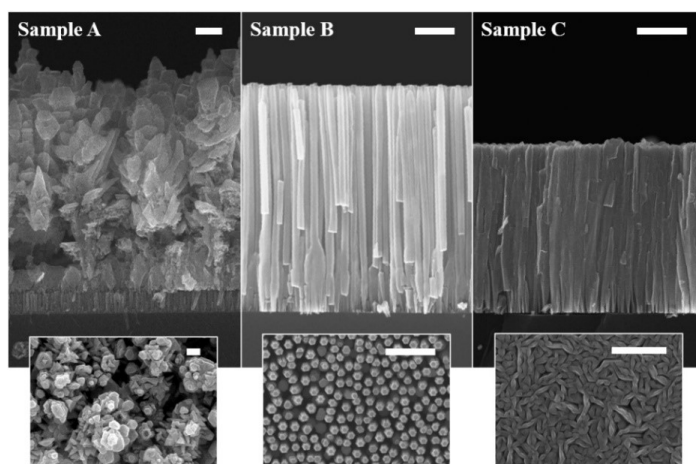


Fig. 1. Typical cross-section SEM images of the samples grown at (a) 600 °C, (b) 650 °C, (c) 670 °C. The insertions demonstrate corresponding plan-view SEM images. The scale bars are 500 nm

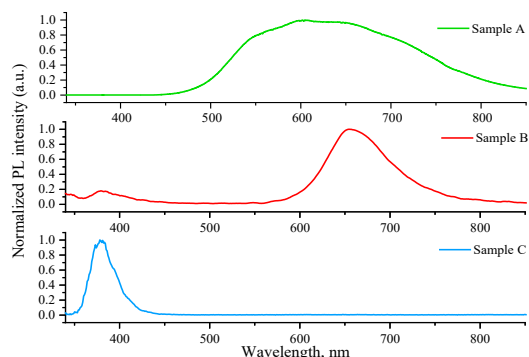


Fig. 2. RT PL spectra of the InGaN nanostructures grown at various substrate temperatures. The green line is the PL of sample A. The red line is the PL of sample B. The blue line is the PL of sample C

Fig. 2 shows the normalized RT PL spectra of the samples. The green line is the RT PL spectrum of sample A grown at the lowest temperature. The sample exhibits a broad emission spectrum in the range from 450 to 850 nm which is explained by the inhomogeneous distribution of In atoms within the structure [9]. In the case of sample B (see the red line in Fig. 2), the RT PL spectrum demonstrates two emission areas: the first with the maximum at 380 nm and the second with the maximum at 656 nm. In the work [7] we have shown that InGaN NWs exhibit spontaneously formed core-shell structure with the In content in the core of about 30–35% and in the shell of about 0–4%. As has been shown, the use of RT PL from the core-shell InGaN NWs can be converted into the actual chemical composition by the Vegard's law with a bowing parameter of 1.43 eV. In this

regard, the InGaN NWs of sample B consist of a spontaneously formed core-shell structure. The first area of RT PL corresponds to the emission from the shell with the In content of about 0–4% and the second area corresponds to the core with the In content of about 44%. The blue line in Fig. 2 is the emission of sample C. The spectrum has a maximum centered at 380 nm that corresponds to an In content of 4%. This indicates that indium adatoms practically do not incorporate into the growing structure under these growth conditions, which is explained by the predominance of the InN thermal decomposition instead of the formation of InGaN nanostructures.

Conclusion

To conclude, we have investigated the influence of the substrate temperature on the structural and optical properties of ternary InGaN alloys. The three-dimensional InGaN nanostructures

with an inhomogeneously distributed In content was grown at 600 °C. The InGaN NWs with the In content of about 40% were grown at 650 °C. The InGaN compact layer with the In content of about 4% was grown at 670 °C. The results can be used to create visible and white light-emitting diodes directly on Si substrates.

REFERENCES

1. Bui H. Q. T., Velpula R. T., Jain B., Aref O. H., Nguyen H.-D., Lenka T. R., Nguyen H. P. T., Full-Color InGaN/AlGaIn Nanowire Micro Light-Emitting Diodes Grown by Molecular Beam Epitaxy: A Promising Candidate for next Generation Micro Displays, *Micromachines*. 10 (8) (2019) 492.
2. Chu S., Li W., Yan Y., Hamann T., Shih I., Wang D., Mi Z., Roadmap on Solar Water Splitting: Current Status and Future Prospects, *Nano Futures*. 1 (2) (2017) 022001.
3. Lin J., Wang W., Li G., Modulating Surface/Interface Structure of Emerging InGaN Nanowires for Efficient Photoelectrochemical Water Splitting, *Advanced Functional Materials*. 30 (52) (2020) 2005677.
4. Ho I., Stringfellow G., Solid Phase Immiscibility in GaInN, *Applied Physics Letters*. 69 (18) (1996) 2701–2703.
5. Kuykendall T., Ulrich P., Aloni S., Yang P., Complete Composition Tunability of InGaN Nanowires Using a Combinatorial Approach, *Nature materials*. 6 (12) (2007) 951–956.
6. Roche E., Andre Y., Avit G., Bougerol C., Castellucci D., Réveret, F., Gil E., Médard F., Leymarie J., Jean T., Dubrovskii V. G., Trassoudaine A., Circumventing the Miscibility Gap in InGaN Nanowires Emitting from Blue to Red, *Nanotechnology*. 29 (46) (2018) 465602.
7. 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.
8. Tabata T., Paek J., Honda Y., Yamaguchi M., Amano H., Growth of InGaN Nanowires on a (111) Si Substrate by RF-MBE, *physica status solidi c*. 9 (3-4) (2012) 646–649.
9. Gridchin V. O., Kotlyar K. P., Reznik R. R., Shevchuk D. S., Kirilenko D. A., Bert N. A., Soshnikov I. P., Cirlin G. E., Peculiarities of the Structural Properties of InxGal-XN Polytype Nanostructures Grown by Molecular-Beam Epitaxy, *Journal of Physics: Conference Series – IOP Publishing*. 1482 (1) (2020) 012014.

THE AUTHORS

GRIDCHIN Vladislav O.
gridchinfo@gmail.com
ORCID: 0000-0002-6522-3673

REZNIK Rodion R.
moment92@mail.ru
ORCID: 0000-0003-1420-7515

KOTLYAR Konstantin P.
konstantin21kt@gmail.com
ORCID: 0000-0002-0305-0156

SHUGABAEV Talgat M.
talgashugabaev@mail.ru
ORCID: 0000-0002-4110-1647

DRAGUNOVA Anna S.
anndra@list.ru
ORCID: 0000-0002-0181-0262

KRYZHANOVSKAYA Natalia V.
nataliakryzh@gmail.com
ORCID: 0000-0002-4945-9803

CIRLIN George E.
george.cirlin@mail.ru
ORCID: 0000-0003-0476-3630

Received 20.07.2022. Approved after reviewing 22.07.2022. Accepted 23.07.2022.