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Optical properties of single InGaN nanowires with core-shell structure

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Abstract. In this work, the photoluminescence of single InGaN NWs with a core-shell structure is investigated along their entire length at RT and 77 K. Multicolor emission, covering the spectral range from 380 to 650 nm, was obtained and described in details. Using the modified Vegard's law, the photoluminescence lines were correlated with the InGaN composition. Based on these results, conclusions about the structural properties and homogeneity of the InGaN NWs along their length were carried out.

Keywords: nanowires, core-shell structure, III-V semiconductors

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Оптические свойства одиночных нитевидных нанокристаллов InGaN со структурой «ядро-оболочка»

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Аннотация. В данной работе исследована фотолюминесценция одиночных InGaN ННК со структурой "ядро-оболочка" вдоль всей их длины при комнатной температуре и при 77 K. Получена и подробно описана многоцветная эмиссия, охватывающая спектральный диапазон от 380 до 650 нм. С помощью модифицированного закона Вегарда линии фотолюминесценции были соотнесены с составом InGaN. На основании этих результатов были сделаны выводы о структурных свойствах и однородности InGaN ННК по их длине.

Ключевые слова: нитевидный нанокристалл, структура "ядро-оболочка", III-V полупроводники

Финансирование: Образцы выращены при поддержке Министерства науки и высшего образования Российской Федерации (государственное задание № 0791-2020-0003). СЭМ исследования образцов проводились при поддержке Министерства науки и высшего образования Российской Федерации (государственное задание № 0791-2020-0002). Поддержка оптических измерений осуществлялась в рамках программы фундаментальных исследований Национального исследовательского университета «Высшая школа экономики» (НИУ ВШЭ).

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Introduction

InGaN ternary compounds are of considerable interest as materials for creation of solid-state light sources due to the possibility of regulating their emission energy from ultraviolet to near-infrared spectral range by changing the chemical composition [1]. A currently urgent problem is to obtain a compound with a high In content that has an emission energy lying in the green and red spectra. This will make it possible to create a highly efficient white light-emitting diode by mixing the base colors, without using phosphor-based wavelength conversion [2]. However, obtaining high crystal quality InGaN layers is complicated due to solid phase immiscibility of InGaN, which imposes restrictions on obtaining homogeneous layers with In concentration x_{In} = 0.2–0.8. This peculiarity is caused by a significant mismatch of lattice constants between InN and GaN [3].

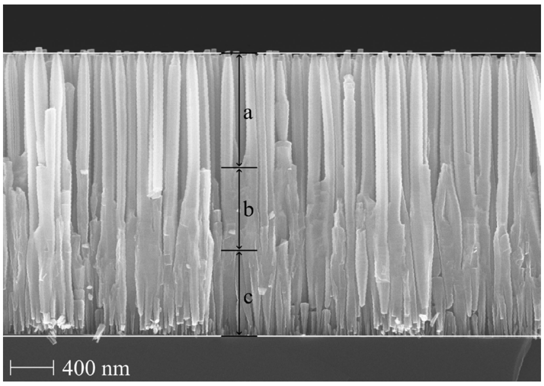

Fig. 1. Cross section SEM image of the InGaN NWs

In addition, the growth of InGaN layers with low density of structural defects is difficult due to the lack of substrates matched to the layer by lattice parameters. These problems can be solved by synthesizing InGaN nanowires (NWs) [4]. It has been shown that synthesis of InGaN NWs is possible over the entire range of chemical composition [5]. In particular, InGaN NWs grown by molecular beam epitaxy (MBE) on a Si substrate can exhibit a spontaneously formed core-shell structure [6]. It should be noted that the chemical composition and morphology of NWs significantly depend on the growth temperature [6]. In particular, the morphology can transform from coalesced nanostructures to spatially separated NWs with the spontaneously formed core-shell structure. However, the literature contains practically no data on the structural and optical properties of InGaN NWs, as well as their homogeneity along the entire length. The study of the optical properties of single NW allows us to estimate their structural and optical properties along the entire length, the presence or absence of localized states within the NWs, the distribution of chemical composition along the NWs.

In this paper photoluminescence of single InGaN NW grown on Si substrate and having core-shell structure is studied along its entire length at different temperatures.

Samples 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 a nitrogen plasma source. Growth was performed on an atomically clean Si surface under N-rich conditions at equal fluxes of In and Ga and a substrate temperature of 660 °C [7].

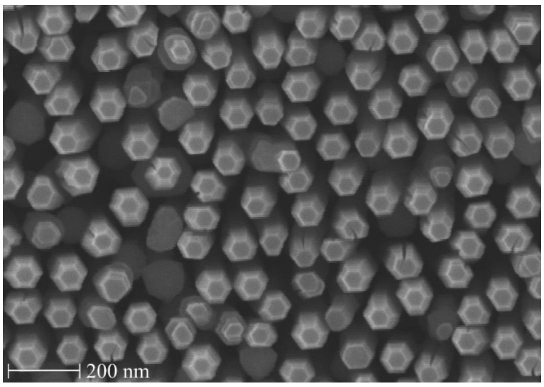

Fig. 2. Plan-view SEM image of the InGaN NWs

The morphology of the samples was studied by scanning electron microscopy (SEM Supra 25 Zeiss). The optical properties of the samples were studied by photoluminescence (PL) microscopy. PL of the samples was excited by a He-Cd laser (CW, λ = 325 nm), focused into the spot of 0.5 mm using a 50X Mitutoyo Plan Apo NUV HR objective. The positioning of the laser spot at a specific point on the NW was realized by precision objective movement and surface visualization with an external lamp and a video camera. The PL signal was detected using a standard lock-in technique by single-channel Si detector and Stanford Research Systems SR810 Lock-In Amplifier or by Symphony II CCD camera. The light was spectrally separated by SOL instruments MS 5204i monochromator with a 1200 gr/mm grating. The measurements were done at room temperature and at 77 K. For low temperature studies, the sample was mounted into the open-cycle nitrogen cryostat Janis ST-500.

Fig. 1 shows a cross section SEM image of initially grown InGaN NWs. This image shows that the NWs have a maximum length of about 2.6 μm and are morphologically heterogeneous. Along the length of the nanowires, three different regions can be seen: an upper part spatially separated from other NWs (*a*), a middle part coalesced with other NWs (*b*), and a lower part, tapering to the base (*c*). The upper part separated from other NWs have a spontaneously formed core-shell structure, as evidenced by our previous TEM measurements and wedge-shaped cracks at the NWs shown in Fig. 2 [7].

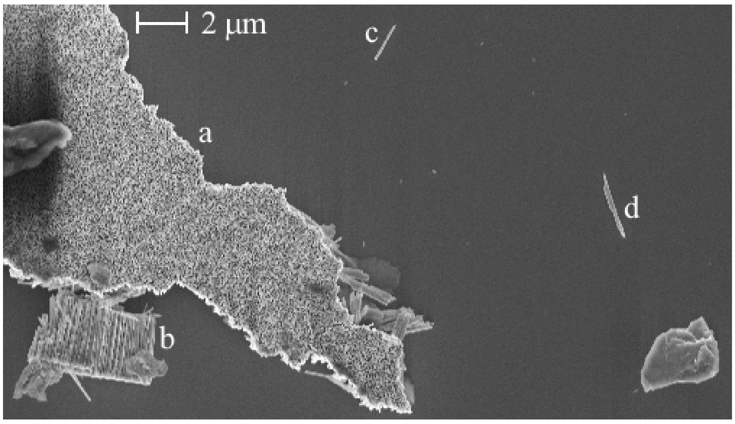

Fig. 3. SEM image of NWs transferred to Si (111) substrate

Results and Discussion

To study the optical properties of single NW, the NWs were transferred to a pure Si (111) substrate by the method described in [8]. Arrays of InGaN NWs were also transferred to the Si (111) substrate. Fig. 3 shows the SEM image of the transferred NWs. We observe an array of vertically oriented InGaN NWs (*a*), an array of horizontally oriented InGaN NWs (*b*), and individual NWs of different lengths (*c*, *d*). In this work, we studied NWs having a length closest to the maximum for the given sample (*d*). The length of the NW under study (*d*) is ~2.6 mm. The other NWs have smaller length (*c*) due to the damage during the transferring process and they were out of the scope of this work. Fig. 4 shows an optical image of the photoluminescence of NW (*d*) optically pumped by He-Cd laser. Along the length of the NW, we put three points, where its optical properties were investigated. As it is seen from Fig. 4, the photoluminescence color changes continuously from blue to red.

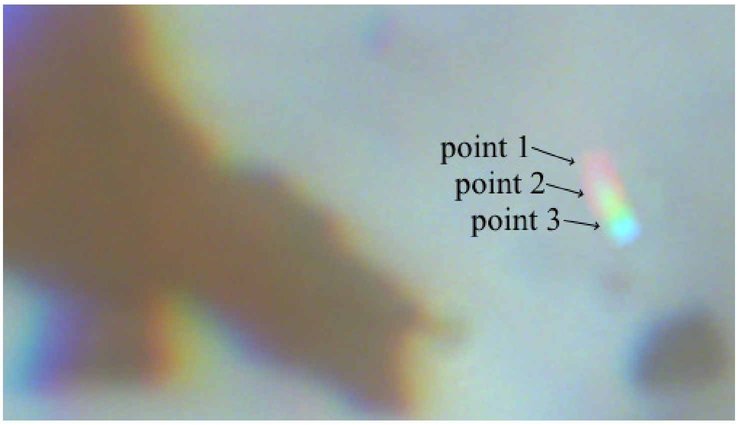

Fig. 4. Optical image of NW under optical pumping

Photoluminescence spectra of the single NW were obtained in three different points, at room temperature (Fig. 6) and at 77 K (Fig. 7). The first point corresponds to the top of the NW, the second to the center, and the third to the bottom. Visually, red color dominates in point 1. In the PL spectrum obtained at this point, the most intense peak has a spectral position of 656 nm (1.89 eV) at RT and 654 nm (1.90 eV) at 77 K. When the laser is focused in point 2, we see PL of different colors from yellow to green. The PL spectrum plot of the second point has two lines, with maxima at 507 nm (2.45 eV) and at 600 nm (2.07 eV) at RT (500 nm (2.48 eV) and 650 nm (1.91 eV) at 77 K), and a lower intensity peak located near 378 nm (3.28 eV) at RT and 77 K. At point 3, the PL has one well-defined peak located at 507 nm (2.45 eV) at RT and at 501 nm (2.48 eV) at 77 K.

It is possible to correlate spectral positions of PL lines with the structural properties of the InGaN NWs. The composition dependence (x) of the bandgap at RT can be described using modified Vegard's law including a quadratic term depending on a bowing parameter C:

$$E_g^{InGaN} = xE_g^{InN} + (1-x)E_g^{GaN} - x(1-x)C,$$

where E_g^{InN} is used as 0.7 eV [9], E_g^{GaN} is used as 3.43 eV [10], the bowing parameter C is 1.43 eV [9]. The graph of obtained dependence shown in Fig. 7. The dots indicate the spectral position of obtained PL peaks. The graph shows that there are three $In_xGa_{1-x}N$ compositions with In concentration of 4%, 26%, and 43% in the studied NW, with the In content increasing from the bottom of the NW to its top. In the bottom region, tapering to the base, the In content is ~ 4%. In the middle region, we observe coalescence of the NWs. In this part of the NW, the In content is ~ 26%. In the upper region, where NWs are separated from each other, and core-shell structure is formed the In content in the core is ~ 43%. We assume the alloy variation in the InGaN NWs is explained by a decrease in the effective growth temperature with increasing NW length. This assumption is confirmed by our previous work [6], where small temperature changes had a main influence on both the chemical composition and morphology of InGaN NWs. This is also confirmed by the model proposed in [11] that shows the presence of a nonuniform temperature distribution in GaN/InGaN NWs. Another reason is the huge lattice mismatch between InN and GaN resulting in internal strains and phase separation in InGaN with a high In content [12].

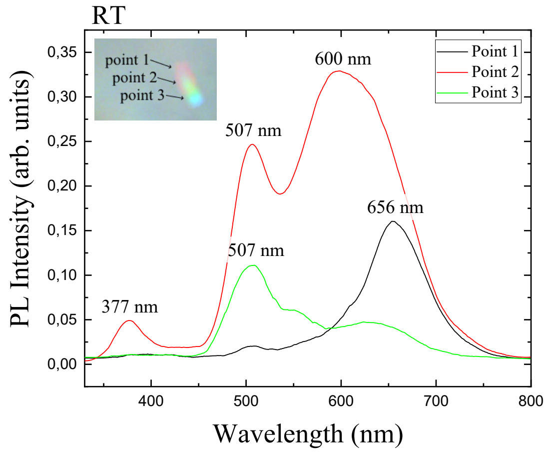

Fig. 5. Photoluminescence spectra obtained at different points of InGaN NW at room temperature

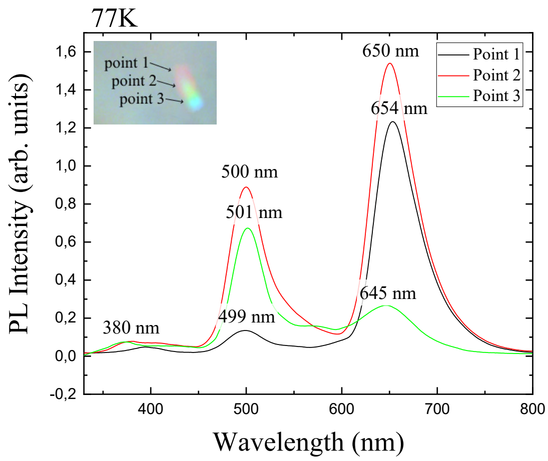

Fig. 6. Photoluminescence spectra obtained at different points of InGaN NW at 77 K

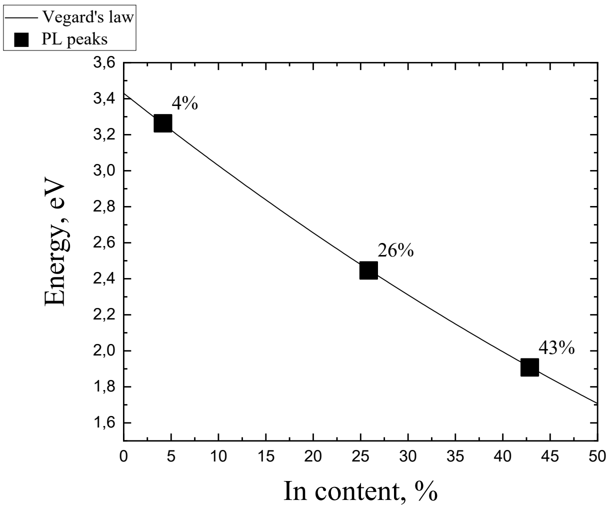

Fig. 7. Composition dependence of the InGaN bandgap at room temperature

Conclusion

To conclude, for the first time, multicolor emission from a single InGaN NW, covering the spectral range from 380 nm to 650 nm were obtained. Observing the sample under optical pumping, it is visually noticeable that the PL color consistently changes from red to blue. PL spectra for different points of the sample were obtained, and the spectral position of the PL intensity maxima was compared with the InGaN composition. Obtained results correlate well with TEM studies carried out earlier [6].

Acknowledgments

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