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Physical properties of InGaAs quantum dots in AlGaAs nanowires synthesized on silicon at different growth temperatures

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Abstract. In this work, we have studied the physical properties of InGaAs quantum dots (QDs) in AlGaAs nanowires (NWs) synthesized on silicon at different temperatures. The results of the studies have shown that, a decrease in the growth temperature leads to an increase in the mole fraction of indium in the InGaAs QD solid solution. In this case, the number of defects in QDs increases significantly due to an increase in the mismatch in the crystal lattices parameters of NWs and QDs.

Keywords: III-V compounds, silicon, nanowires, quantum dots, molecular beam epitaxy

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Материалы конференции
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Физические свойства InGaAs квантовых точек в AlGaAs нитевидных нанокристаллах, ситезировнных на кремнии при разных ростовых температурах

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Аннотация. В работе представлены результаты экспериментальных исследований физических свойств InGaAs КТ в AlGaAs ННК, синтезированных на кремнии при различных температурах. Результаты исследований показали, что, как и ожидалось, снижение температуры роста приводит к увеличению мольной доли индия в твердом растворе InGaAs КТ. При этом количество дефектов в КТ значительно возрастает из-за увеличения несоответствия параметров кристаллических решеток ННК и КТ.

Ключевые слова: соединения III-V, кремний, нитевидные нанокристаллы, квантовые точки, молекулярно-пучковая эпитаксия

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Introduction

Nowadays, semiconductor direct-gap nanostructures based on III-V compounds attract increased interest of researchers due to their unique optical, mechanical, and electrical properties [1]. Moreover, the possibility of these objects integration with silicon technology has opened up wide prospects for creating applications based on them [2]. Of particular interest are combinations of III-V materials of different dimensions on the silicon surface, for example, nanowires (NWs) with quantum dots (QDs) [3]. The development of modern methods of synthesis, such as molecular-beam epitaxy (MBE), makes it possible to controllably synthesize QDs in NWs with specified sizes and surface density. In our previous works, we successfully synthesized AlGaAs NWs with GaAs QDs on a silicon surface for the first time, and it was shown that a change in the QDs growth time leads to a shift of the corresponding photoluminescence (PL) line [4]. In addition, the results of autocorrelation measurements have shown that the formed QDs are sources of single photons, which makes them promising for quantum cryptography and other applications [4]. Nevertheless, to increase the range of applications based on such nanostructures, e.g., for telecom wavelengths, it is necessary to expand the range of QDs materials for shifting emission of single photon sources to a longer wavelength region towards 1.3–1.5 μm. At present, there is a number of papers reporting on the synthesis and properties of InGaAs QDs in GaAs NWs [5–7]. However, in most cases such nanostructures are grown on GaAs substrates and exhibit PL spectra only at low temperatures due to the low localization of charge carriers. In this case, the long-wavelength shift of the PL spectra is limited by a desorption of indium adatoms from the surface at the growth temperature of GaAs NWs (500-600 °C). In [8], we synthesized AlGaAs NWs with InGaAs QDs on a silicon surface, for the first time, at growth temperatures of 320–510 °C. Due to the fact that the band gap of AlGaAs exceeds that of GaAs, the localization of charge carriers in this case has a higher value. However, PL spectra at room temperature in the region of 1.3 μm were observed only from structures synthesized at high temperatures. Therefore, we made the assumption that as the growth temperature decreases, the number of indium adatoms embedded in QDs increases. In turn, this leads to an increase in the mismatch in lattice constants between QDs and NWs, thereby may leads to the appearance of structural defects in QDs.

In this paper we present the results of experimental studies of the structural properties of AlGaAs NWs with InGaAs QDs synthesized by MBE on a silicon surface at different temperatures.

Materials and Methods

Growth experiments were carried out using Riber 21 MBE setup equipped with the effusion Ga, Al, In and As_4 cells as well as a separate metallization chamber for Au deposition, which allows one to transfer the samples to the growth chamber with no vacuum brake. The growth procedure is described in details in [8]. Prior to the growth the fabrication of Au catalyst arrays was performed. First, the oxide-free Si surface was achieved by wet chemical treatment in $HF{:}H_2O$ solution and then annealing at a temperature of 950 °C in the metallization chamber. Thereafter, the substrate temperature was decreased to 550 °C and deposition of ~1 nm thick Au film followed by 1 min exposure time to improve the droplets homogeneously. Then, the substrates were transferred to the growth chamber. Upon reaching the growth temperature, which was set at 400 °C and 510 °C in our experiments, the formation of AlGaAs NWs with nominal Al content $x = 0.3$ was initiated by simultaneous opening of Al, Ga, As sources. NWs growth processes lasted 25 min, whereas the formation of InGaAs insertion was realized at 20 min by short-term (20 s) switching of Al to In fluxes. The material fluxes from all sources were constant throughout the growth process and were corresponded to the growth rates of planar layers 0.5, 0.3 and 0.5 monolayers per second (ML/S) for Ga, Al and In fluxes, respectively, according to the preliminary calibrations on a separate substrate. The NWs growth process was controlled *in situ* by reflection high-energy diffraction (RHEED). It should be noted, that RHEED revealed the wurtzite crystal phase of AlGaAs NWs throughout their growth except at the very beginning (~1 min).

The samples obtained were characterized by scanning electron microscopy (SEM), transmission electron microscopy (TEM) combined with energy-dispersive *X*-ray (EDX) analysis. Optical properties of NWs were studied with the use of the macro-photoluminescence technique at room temperature.

Results and Discussion

Figure 1 shows the SEM images of the samples grown at: (*a*) 510 °C; (*b*) 400 °C. As can be seen from the figure, a decrease in the growth temperature leads to a change in the geometric parameters of NWs: a decrease in the height and an increase in the diameter. The reason for that is a significant decrease in the migration speed of Al adatoms over the surface, which restricts the migration of Ga adatoms. It should be noted that NWs are mainly formed in <111> direction which indicates their epitaxial relation to Si(111) substrate.

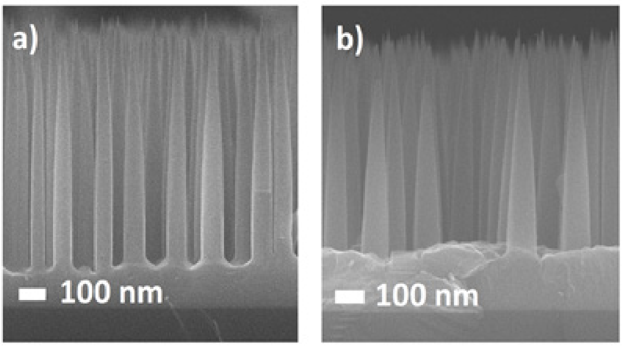

Fig. 1. Typical SEM images of AlGaAs NWs with InGaAs QDs grown on silicon at growth temperature of: (*a*) 510 °C; (*b*) 400 °C

Figure 2 shows typical PL spectra measured at room temperature from InGaAs QDs in AlGaAs NWs grown at 510 °C and 400 °C. It can be seen from the figure that the sample grown at 510 °C exhibit PL spectra at room temperature in a wide wavelength range from 850 to 1300 nm. Such a wide range of emission is associated with both the inhomogeneity of the QDs sizes in the NWs array and with the formation of several InGaAs nanoobjects in the AlGaAs NW bodies [8]. It is important to note, that the observation of PL spectra up to room temperature indicates the high optical quality of the structures. In turn, the intensity of the PL spectrum from the sample grown at 400 °C is at the noise level or is completely absent at room temperature. This may indicate a significant number of nonradiative recombination centers in QDs.

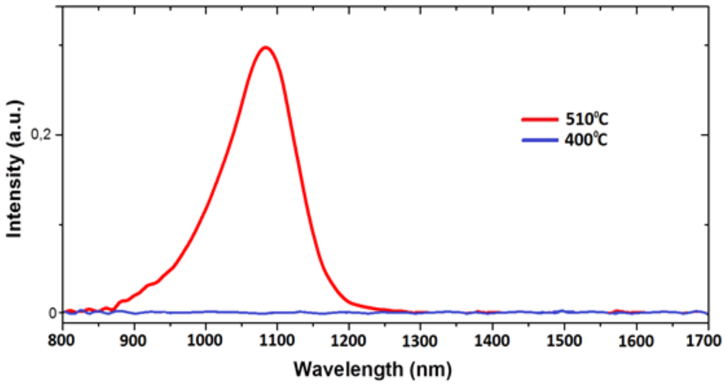

Fig. 2. Typical PL spectra measured at room temperature from InGaAs QDs in AlGaAs NWs grown on silicon at 510 °C and 400 °C

Typical TEM images of dispersed on a carbon grid single InGaAs QD in AlGaAs NW grown at the substrate temperatures of 510 °C and 400 °C are shown in figures 3, *a* and 3, *b*, respectively. It is seen that the QD grown at 510 °C contains only a few monolayers of the zinc blende structure in the wurtzite crystallographic phase of QD, which is associated with the features of the AlGaAs NWs synthesis. Studies of the grown at 510 °C InGaAs QD composition by the EDX method showed that the molar fraction of indium in QD is ~ 22%. As expected, the grown at 400 °C QD is highly dislocated and contains a large number of structural defects. The results of studies by the EDX method showed that the mole fraction of In in the solid solution of this QD is ~ 50%. Thus, the mismatch in the lattice constant parameters in the latter case increases significantly, leading to an increase in the number of nonradiative recombination centers in QDs, which is in good agreement with our pre-assumptions. One of the possible ways to overcome this problem can be the synthesis of small-size QDs. According to [9], when synthesized in a mismatched QD material with a size smaller than the critical one, the QD does not contain any structural defects. The results of corresponding studies will be presented in the following works.

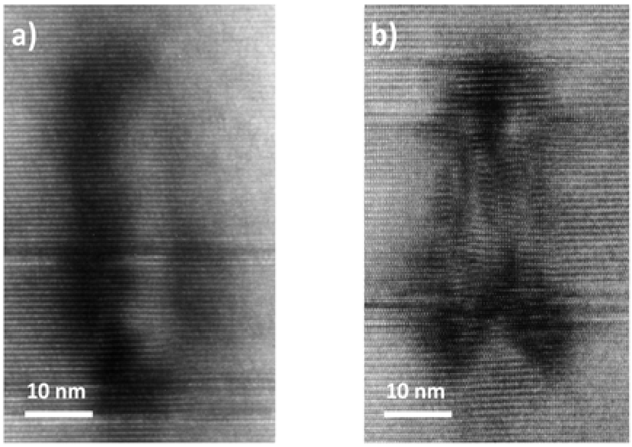

Fig. 3. Typical TEM images of single InGaAs QD in AlGaAs NW grown at the substrate temperatures of: 510 °C (*a*); 400 °C (*b*)

Conclusion

To conclude, we have studied the physical properties of InGaAs QDs in AlGaAs NWs synthesized on silicon at different temperatures. The results of the studies have shown that, as expected, a decrease in the growth temperature leads to an increase in the mole fraction of indium in the InGaAs QD solid solution. In this case, the number of defects in QDs increases significantly due to an increase in the mismatch in the crystal lattices parameters of NWs and QDs. The results of the proposed method for solving this problem will be presented in the following works.

REFERENCES

1. **Dubrovskii V. G., Cirlin G. E., Ustinov V. M.,** Semiconductor nanowhiskers: synthesis, properties, and applications, Semiconductors. 43 (12) (2009) 1539–1584.

2. **Mertensson T., Svensson C. P. T., Wacaser B. A., Larsson M. W., Seifert W., Deppert K., Gustafsson A., Wallenberg L. R., Samuelson L.,** Epitaxial III–V nanowires on silicon, Nano Letters. 4 (10) (2004) 1987–1990.

3. **Dalacu D., Mnaymneh K., Lapointe J., Wu X., Poole P. J., Bulgarini G., Zwiller V., Reimer M. E.,** Ultraclean emission from InAsP quantum dots in defect-free wurtzite InP nanowires, Nano Letters. 12 (11) (2012) 5919–5923.

4. **Leandro L., Gunnarsson C. P., Reznik R., Jцns K. D., Shtrom I., Khrebtov A., Kasama T., Zwiller V., Cirlin G., Akopian N.,** Nanowire quantum dots tuned to atomic resonances, Nano Letters. 18 (11) (2018) 7217–7221.

5. **Tatebayashi J., Kako S., Ho J., Ota Y., Iwamoto S., Arakawa Y.,** Growth of InGaAs/GaAs nanowire-quantum dots on AlGaAs/GaAs distributed Bragg reflectors for laser applications, Journal of Crystal Growth. 468 (2017) 144–148.

6. **Heiss M., Ketterer B., Uccelli E., Morante J. R., Arbiol J., Morral A. F.,** Circumventing the Miscibility Gap in InGaN Nanowires Emitting from Blue to Red, Nanotechnology. 22 (19) (2011) 195601.

7. **Makhonin M., Foster A., Krysa A., Fry P., Davies D., Grange T., Walther T., Skolnick M., Wilson L.,** Homogeneous array of nanowire-embedded quantum light emitters, Nano Letters. 13 (3) (2013) 861–865.

8. **Reznik R. R., Ilkiv I. V., Kotlyar K. P., Gridchin V. O., Bondarenko D. N., Lendyashova V. V., Ubyivovk E. V., Dragunova A. S., Kryzhanovskaya N. V., Cirlin G. E.,** Molecular-Beam Epitaxy Growth and Properties of AlGaAs Nanowires with InGaAs Nanostructures, Physica Status Solidi-Rapid Research Letter. (2022) 2200056.

9. **Tonkikh A. A., Tsyrlin G. E., Talalaev V. G., Novikov B. V., Egorov V. A., Polyakov N. K., Samsonenko Yu. B., Ustinov V. M., Zakharov N. D., Werner P.,** Room-temperature 1.5–1.6 μm photoluminescence from InGaAs/GaAs heterostructures grown at low substrate temperature, Semiconductors. 37 (12) (2003) 1406-1410.

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