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MBE growth of InGaN nanowires on SiC/Si(111) and Si(111) substrates: comparative analysis

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In this work, InGaN nanowires with a high In content were grown, for the first time on hybrid SiC/Si substrates and compared with InGaN nanowires grown on Si. It was shown that InGaN nanowires on SiC/Si have lower indium content (by about 10%) compared to the nanowires on Si. The results can be beneficial for studying the growth mechanisms of InGaN nanowires and creating optoelectronic devices in the visible spectral range.

Keywords: InGaN, nanowires, molecular beam epitaxy, SiC/Si, morphological properties, optical properties, miscibility gap, silicon carbide on silicon

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InGaN ternary compounds are of great interest for solid-state light emitters [1] and renewable energy sources [2] since they are direct-band semiconductors with the band gap ranging from 0.7 (InN) to 3.43 eV (GaN) depending on the chemical composition. However, one of the main specific features of the $In_xGa_{1-x}N$ solid solutions is their spinodal decomposition restricting obtaining homogeneous layers with the In content of $x = 0.2$ to 0.8 at the growth temperature of $> 600°C$ [3]. This peculiar feature is caused by a great difference in the In−N and Ga−N bond lengths. In addition, growth of InGaN layers with a low number of structural defects is hampered by unavailability of lattice matched substrates. One of the possible ways to overcome the mentioned problems is synthesis of nanowires (NW). It was shown that the InGaN synthesis in the entire chemical composition range is possible in the case of growing NW [4]. Besides that, efficient relaxation of elastic stresses on the NW walls ensures the growth of almost fully defect-free structures on mismatched substrates. For instance, the GaN NWs on Si [5] and III-N may be synthesized by different epitaxial techniques on amorphous or highly rough substrates with retention of their excellent crystalline and optical characteristics [6,7].

To the best of our knowledge, direct MBE-growth of InGaN NWs has been performed by only a few groups [8–12], and the influence of the growth conditions on physical properties of those nanostructures still remains an open question. Studies [12,13] showed that in a narrow growth temperature range InGaN NWs on Si are formed in the „core−shell“ heterostructure at a high In content in the „core“ (no less than 30%) and low In content in the „shell“ (no more than 4%).

In this work, the possibility of growing InGaN NWs with high In content on hybrid SiC/Si(111) substrates has been shown for the first time, and comparative analysis with InGaN NWs grown on the Si(111) substrate has been conducted.

Boron-doped Si(111) wafers were used for the growth experiments. First a SiC layer was formed on a half of wafers by the method of concerted atom displacement proposed by Kukushkin et al. [14], at the temperature of 1310°C and CO pressure of 0.4 Torr. The time of the layer formation was 20 min. Fig. 1, *a* presents a typical scanning electron microscopy (SEM) image of the prepared substrate with a SiC layer. Under the layer there are pores that arise from silicon vacancies and are responsible for the relaxation of elastic strains induced by the Si/SiC lattice mismatch. The average values of thickness and porosity are as follows: the SiC layer thickness is ~ 150 nm, the porous layer thickness is $\sim 3\,\mu$m, the bulk porosity is $\sim 50\%$. The SiC layer is mainly the 4*H* polytype, which was found out from photoluminescence (PL) measurements at the liquid helium temperature; the maximal emission was observed in the spectrum at 357.57 nm (Fig. 1, *b*) [14].

The InGaN NW growth experiments were carried out using the molecular-beam epitaxy setup Riber Compact 12 at the following growth parameters. The Si substrates loaded into the growth chamber were heated to 950°C and held at this temperature for 20 min for thermal cleaning. Next, the substrate temperature was reduced to 650°C, and the nitrogen plasma source was initiated. The source

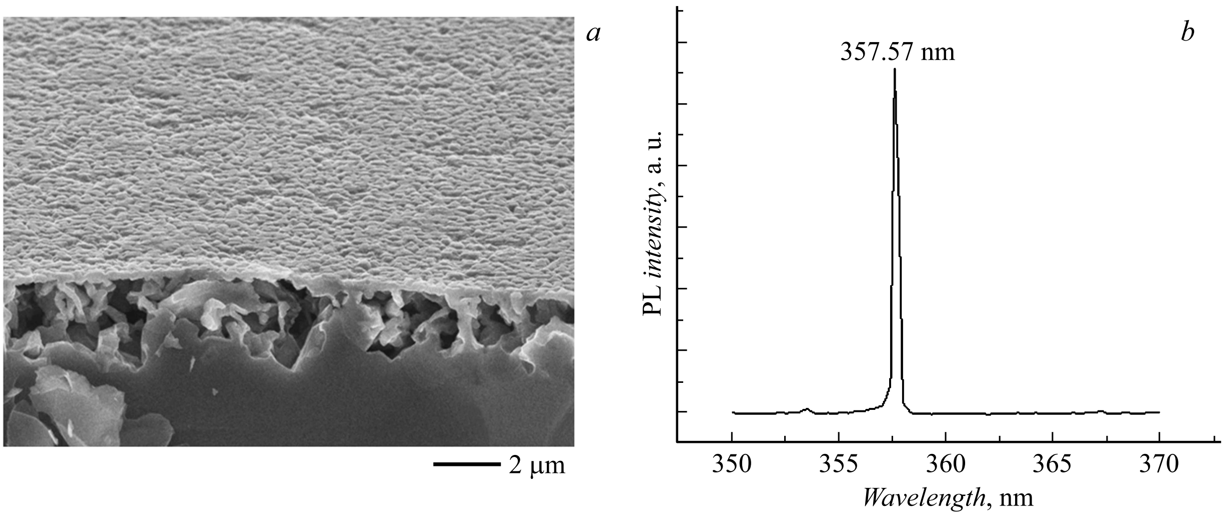

Figure 1. *a* — Typical isometric SEM image of the SiC/Si substrate; *b* — PL of the SiC/Si substrate at the liquid helium temperature.

power was 450 W at the nitrogen flow of 0.4 sccm. The power was determined based on the maximum of the plasma glow intensity. Upon the pressure stabilization at $7.4 \cdot 10^{-6}$ Torr, the indium and gallium sources were opened simultaneously. The element flows pre-measured with the Bayard-Alpert gauge were $1 \cdot 10^{-7}$ Torr for each metal. The InGaN NW growth lasted for 21 h. InGaN NWs on the SiC/Si substrate were grown at the same growth parameters.

The surface condition and NW formation process were monitored by the method of reflected high-energy electron diffraction. On the completion of growth, point diffraction corresponding to the wurtzite crystal structure was observed for both InGaN on Si and InGaN on SiC/Si.

Morphological properties of the samples were analyzed with a SEM Supra 25 Zeiss. Optical properties were studied by the PL at room temperature and liquid helium temperature by using a He–Cd-laser (325 nm) at the excitation power of 6.5 mW. The signal was detected with the Sol Instruments MS5204i monochromator and silicon photodetector.

Fig. 2 presents the typical isometric SEM-images of NWs grown on the Si substrate (*a*) and SiC/Si substrate (*b*). The respective inserts present plane view SEM images of the NWs. As one can see, NWs on both Si and SiC/Si have similar morphologies: NWs arranged tightly to each other with the average surface density of $7 \cdot 10^9\,\mathrm{cm}^{-2}$, length of $2.8\,\mu$m and diameter 100 nm. It should be noted that NWs often are coalesced at their bases at the height of ~ 280 nm from the substrate surface. A similar tendency of the morphology changes was reported in paper [12].

Fig. 3 presents PL spectra of the grown samples. The PL integral intensity of NWs on SiC/Si is 5 times higher than that of NWs on Si, which indicates their better crystallinity. Both spectra exhibit two PL regions: a shortwave one with the maximum at 380 nm and a longwave one with the maximum at 380 nm for NWs on SiC/Si and 657 nm for NWs on Si. As shown earlier [12], InGaN NWs grown on the Si substrate exhibit core-shell heterostructure and their shortwave PL signal corresponds to the shell emission, while the longwave one corresponds to the core emission. Since NWs grown on SiC/Si have a similar PL spectrum, we assume the existence of a similar core–shell structure within them. As per the chemical composition estimates for ternary solutions $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N}$ according to the modified Vegard's law [14], $E_{\mathrm{InGaN}} = xE_{\mathrm{InN}} + (1-x)E_{\mathrm{GaN}} - bx(1-x)$, where E_{InN} is the InN optical band gap width, E_{GaN} is the GaN optical band gap width, b is the bending parameter, the In content in the core of NWs on Si is $\sim 44\%$, while that for NWs on SiC/Si is $\sim 33\%$. The estimation was carried out using the following quantities: $E_{\mathrm{InN}} = 0.7$ eV, $E_{\mathrm{GaN}} = 3.43$ eV, $b = 1.43$ eV. The parameter b value implies the presence of the structure internal stresses as per [15] and is well consistent with the chemical composition measurements of InGaN NWs with the core–shell structure [12]. To our mind, the difference in chemical compositions of NWs grown on Si and SiC/Si is caused by the following reasons. The first one consists in essential differences between the parameters and lattice symmetry of Si and InGaN lattice in NWs.

Based on the Vegard's law $a_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N}} = xa_{\mathrm{InN}} + (1-x)a_{\mathrm{GaN}}$ [16], it is possible to demonstrate that mismatch of hexagonal lattices parameters a $\mathrm{In}_{0.44}\mathrm{Ga}_{0.56}\mathrm{N}$ (3.34 Å) and $\mathrm{In}_{0.33}\mathrm{Ga}_{0.67}\mathrm{N}$ (3.3 Å) is less significant and equals only 1.2%. However, the mismatch of the Si lattice constant (in the (111)) direction of 3.84 Å and that of $\mathrm{In}_{0.44}\mathrm{Ga}_{0.56}\mathrm{N}$ is $\sim 13\%$. A similar ratio between the $\mathrm{In}_{0.33}\mathrm{Ga}_{0.67}\mathrm{N}$ and 4*H*-SiC (3.081 Å) lattice constants is $\sim 7.1\%$ [16]. Under such conditions, NWs grown on SiC/Si will be exposed to compressing strains, contrary to tension strains acting on NWs grown on Si. The other reason

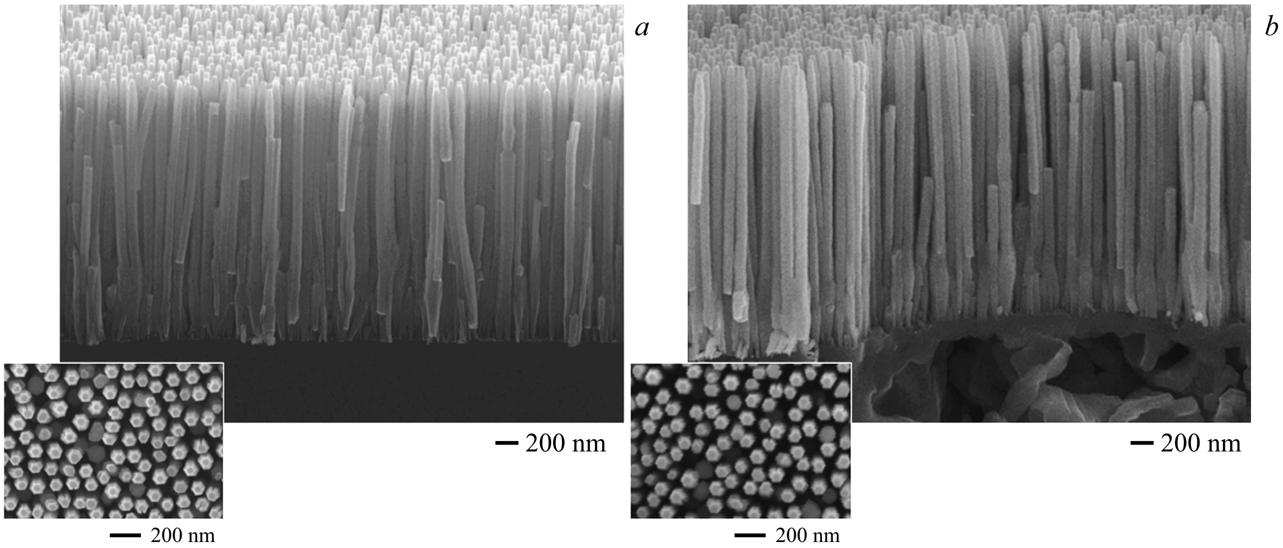

Figure 2. Typical isometric SEM-images of the InGaN NWs grown on the Si substrate (*a*) and SiC/Si substrate (*b*). The inserts contain SEM-images (top view).

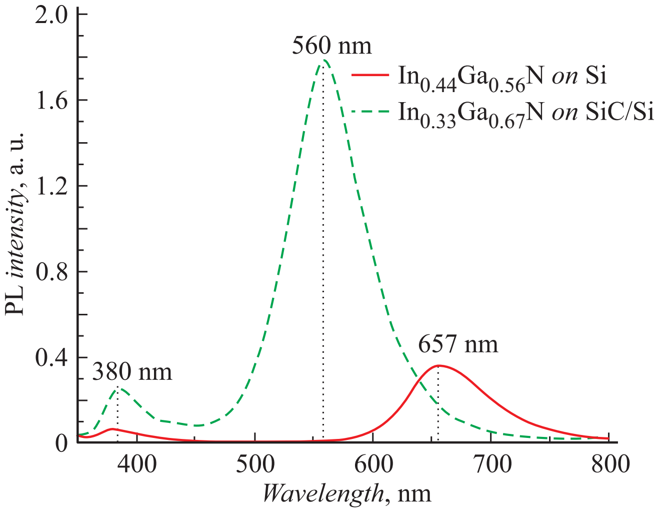

Figure 3. Room-temperature PL spectra of InGaN NW on Si (solid line) and SiC/Si (dashed line).

for that effect is thermal deformations arising between SiC/pores/Si, SiC/InGaN, Si/InGaN and InGaN core/shell. To our opinion, these reasons lead to the fact that the In content in NWs on Si is higher than that in NWs on SiC/Si.

Thus, we have grown InGaN NWs with the core–shell structure and high In content on the Si substrates and hybrid substrates SiC/Si(111). InGaN NWs grown on different substrates with all else growth parameters being equal exhibit similar surface morphologies. The PL maximum of InGaN on SiC/Si is shifted by ~ 100 nm to the shortwave regions, which we explain by a lower In content in the NW core. The results can be beneficial for studying the growth mechanisms of InGaN nanowires and creating optoelectronic devices in the visible spectral range.

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Conflict of interests

The authors declare that they have no conflict of interests.

References

[1] H.Q.T. Bui, R.T. Velpula, B. Jain, O.H. Aref, H.-D. Nguyen, T.R. Lenka, H.P.T. Nguyen, Micromachines, **10** (8), 492 (2019). DOI: 10.3390/mi10080492
[2] J. Ramanujam, A. Verma, B. González-Díaz, R. Guerrero-Lemus, C. del Cañizo, E. García-Tabarés, I. Rey-Stolle, F. Granek, L. Korte, M. Tucci, J. Rath, U.P. Singh, T. Todorov, O. Gunawan, S. Rubio, J.L. Plaza, E. Diéguez, B. Hoffmann, S. Christiansen, G.E. Cirlin, Prog. Mater. Sci., **82**, 294 (2016). DOI: 10.1016/j.pmatsci.2016.03.005
[3] I. Ho, G.B. Stringfellow, Appl. Phys. Lett., **69** (18), 2701 (1996). DOI: 10.1063/1.117683
[4] E. Roche, Y. Andre, G. Avit, C. Bougerol, D. Castelluci, F. Rѓveret, E. Gil, F. Médard, J. Leymarie, T. Jean, V.G. Dubrovskii, A. Trassoudaine, Nanotechnology, **29** (46), 465602 (2018). DOI: 10.1088/1361-6528/aaddc1
[5] V.G. Dubrovskii, G.E. Cirlin, V.M. Ustinov, Semiconductors, **43** (12), 1539 (2009).
DOI: 10.1134/S106378260912001X].

[6] M.A. Johar, H.-G. Song, A. Waseem, M.A. Hassan, I.V. Bagal, Y.-H. Cho, S.-W. Ryu, Appl. Mater. Today, **19**, 100541 (2020). DOI: 10.1016/j.apmt.2019.100541

[7] V.O. Gridchin, K.P. Kotlyar, R.R. Reznik, L.N. Dvoretskaya, A.V. Parfen'eva, I.S. Mukhin, G.E. Cirlin, Tech. Phys. Lett., **46** (11), 1080 (2020). DOI: 10.1134/S1063785020110061.

[8] R.R. Reznik, K.P. Kotlyar, N.V. Kryzhanovskaya, S.V. Morozov, G.E. Cirlin, Tech. Phys. Lett., **45** (11), 1111 (2019). DOI: 10.1134/S1063785019110129.

[9] H. Chen, P. Wang, H. Ye, H. Yin, L. Rao, D. Luo, X. Hou, G. Zhou, R. Nötzel, Chem. Eng. J., **406**, 126757 (2021).

[10] T. Tabata, J. Paek, Y. Honda, M. Yamaguchi, H. Amano, Phys. Status Solidi C, **9** (3-4), 646 (2012). DOI: 10.1002/pssc.201100446

[11] A. Vajpeyi, A. Ajagunna, K. Tsagaraki, M. Androulidaki, A. Georgakilas, Nanotechnology, **20** (32), 325605 (2009). DOI: 10.1088/0957-4484/20/32/325605

[12] V.O. Gridchin, K.P. Kotlyar, R.R. Reznik, A.S. Dragunova, N.V. Kryzhanovskaya, V.V. Lendyashova, D.A. Kirilenko, I.P. Soshnikov, G.E. Cirlin, Nanotechnology, **32** (33), 335604 (2021). DOI: 10.1088/1361-6528/ac0027

[13] I.P. Soshnikov, K.P. Kotlyar, R.R. Reznik, V.O. Gridchin, V.V. Lendyashova, A.V. Vershinin, V.V. Lysak, D.A. Kirilenko, N.A. Bert, G.E. Cirlin, FTP, **55** (9), 785 (2021). DOI: 10.21883/FTP.2021.09.51295.25 (in Russian)

[14] S.A. Kukushkin, A.V. Osipov, Phys. Solid State, **50** (7), 1238 (2008). DOI: 10.1134/S1063783408070081.

[15] G. Orsal, Y. El Gmili, N. Fressengeas, J. Streque, R. Djerboub, T. Moudakir, S. Sundaram, A. Ougazzaden, J.P. Salvestrini, Opt. Mater. Express, **4** (5), 1030 (2014). DOI: 10.1364/OME.4.001030

[16] D. Zhu, D.J. Wallis, C.J. Humphreys, Rep. Prog. Phys., **76** (10), 106501 (2013). DOI: 10.1088/0034-4885/76/10/106501