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MBE growth of wurtzite AlGaAs nanowires with zinc-blende insertions

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Abstract. In this work we present the experimental results on the molecular-beam epitaxy growth of wurtzite AlGaAs nanowires with nanoscale zinc-blende insertions on silicon substrate. Structural characterization confirmed the formation of zinc-blende nanoscale segments within the wurtzite nanowire matrix. Autocorrelation function measurements for emission at 710 nm have shown the characteristic dip at zero time delay, which indicates that the synthesized nanostructures are sources of single photons.

Keywords: Nanowires, AlGaAs, quantum dots, wurtzite, molecular-beam epitaxy

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Материалы конференции

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Рост методом МПЭ вюрцитных нитевидных нанокристаллов AlGaAs с включениями кубической кристаллографической фазы

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Аннотация. В работе представлены результаты экспериментальных исследований по росту методом молекулярно-пучковой эпитаксии вюрцитных AlGaAs нитевидных нанокристаллов с наноразмерными вставками кубической кристаллографической



фазы на поверхности кремния. Результаты исследований структурных свойств подтвердили формирование вставок кубической фазы в вюрцитную структуру ННК. Результаты измерений автокорреляционной функции для излучения на длине волны 710 нм продемонстрировали характерный провал при нулевой временной задержке, что характеризует синтезированные наноструктуры как источники одиночных фотонов.

Ключевые слова: Нитевидные нанокристаллы, AlGaAs, квантовые точки, вюрцит, молекулярно-пучковая эпитаксия

Финансирование: Работа выполнена при поддержке СПбГУ, шифр проекта 122040800254-4. Структурные свойства синтезированных образцов были изучены с помощью оборудования Междисциплинарного ресурсного центра по направлению «Нанотехнологии» Санкт-Петербургского государственного университета. Измерения автокорреляционной функции были выполнены с помощью оборудования ресурсного центра Оптические и лазерные методы исследования вещества Санкт-Петербургского государственного университета.

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Introduction

Nowadays, III-V semiconductor nanowires (NWs) are promising platform for nanoscale devices due to their unique optoelectronic, electrophysical, transport and other properties [1, 2]. Recent advances in NWs formation enable integration of quantum dots (QDs) in the NWs matrix, allowing precise control over QDs' size and position, which define physical properties of QDs. In turn, III-V QDs are particularly promising as single photon sources, essential components for quantum communication and computing [3]. Moreover, the unique morphology of NWs allows the synthesis of such nanostructures directly on the silicon surface [4, 5]. However, the growth of QDs in NWs based on different material systems may come with a set of challenges, due to the possible reservoir effect during catalytic growth or significant mismatch of lattice constants. It has been shown previously that varying the growth conditions during NWs synthesis, for example III/V flux ratio, enables reversible alternation of the catalyst droplet's contact angle and change the crystallographic phase of the growing NW segment [6]. Furthermore, it was demonstrated that bands offset between the wurtzite (WZ) and zinc-blende (ZB) crystal phases can lead to an indirect electron-hole pair recombination [7] and formation of QDs within the single material system for III-V NWs, including GaAs, InAs and InP NWs, which can be sources of single photons [8]. However, to expand the number of applications, it is necessary to increase the range of compositions for such nanostructures. Previously, we have shown the possibility of Au-catalyzed WZ AlGaAs NWs on silicon for the first time and demonstrated their potential as a promising material system for optoelectronic applications [9, 10]. Formation of crystal phase QDs in such NWs could further increase the range of applications based on this material.

In this work we demonstrate the results of experimental studies on the Au-catalyzed WZ AlGaAs NWs with ZB nanoscale insertions growth by molecular-beam epitaxy (MBE) on Si(111). It was shown for the first time that AlGaAs NWs could be sources of single photons according to autocorrelation function measurements results.

Materials and Methods

Growth experiments were carried out using a Riber Compact 21 MBE setup equipped with Ga and Al effusion cells, As cracker source and additional metallization chamber. The one-side polished *p*-type Si wafers with surface orientation (111) were used as the substrates for

MBE growth. Initially, native oxide layer was removed by wafer etching in aqueous solution of hydrofluoric acid (10:1). Then the substrate was loaded into the metallization chamber where it was annealed at 850 °C for 20 minutes to remove residual oxide layer. Then substrate temperature was decreased to 550 °C and a 1 nm layer of Au was deposited onto the surface. The substrate was kept at the same temperature for one minute to form gold droplets on substrate. On the next stage the substrate was transferred into the growth chamber without breaking ultra-high vacuum conditions. In growth chamber the substrate temperature was increased to 510 °C and As shutter was open. After stabilization of As flux, Al and Ga shutters were opened simultaneously for 20 minutes initiating AlGaAs NWs growth. In order to form ZB segments As flux was periodically decreased from $1 \cdot 10^{-5}$ Torr to $4 \cdot 10^{-6}$ Torr by source aperture modulation from 100% to 45% for 5 seconds. The aperture response time was ~1 second. Although As is not pumped instantly from growth chamber, the growth conditions on the substrate surface change rapidly from group V-stabilized to group III-stabilized and back [8]. The fluxes from the Ga and Al sources corresponded to AlGaAs layer growth rate of 1 monolayer per second at a Ga/Al ratio of 0.5/0.5 according to preliminary calibrations.

Morphological properties of grown samples were studied using a Carl Zeiss Supra 25 scanning electron microscopy (SEM) system. Structural properties of single NWs were studied using Jeol 2100 transmission electron microscopy (TEM) system. Correlation measurements were performed using the optical stand equipped with the cryostat, monochromator, lasers for excitation of nanostructures and single photon detectors.

Results and Discussion

Figure 1, *a* demonstrates the typical SEM image of grown AlGaAs NWs array on Si(111) substrate. As can be seen from the figure, AlGaAs NWs formed strictly in [111] direction which indicates their epitaxial bond with the substrate. The average height of the NWs is 1.4 μm . It should be noted that the NWs have a non-uniform diameter in height – 140 nm at the base and 30 nm at the top of the NWs. The average NWs surface density is $\sim 2 \cdot 10^7 \text{ cm}^{-2}$ which allows exciting a single NW using the laser with a spot area of 1 μm .

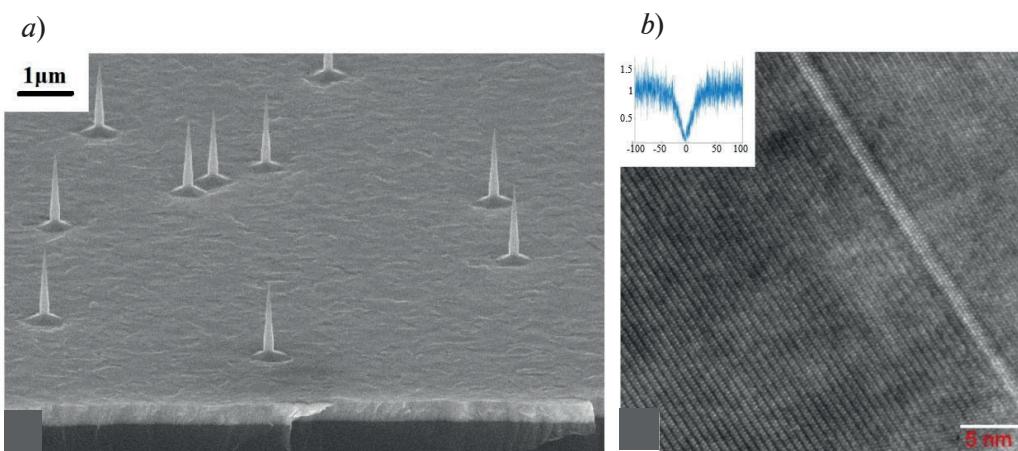


Fig. 1. Typical SEM image of grown AlGaAs NWs array on Si(111) substrate (*a*). Typical TEM image of the AlGaAs NW segment. The insert shows the results of the autocorrelation function measurements for emission at the wavelength of 710 nm (*b*)

Figure 1, *b* shows typical TEM image of the AlGaAs NW segment. It can be seen that NW segment demonstrates predominately WZ crystal structure with a presence of nanoscale ZB insertion. The height of the ZB insertion is ~ 2 nm. The insert to Fig. 1, *b* shows the results of the autocorrelation function measurements for photoluminescence emission at the wavelength of 715 nm, corresponding to AlGaAs [9]. The characteristic dip of the function at zero time indicates that the synthesized nanostructures are sources of single photons. It should be noted that the high noise level is associated with the short measurement time.



Conclusion

In summary, we demonstrated the MBE growth of WZ AlGaAs NWs with ZB insertions on the silicon surface using Au as a catalyst. The height of the ZB insertion studied using TEM is ~ 2 nm. The characteristic dip of the autocorrelation function at zero time indicates that the synthesized nanostructures are sources of single photons. These results open up new prospects for creating devices in the field of quantum technologies.

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