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Influence of the wet-chemical treatment on the optical and structural properties of core-shell InGaN nanowires

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Abstract. We study the influence of the shell in InGaN nanowires with spontaneously formed core-shell structure on their optical and morphological properties. It is shown that removing the shell from the initial nanowires induces the photoluminescence enhancement and changes their spectrum emission. Our research shows that etching the shell of these nanowires nanocrystals leads to their deviation from the vertical position.

Keywords: core-shell InGaN nanowires, etching of nanowires, photoluminescence enhancement, molecular beam epitaxy

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Влияние химического травления на оптические и структурные свойства нитевидных нанокристаллов InGaN ядро-оболочка

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Аннотация. В данной работе исследуется влияние химического травления нитевидных нанокристаллов InGaN со спонтанно-сформированной структурой ядро-оболочка на их оптические и структурные свойства. Продемонстрировано, что удаление оболочки у исходных нитевидных нанокристаллов приводит к увеличению интегральной и пиковой интенсивностей фотолюминесценции. Проведенные исследования показывают, что травлением оболочки данных нитевидных нанокристаллов, приводит к их отклонению от вертикального положения.

Ключевые слова: нитевидные нанокристаллы InGaN, ядро-оболочка, травление нитевидных нанокристаллов, усиление фотолюминесценции, молекулярно-пучковая эпитаксия

Финансирование: Экспериментальные образцы были синтезированы при поддержке Министерства науки и высшего образования Российской Федерации (госзадание № 0791-2023-0004). А.К. выражает благодарность Российскому научному фонду (грант № 24-12-00225) за поддержку экспериментов по исследованию оптических свойств синтезированных образцов. Исследования морфологических свойств выращенных образцов были выполнены при поддержке СПбГУ, шифр проекта 87465891. 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).

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Introduction

InGaN nanowires (NWs) are prospective candidates for fabrication ultra-high resolution micro- and nano- light-emitting devices over a wide range of wavelengths [1, 2]. InGaN NWs can be grown on lattice-mismatched substrates without forming structural defects, in particular on cheap and available Si substrates [3]. The synthesis of ternary InGaN compounds in the morphology of nanowires allows to obtain nanostructures with necessary emission wavelength in the visible range due to all possible variations in the In content [4, 5]. Moreover, orange and red-emitting InGaN nanocolumns with a high density of localized states due to large In fluctuation was shown to potential achieve high luminescence efficiency [6].

Recently, it was shown that InGaN NWs grown by plasma-assisted molecular beam epitaxy spontaneously formed with the core-shell structure [5], where the core contains 35–40 % indium ($In_{0.35-0.40}Ga_{0.65-0.60}N$), and the shell is almost pure GaN with 0–4 % In content. In this work, for the first time, we study the influence of the shell of the NWs on their photoluminescence (PL) properties. We determine that a decrease in the diameter of core-shell InGaN nanowires induced by the wet-chemical treatment leads to their deviation from the vertical position.

Materials and Methods

The InGaN NWs were grown directly on p-type Si(111) substrates using Riber Compact 12 molecular beam epitaxy setup, equipped with Ga, In effusion cells, and a nitrogen plasma source. Prior to growth, the substrates were heated up to a temperature of 920 °C and annealed for 20 min to remove the native oxide. The residual pressure in the chamber was $7 \cdot 10^{-8}$ Torr. Then, the substrate temperature was decreased and a plasma source was turned on. After reaching the temperature of 650 °C, gallium and indium shutters were opened simultaneously. The growth time of InGaN NWs was 20 hours.

We subjected the initial NW arrays to chemical treatment. Wet chemical etching was carried out in the solution KOH:H_2O (1:5) at a temperature of 75 °C to remove the GaN shell. The etching time varied from 20 to 80 seconds.

The morphology of the samples was studied by scanning electron microscopy (SEM Supra 25 Zeiss). The NW array was investigated with the use of micro-photoluminescence (PL) spectroscopy. The measurements were carried out on Jobin Yvon Horiba LabRAM HR 800 spectrometer equipped with a 100× magnification objective (N.A. = 0.9), camera and a stage with piezoelectric controllers for precise positioning of the laser beam. The source of optical excitation was a 532 nm laser, operating in a continuous mode. The optical system focuses the excitation into the beam with a diameter of about 1 μm.

Results and Discussion

Figure 1, *a* demonstrates schematical image of initial NWs (before etching). The typical SEM images of the grown NWs are presented in Fig. 1, *b*. It is clearly seen that the array contains both separated and coalesced NWs with an average height of about 2.7 μm. The spatially separated part of NWs have a core-shell structure with a shell thickness of about 20–30 nm and core diameter d_3 30–40 nm [5, 7]. The core-shell NW diameters are not constant and decrease from 100–130 nm (d_1) at the substrate surface up to 55–65 nm (d_2) at the tops of NWs.

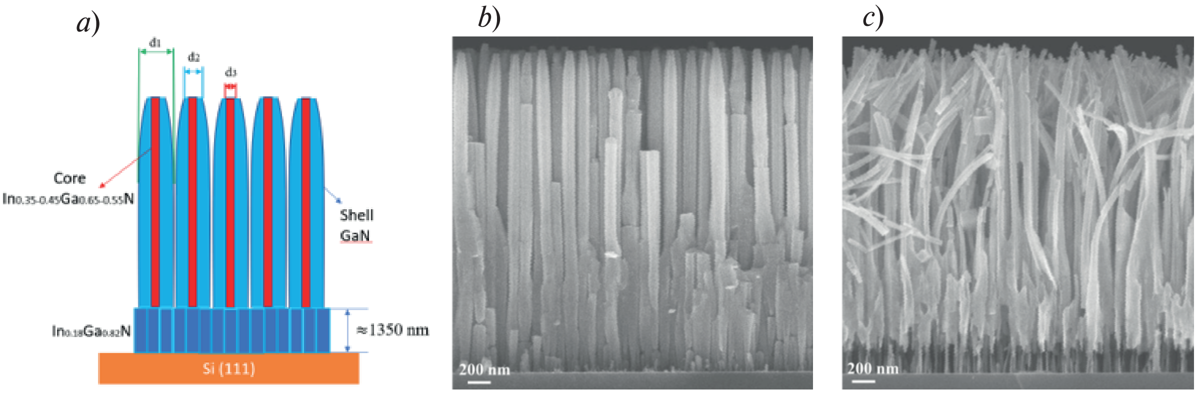

Fig. 1. Schematic image of the NW array before etching (*a*); typical cross-section SEM images of initial InGaN NWs (*b*) and after 50 s of etching (*c*)

The average values of d1 and d_2 decreased to 66 nm and 42 nm, respectively, after 60 seconds of etching (Fig. 1, *c*). Note that this value of d_2 is close to the core diameter d_3 30–40 nm. In addition, we observe a deviation of NWs from the growth direction in the range from 0 to 60 degrees that induced by the thinning of the shells.

Figure 2 demonstrates room-temperature PL spectra of sample with different etching times:

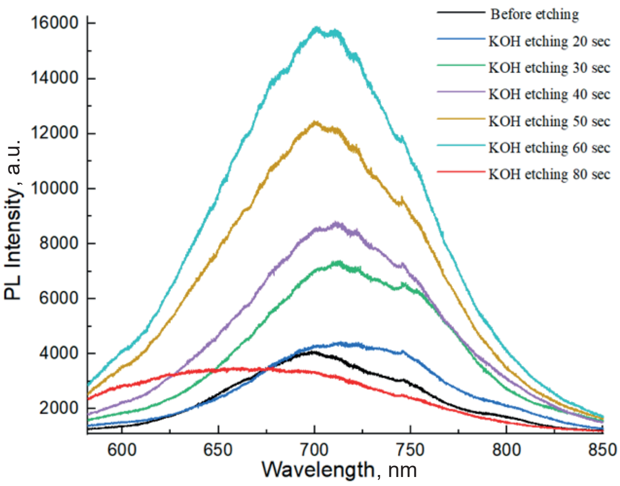

Fig. 2. Room-temperature PL spectra of the NW array with different etching times

The grown NWs have a noticeable PL in the range of 600–800 nm. We attribute the relatively broad PL spectrum to the compositional fluctuations of the indium content within the NWs. Figure 2 shows an increase in peak and integral PL intensity by a factor of 3.5 with increasing etching time from 0 to 60 seconds. Noticeable PL quenching and a shift of the PL peak to short wavelengths are observed after 80 seconds of etching that is explained by the sufficient core etching. Detailed mechanisms and reasons for the PL enhancement as a result of InGaN core-shell NWs etching will be presented in further studies.

Conclusion

This work should be useful for understanding the influence of shell on the luminescent properties of core-shell InGaN NWs. The PL intensity enhancement was observed in the case of etched NWs. Moreover, our results showed that a decrease in the diameter of core-shell NWs induced by treatment leads to their deviation from the vertical position. The results obtained can be useful for fabrication efficient LEDs based on molecular beam epitaxy growth InGaN NWs.

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