Formation of InAs nanoislands on silicon surfaces and heterostructures based on them

© I.V. Ilkiv[1,2], V.V. Lendyashova[1,3], B.B. Borodin[3], V.G. Talalaev[4], T. Shugabaev[1], R.R. Reznik[2], G.E. Cirlin[1,5]

[1] Alferov University,
194021 St. Petersburg, Russia
[2] St. Petersburg State University,
199034 St. Petersburg, Russia
[3] Ioffe Institute,
194021 St. Petersburg, Russia
[4] Martin Luther University Halle-Wittenberg,
06108 Halle, Germany
[5] ITMO University,
197101 St. Petersburg, Russia
E-mail: fiskerr@ymail.com

Received May 5, 2023
Revised June 29, 2023
Accepted July 6, 2023

Experimental results of studying the InAs islands formation of silicon surface by molecular beam epitaxy are presented. It has been found that, InAs islands with both bimodal and uniform size distributions can be formed depending on the Si surface relief and the presence of nanopits. The possibility of fabricating heterostructures with InAs quantum dots demonstrating photoluminescence in the region of 1.65 μm, was showed.

Keywords: Quantum dots, molecular beam epitaxy, semiconductors, silicon, heterostructures.

1. Introduction

The integration of $A^{III}B^{V}$-based semiconductor heterostructures on silicon is of significant interest due to the prospects for creating optoelectronic integrated circuits based on them with higher performance and low power consumption [1,2]. Despite the progress achieved in the development of semiconductor wafer bonding methods [3], monolithic integration by direct synthesis of $A^{III}B^{V}$ nanostructures on silicon substrates continues to be of high scientific interest. To date, certain successes have already been achieved in the production of $A^{III}B^{V}$ heterostructures on silicon, including the creation of injection lasers with continuous operation at room temperatures on their basis [4]. However, the technology for growing buffer layers with high crystalline quality, primarily with a low density of threading dislocations and the absence of antiphase domains, is still very resource-intensive and represents a high technological complexity. In particular, the main problems are the need to form thick (of the order of $3-5\,\mu$m) buffer layers with a gradient composition [5,6] and elastically stressed superlattices [7,8], the use of Si(100)-substrates with a deviation of $4-6^\circ$ in the [110] direction [7], etc. In this regard, research aimed mainly at the use of structures with an active region based on arrays of self-organizing quantum dots (QDs), which are less sensitive to defects, is becoming increasingly relevant. At the same time, the unique properties of quantum dots associated with the relaxation of elastic stresses can also be used to create heterostructures with $A^{III}B^{V}$ QDs monolithically integrated in a silicon layer [9–12].

This study is devoted to investigating the processes of formation of InAs nanoislands on a silicon surface using the molecular beam epitaxy (MBE) method, as well as their subsequent overgrowth with the aim of forming on their basis heterostructures with InAs QDs integrated in the silicon layer.

2. Experimental methods

The nanostructures were synthesized using an Compact 21 EB200 MBE system by Riber equipped with effusion sources for the growth of $A^{III}B^{V}$ compounds, as well as electron beam evaporators for Si and Ge, which in turn made it possible to synthesize heterostructures in a single technological cycle. Si(100)-wafers with a misorientation of 4° were used as substrates. First, liquid-phase chemical etching of the substrates was carried out, followed by thermal annealing in the growth chamber at 900°C to remove the thin oxide layer and to form a buffer Si-layer with a thickness of 50 nm at 600°C. Then, at a substrate temperature of 400°C, self-organizing growth of InAs islands was performed. The In deposition rate was 0.1 Å/s, and the flux ratio was V/III ~ 20. After this, a technological pause was made to pump out residual arsenic from the growth chamber and the cover Si-layer was formed. Upon completion of the growth, samples with InAs QDs in silicon were cooled to room temperature and unloaded to study the optical properties using low-temperature photoluminescence (PL). The measurements were carried out in a closed-cycle cryostat cooled to

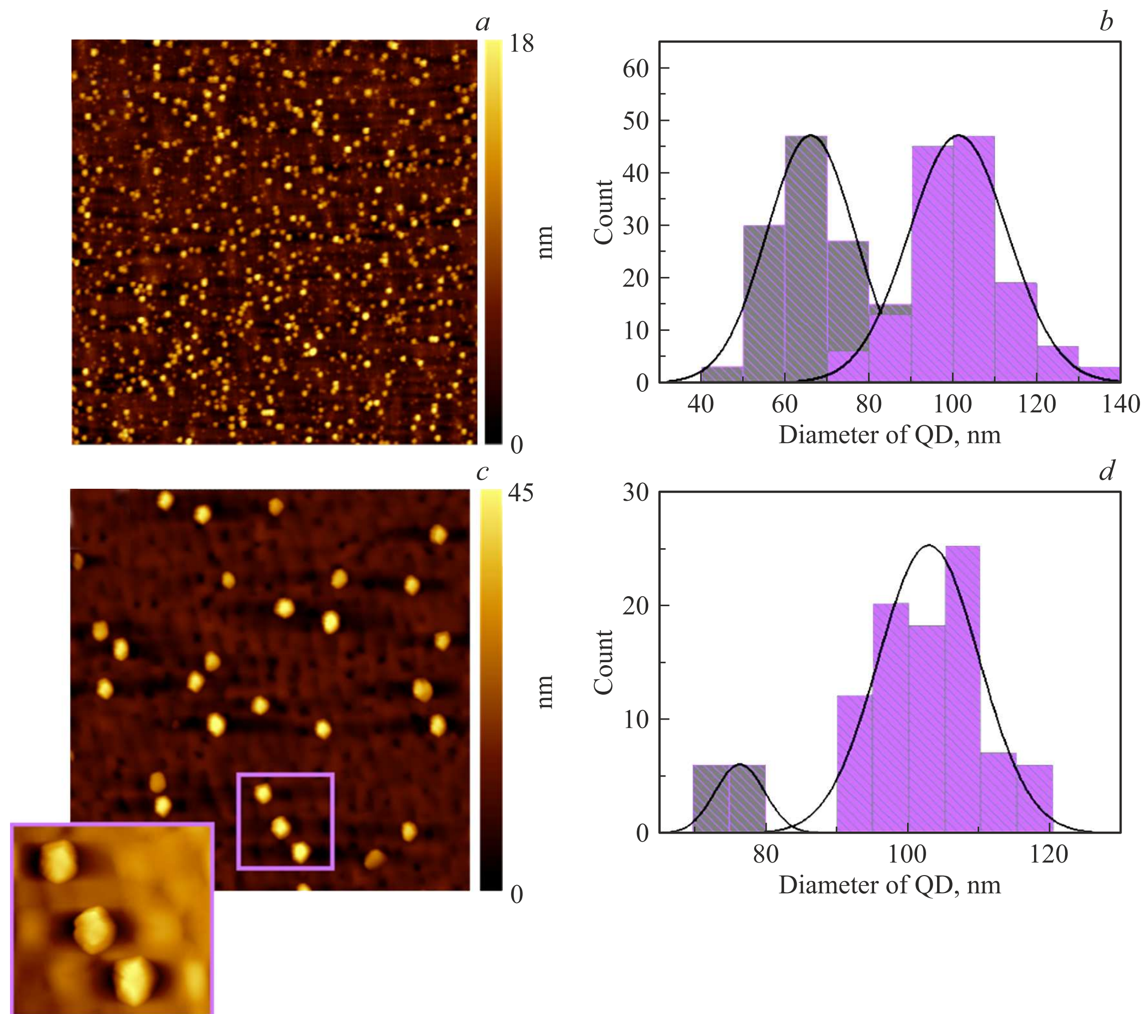

Figure 1. AFM-images ($5 \times 5\,\mu$m) of Si-surface after deposition of an InAs-layer with a thickness of 0.6 ML (*a*) and 1 ML (*c*). Bar charts of size distribution of InAs nanoislands formed after deposition of an InAs layer with a thickness of 0.6 ML (*b*) and 1 ML (*d*).

10 K. Pumping was implemented by laser radiation at a wavelength of 405 nm, with a radiation power density of $\sim 30\,\mathrm{W/cm^2}$. PL spectra were recorded using an InGaAs photodetector. The width of the entrance and exit slits of the monochromator was 1–1.5 mm. The signal accumulation time was 0.5 s.

To study the morphological features of InAs islands, samples without the Si cover layer were synthesized. For this purpose, a Ntegra Aura atomic force microscope (AFM) was used, operating in semi-contact mode using silicon probes (HANC, TipsNano) with a tip curvature radius of < 10 nm.

3. Results and discussion

To date, studies have already been presented on the synthesis of InAs islands on the surface of silicon wafers, where the effect of the main growth parameters (growth temperature, flux ratio, etc.) on the processes of island formation was studied, however, the effect of the Si surface morphology is known to a lesser extent. In addition, only in a few studies [11,12] the authors have succeeded to form heterostructures of sufficiently high crystalline quality that exhibit photoluminescence. It should be noted that one of the possible reasons for the low optical quality of such heterostructures may be related to the formation of InAs islands with relatively large sizes, which are often dislocated [10]. Therefore, in this study, it was decided to first investigate the possibility of forming InAs islands with relatively small sizes.

A characteristic feature of the formation of islands in mismatched systems of materials is a change in diffraction patterns. Analysis of reflection high-energy electron diffraction (RHEED) images *in situ* has shown that the point reflections in the RHEED pattern of Si(100) with the (2×1) superstructure begin to occur after the deposition of ~ 0.7 monolayers (ML) of InAs. This is indicative of the fact that the formation of InAs islands under the chosen growth conditions takes place within the framework

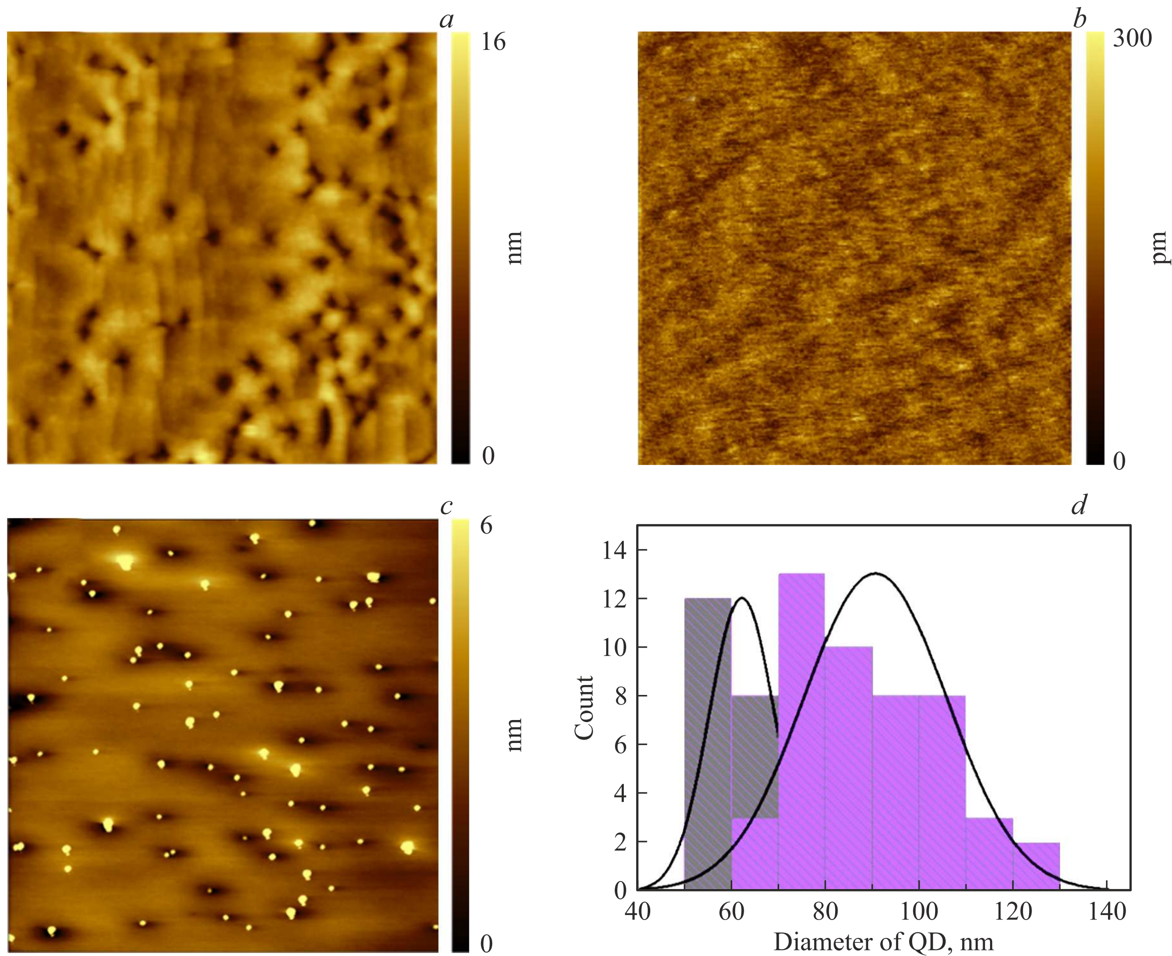

Figure 2. AFM-images ($1 \times 1\,\mu$m) of the silicon buffer layer surface before (*a*) and after (*b*) thermal annealing. AFM-image ($5 \times 5\,\mu$m) of InAs nanoislands formed on a smooth Si surface after deposition of an InAs layer with a thickness of 0.6 ML (*s*) and the corresponding bar chart of islands distribution by size (*d*).

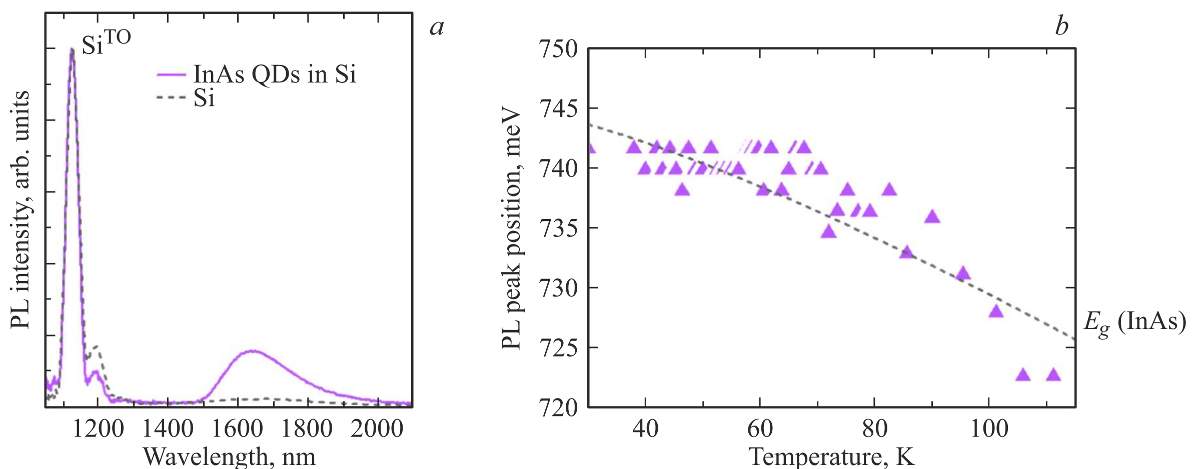

Figure 3. PL spectrum of InAs QDs integrated in the silicon layer (0.6 ML) and the original Si-substrate (*a*). The spectrum of silicon contains peaks of interband transitions with phonons involvement, the most intense of which is marked as SiTO in the figure. Graph of the position of the photoluminescence peak of the heterostructure as a function of temperature (*b*). The triangular points on the graph represent the experimental values, the dashed line shows values calculated using the Varshney formula at $\alpha = 3.2 \cdot 10^{-4}\,\text{eV} \cdot \text{K}^{-1}$, $\beta = 93\,\text{K}$.

of the Volmer–Weber mechanism [13]. Figure 1, *a* shows AFM-images of the surface of samples with a thickness of deposited material equal to 0.6 ML. It can be seen that as a result of the deposition of InAs, nanoscale islands of a hemispherical shape were formed on the silicon surface. It should be noted that the islands had a bimodal distribution by size. Relatively small islands with a transverse size of 68 ± 15 nm and a height of 2–4 nm had a surface density of $4 \cdot 10^{10}\,\text{cm}^{-2}$. The dimensions of large islands were 100 ± 10 nm (transverse size) and 6–12 nm (height), and their density was $2.3 \cdot 10^{9}\,\text{cm}^{-2}$. With an increase in the effective thickness of the deposited material to 1 ML (see Figure 1, *c*), the average size of nanoislands increased, while the typical bimodal size distribution remained the same. In addition, the shape of the islands has changed and acquired a pronounced faceting (see the inset to Figure 1, *c*). Also, it should be noted that the ratio of small islands number to large islands number has decreased significantly. This fact may be due to the coalescence of small islands during the InAs deposition process.

The formation of islands with a bimodal distribution often occurs in InAs/InP, InAs/GaAs material systems at the initial stage of growth and, as a rule, is associated with the presence of energy barriers [14,15]. In our case, an assumption was made that one of the possible reasons for the appearance of energy barriers may be related to the presence of nanowells on the surface of the silicon buffer layer formed during the process of silicon homoepitaxy [16,17]. Apparently, the nucleation of nanoislands with smaller sizes occurs predominantly in wells, and large islands are formed on atomically smooth areas of the silicon surface due to a larger area of material collection, i. e. greater diffusion flow of indium adatoms into the forming nanoisland. To study this process in more detail, experiments were carried out on the synthesis of InAs nanoislands on a smooth Si-surface. To produce a smooth surface free from nanowells, the substrates with the formed Si buffer layer were thermal annealed *in situ* at a temperature of 1100°C for 20 min (Figure 2, *b*). As a result of the experiments, it was demonstrated that the bimodal distribution of islands by size became less pronounced (Figure 2, *d*). At the same time, the average size of the islands increased, and the density decreased by an order of magnitude. Thus, InAs nanoislands formed on a smooth Si surface are more uniform in size.

To carry out optical measurements, heterostructures with InAs islands completely integrated in silicon by epitaxial overgrowth were synthesized. The cover S-layer was formed in two stages: (1) — low-temperature (400°C) deposition of a S-layer with a thickness of 10 nm after the formation of InAs islands in order to fix their position and prevent the evaporation of dot material from the surface, (2) — growth of a 20 nm thick S-layer with a higher crystalline quality at an increased (500°C) substrate temperature. PL spectra of heterostructures with InAs QDs measured at a temperature of 10 K (the effective thickness of the deposited material is 0.6 ML) are shown in Figure 3, *a*. The spectrum exhibits a low-intensity peak in the region of 1.65 μm, which was not detected on the reference samples with a Si buffer layer and is apparently due to the photoluminescence of InAs QDs. It should be noted that this peak is quite wide and asymmetrical. In this case, the presence of a long-wavelength shoulder can be associated with the bimodal size distribution of InAs QDs. In turn, the PL signal from heterostructures with InAs QDs with a deposited material thickness of 1–2 ML could not be detected, which confirms the defectiveness of relatively large InAs QDs.

Figure 3, *b* shows the experimentally obtained temperature dependence of the position of the PL peak maximum on the InAs QD and the InAs band gap calculated by the Varshney formula [18] using $\alpha = 3.2 \cdot 10^{-4}\,\text{eV} \cdot \text{K}^{-1}$ and $\beta = 93$ K coefficients. It can be seen that the temperature dependence of the PL maximum position reproduces the change in the InAs band gap in the range of 0–100 K. In addition, the obtained dependence is well consistent with the experimental data of other studies devoted to the investigation of optical properties of InAs QDs [19–21].

4. Conclusion

As a result of the studies performed, the possibility of growing InAs nanoislands on the silicon surface was demonstrated. It has been demonstrated that, depending on the relief and the presence of nanowells on the Si-surface, InAs islands with both bimodal and uniform size distributions can be formed. Using the two-stage overgrowth with silicon, the possibility has been demonstrated of forming heterostructures with InAs quantum dots integrated in silicon, which can exhibit photoluminescence in the region of 1.65 μm.

Funding

This study was supported financially by the Ministry of Science and Higher Education of the Russian Federation (0791-2023-0004). Optical measurements were conducted within research grant of SPb State University No. 94033852. The investigation of morphology of InAs-islands was supported by the Ministry of Science and Higher Education of the Russian Federation, research project No. 2019-1442 (research topic code FSER-2020-0013).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] D. Thomson, A. Zilkie, J. E. Bowers, T. Komljenovic, G.T. Reed, L. Vivien, D. Marris-Morini, E. Cassan, L. Virot, J.M. Fédéli, J.M. Hartmann, J.H. Schmid, D.X. Xu, F. Boeuf, P. O'Brien, G.Z. Mashanovich, M.N. Nedeljkovic. J. Optics, **18** (7), 073003 (2016).

[2] X. Chen, M. M. Milosevic, S. Stanković, S. Reynolds, T.D. Bucio, K. Li, D.J. Thomson, F. Gardes, G.T. Reed. Proc. IEEE, **106** (12), 2101 (2018).
[3] A. Sakanas, E. Semenova, L. Ottaviano, J. Mørk, K. Yvind. Microelectron. Eng., **214**, 93 (2019).
[4] C. Jiang, H. Liu, J. Wang, X. Ren, Q. Wang, Z. Liu, B. Ma, K. Liu, R. Ren, Y. Zhang, S. Cai, Y. Huang. Appl. Phys. Lett., **121** (6), 061102 (2022).
[5] H. Tanoto, S.F. Yoon, K.L. Lew, W.K. Loke, C. Dohrman, E.A. Fitzgerald, L.J. Tang. Appl. Phys. Lett., **95** (14), 141905 (2009).
[6] W.K. Loke, Y. Wang, Y. Gao, L. Khaw, K.E.K. Lee, C.S. Tan, E.A. Fitzgerald, S.F. Yoon. Mater. Sci. Semicond., **146**, 106663 (2022).
[7] B. Kunert, Y. Mols, M. Baryshniskova, N. Waldron, A. Schulze, R. Langer. Semicond. Sci. Technol., **33** (9), 093002 (2018).
[8] J.C. Norman, D. Jung, Z. Zhang, Y. Wan, S. Liu, C. Shang, R.W. Herrick, W.W. Chow, A.C. Gossard, J.E. Bowers. IEEE J. Quant. Electron., **55** (2), 1 (2019).
[9] G.E. Cirlin1, V.G. Dubrovskii, V.N. Petrov, N.K. Polyakov, N.P. Korneeva, V.N. Demidov, A.O. Golubok, S.A. Masalov, D.V. Kurochkin, O.M. Gorbenko, N.I. Komyak, V.M. Ustinov, A.Yu. Egorov, A.R. Kovsh, M.V. Maximov, A.F. Tsatsul'nikov, B.V. Volovik, A.E. Zhukov, P.S. Kop'ev, Zh.I. Alferov, N. N. Ledentsov, M. Grundmann, D. Bimberg. Semicond. Sci. Technol., **13** (11), 1262 (1998).
[10] M. Benyoucef, M. Usman, J.P. Reithmaier. Appl. Phys. Lett., **102** (13), 132101 (2013).
[11] R. Heitz, N.N. Ledentsov, D. Bimberg, A.Yu. Egorov, M.V. Maximov, V.M. Ustinov, A.E. Zhukov, Zh.I. Alferov, G.E. Cirlin, I.P. Soshnikov, N.D. Zakharov, P. Werner, U. Gösele. Appl. Phys. Lett., **74** (12), 1701 (1999).
[12] C. Bru-Chevallier, A. El Akra, D. Pelloux-Gervais, H. Dumont, B. Canut, N. Chauvin, P. Regreny, M. Gendry, G. Patriarche, J.M. Jancu, J. Even, P. Noe, V. Calvo, B. Salem. J. Nanosci. Nanotechnol., **11** (10), 9153 (2011).
[13] Z. M. Zhao, O. Hul'ko, H.J. Kim, J. Liu, T. Sugahari, B. Shi, Y.H. Xie. J. Cryst. Growth, **271** (3-4), 450 (2004).
[14] B. Bansal, M.R. Gokhale, A. Bhattacharya, B.M. Arora. J. Appl. Phys., **101** (9), 094303 (2007).
[15] X.B. Su, Y. Ding, B. Ma, K.L. Zhang, Z.S. Chen, J.L. Li, X.R. Cui, Y.Q. Xu, H.Q. Ni, Z.C. Niu. Nanoscale Res. Lett., **13**, 1 (2018).
[16] E.N. Yitamben, R.E. Butera, B.S. Swartzentruber, R.J. Simonson, S. Misra, M.S. Carroll, E. Bussmann. New J. Phys., **19** (11), 113023 (2017).
[17] N.A. Fominykh, M.S. Sobolev, I.V. Ilkiv, D.V. Mokhov, T.N. Berezovskaya, A.D. Bouravleuv. J. Phys.: Conf. Ser., **1695** (1), 012006 (2020).
[18] Y.P. Varshni. Phys. Rev. B, **2** (10), 3952 (1970).
[19] W.H. Jiang, X.L. Ye, B. Xu, H.Z. Xu, D. Ding, J.B. Liang, Z.G. Wang. J. Appl. Phys., **88** (5), 2539 (2000).
[20] I. Yeo. J. Dong Song, J. Lee. Appl. Phys. Lett., **99** (15), 151909 (2011).
[21] R. Heitz, I. Mukhametzhanov, A. Madhukar, A. Hoffmann, D. Bimberg. J. Electron. Mater., **28**, 520 (1999).

Translated by Y.Alekseev