Morphology transformation of InGaN nanowires grown on Si substrate by PA-MBE

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Abstract. The influence of the growth time on the structural properties of InGaN nanowires grown on Si substrate by plasma-assisted molecular beam epitaxy are studied. Under appropriate other growth conditions, the growth for 2h leads to the formation of separated nanowires, whereas the growth for 2h 30min and 3h leads to the formation of nanostructures such as nano-umbrellas. The separated NWs exhibit a photoluminescence spectrum with maxima at about 590 nm, whereas the nano-umbrellas show two pronounced photoluminescence lines at 421 and 619 nm.

1. Introduction
Nowadays, InGaN ternary compounds have proven themselves well for the creation of light-emitting devices and renewable energy sources due to their unique properties [1]. First of all, a band gap energy of InGaN can vary from 0.7 to 3.43 eV depending on the In content inside the compound [2]. However, the lack of the lattice-matched substrates for InGaN makes it impossible to grow high-quality epitaxial layers with a homogeneously chemical composition. Another challenge is the wide miscibility gap of InGaN [3] significantly complicating the formation of InGaN ternary compounds with high In content. A promising approach to overcome these limitations is the growth of nanowire (NWs) structures. It has been demonstrated that the elastic strain relaxation on the nanowire sidewalls can help to overcome the InGaN miscibility gap and achieve a better band gap tunability than in a bulk alloy [2,4,5]. Another advantage of the elastic strain relaxation on the nanowire sidewalls is the possibility of the growth of practically defect-free NWs on substrates with different lattice constants and different thermal expansion coefficients [6,7]. In this regard, the growth of NWs on silicon substrates is one the most promising approach from the point of view of wide applications ones and low cost.

Recently, we have shown that during a plasma-assisted molecular beam epitaxy the InGaN nanowires with spontaneously formed core-shell structures are grown at a narrow growth temperature range, and the core contains high In mole fraction (about 35 %), whereas the shell contains no more than 4 % of In [8,9]. Also, it was obtained that at the lower growth temperature in comparison with ones of InGaN NWs the InGaN nanostructures with three-dimensional morphology can be grown. However, there is
still a lack of information about the influence of the growth conditions on the physical properties of these nanostructures. In this work, the influence of the growth time on the formation of InGaN nanowires is studied.

2. Experimental
The experiments of the InGaN NWs growth were carried out using Riber Compact 12 MBE system equipped with indium (In) and gallium (Ga) effusion cells and a nitrogen source. To grow the NWs, p-type Si (111) substrates were used. Initially, the unprepared substrate was transferred into the growth chamber and the substrate temperature was set to 950 °C for thermal treatment. Next, the substrate temperature was reduced to the actual growth temperature set at 600 °C and the nitrogen plasma source was ignited and the N flux was set to 0.4 sccm and 450 W. Finally, Ga and In sources were simultaneously opened. The beam equivalent pressures of Ga and In were equal to each other and set to 10⁻⁷ Torr. A series of experiments was carried out with growth time in the range from 2h to 3h.

The structural properties of the samples are examined using scanning electron microscopy SUPRA 25 (C. Zeiss SMT Ltd). Photoluminescence (PL) was excited with a helium–cadmium (He-Cd) metal-vapor laser with a wavelength of 325 nm. 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"). The measurements were performed at room temperature.

3. Results and discussion
Figure 1 shows SEM images of the InGaN nanowires grown in (a) 2 hours, (b) 2 hours 30 minutes, (c) 3 hours. The insertions below demonstrate plan-view images of corresponding samples.

![SEM images of InGaN NWs](image)

Figure 1. SEM images of InGaN NWs: (a) bird’s eye view of sample grown at 2h; (b) bird’s eye view of sample grown at 2h 30min; (c) bird’s eye view of sample grown at 3h. The insertions below demonstrate plan-view images of corresponding samples. The scale bars correspond to 200 nm.

As can be seen from figure 1(a), the NWs with an average length of 350 nm and an average diameter of 40 nm are grown for 2h. When the growth time was increased to 2h 30min, the upper part of NWs was transformed to nano-umbrellas. The average length of these NWs is 400 nm, and the average diameter is 50 nm. When the growth time was increased to 3h, the morphology of the NWs is changed significantly: a nanotubes layer with an average length 350 nm and average lateral size of 50 nm was formed; above the nanotube layer the InGaN nanostructures with developed morphology, an average length of 100 nm and an average diameter of 150 nm are grown. The nature of these morphology
transformations is most likely the local In-rich conditions (In accumulation) in the upper part of NWs [10].

Figure 2(a) shows the results of PL measurements of samples grown for 2h and 2h 30min. The insert in figure 2(a) demonstrates the dependence of the PL maxima on the growth time. Samples grown for 2h and 2h 30min exhibit similar PL spectra with an intense emission of about 590 nm and a weak emission band in the range from 700 to 900 nm, which is apparently explained by the In accumulation in the upper part of NWs. It should be noted that the small deviation of PL maxima of these samples is associated with the In fluctuation within the NWs.

Figure 2(b) demonstrates the PL spectrum of sample grown for 3h and the insert with the dependence of the integral intensity on the growth time. The main photoluminescence line is shifted by ~30 nm and located at 619 nm, which is apparently associated with the accumulation of In atoms in the upper part of NWs. Also, the addition emission line is detected at 421 nm, which indicates spinodal decomposition in grown NWs and is often observed in InGaN ternary compounds with 0.2-0.8 In mole fraction [3].

4. Conclusion
In the work, we studied the influence of the growth time on the formation of InGaN nanowires during plasma-assisted molecular beam epitaxy. It has been shown that the growth time can significantly affect the structural properties of NWs. In particular, under appropriate growth conditions, the growth for 2h leads to the formation of separated NWs, whereas the growth for 2h 30min and 3h leads to the formation of nanostructures such as nano-umbrellas. Optical studies have shown that samples grown for 2h and 2h 30min exhibit intense PL area of about 590 nm and a weak emission band in the range from 700 to 900 nm, while the sample grown for 3h exhibits two pronounced PL maxima at 421 and 619 nm, which indicated spinodal decomposition in these nano-umbrellas.

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