Stimulated emission in the InAs/InAsSb/InAsSbP heterostructures with asymmetric electronic confinement

© A.A. Semakova [1], M.S. Ruzhevich [2], V.V. Romanov [1], N.L. Bazhenov [1], K.D. Mynbaev [1,2], K.D. Moiseev [1]

[1] Ioffe Institute,
194021 St. Petersburg, Russia
[2] ITMO University,
197101 St. Petersburg, Russia
E-mail: antonina.semakova@mail.ioffe.ru

Received June 29, 2022
Revised July 6, 2022
Accepted August 4, 2022

The electroluminescent characteristics of the InAs/$InAs_{1-y}Sb_y$/InAsSbP asymmetric light-emitting diode heterostructures with high InSb mole fraction in the active region ($y > 0.09$) in the temperature range 4.2−300 K have been studied. Stimulated emission was achieved in the wavelength range 4.1−4.2 μm at low temperatures ($T < 30$ K). It was found that the electroluminescence spectra were formed as a result of the superposition of contributions from different channels of radiative recombination of charge carriers near the type II heterointerface. The effect of the quality of the type II InAsSb/InAsSbP heterojunction on the radiative interface transitions with an increase in the content of InSb in the ternary solid solution is considered.

Keywords: heterojunctions, InAs, antimonides, electroluminescence, light-emitting diodes.

DOI: 10.21883/SC.2022.09.54131.9925

1. Introduction

The continued interest in heterostructures based on narrow-gap $A^{III}B^V$ solid solutions is attributable both to their physical properties [1] and to the wide range of applicability of such structures in various optoelectronic devices operating in the middle infrared (IR) spectral range (2−6 μm) [2,3]. LED heterostructures based on InAs(Sb, P) solid solutions are promising radiation sources for ecological monitoring and medical diagnostics systems [4,5]. The issue of enhancing the operating efficiency of LEDs of the middle IR range remains topical. The currently available practical solutions for improving the output characteristics of LEDs often make the design of device structures more complicated [6,7]. At the same time, a thorough examination of recombination processes in narrow-gap heterostructures (specifically, at low temperatures) may help reveal new opportunities for enhancement of the parameters of optoelectronic devices. It was demonstrated in [8] that symmetric heterostructures with high energy barriers and a narrow-gap $InAs_{1-y}Sb_y$ ($y = 0.12$) active region exhibit superluminescence at $T = 10{-}35$ K with a full width at half maximum (FWHM) of the emission line of ~ 50 nm. A similar effect in the indicated temperature interval was also observed in the 3.4−3.6 μm spectral range in asymmetric narrow-gap heterostructures with an active region based on $InAs_{1-y}Sb_y$ solid solutions with compositions $y < 0.09$ [9].

In the present study, we report the observation of stimulated emission in electroluminescence (EL) spectra of asymmetric n-InAs/$InAs_{1-y}Sb_y$/p-InAsSbP LED heterostructures with narrow-gap layer compositions $y > 0.09$. The key feature of the design of these samples is the asymmetry of energy jumps at the interfaces of semiconductor compounds forming an epitaxial heterostructure. Therefore, potential barriers for oppositely directed carrier fluxes injected into the active region of a heterostructure may be formed in such a way that a jump in the valence band at one heterointerface would confine holes in the active region, while a jump in the conduction band at the other heterointerface of the active region would block the motion of electrons. As is known, an increase in the molar fraction of InSb in a ternary solution translates into a greater lattice mismatch between an InAsSb epitaxial layer and an InAs substrate. This has a considerable effect on the formation of the heterointerface between an active InAsSb layer and a barrier InAsSbP layer that is matched in the lattice constant with an InAs substrate [10]. Thus, features inducing the formation of additional recombination channels may emerge at a relatively minor band discontinuity at the heterointerface [11]. The observation of stimulated emission associated with indirect recombination transitions at a type II heterointerface is reported below.

2. Experiment

Asymmetric $InAs_{1-y}Sb_y$/p-InAsSbP heterostructures for experiments were grown by metalorganic vapor-phase epitaxy on undoped n-InAs(001) substrates. The epitaxial growth of InAsSb and InAsSbP layers was performed under atmospheric pressure in a horizontal reactor with resistive heating. The n-InAsSb solid solution of the active region was not doped intentionally; the electron density in it (attributable to the presence of residual impurities) did not exceed $n = 3 \cdot 10^{16}$ cm^{-3}. A barrier p-InAsSbP layer doped with zinc was grown on top of the active region. The molar fraction of InSb in the $InAs_{1-y}Sb_y$ solid solution of the active region was $y = 0.095$ (structure A) and $y = 0.110$ (structure B). LED chips with a circular mesa with diameter $d = 300\,\mu$m were formed using standard lithography and wet chemical etching techniques. The specifics of growth procedure and certain optical and electric properties of such heterostructures fabricated at the temperatures of 77 and 300 K have been reported earlier in [12,13]. In the present study, EL was examined in a wide temperature range ($T = 4.2-300$ K) under pulsed excitation with frequency $f = 1$ kHz and pumping pulse duration $\tau = 2\,\mu$s.

3. Results and discussion

Figure 1 presents the temperature dependences of the position of spectral maxima of EL bands of the studied heterostructures and the calculated band gap (E_g) of $InAs_{1-y}Sb_y$ solid solutions of the active region for compositions $y = 0.095$ and $y = 0.110$. Dependence $E_g(T)$ was calculated in accordance with the Varshni relation [14]:

$$E_g = E_0 - \alpha \cdot T^2 \cdot (T + \beta)^{-1}, \quad (1)$$

where coefficients $\alpha = 2.76 \cdot 10^{-4}$ eV/K and $\beta = 93$ K were close to the corresponding parameters for binary compound InAs. The value of E_0 for solid solution $InAs_{1-y}Sb_y$ was calculated as an interpolation of the fractions of InAs and InSb compounds in accordance with the following expression [15]:

$$E_0 = E_{g_{\mathrm{InAs}}}(1-y) + E_{g_{\mathrm{InSb}}} \cdot y - C_{\mathrm{InAsSb}} \cdot y(1-y), \quad (2)$$

where, according to [16], $E_{g_{\mathrm{InAs}}} = 0.417$ eV, $E_{g_{\mathrm{InSb}}} = 0.235$ eV, and $C_{\mathrm{InAsSb}} = 0.61$ eV at $T = 0$ K.

It can be seen from Fig. 1 that the studied samples had distinct temperature dependences of the photon energy at the EL band maximum. At low temperatures ($T < 50$ K), the experimental energy values deviated significantly from the calculated curve. It should be noted that this deviation was ~ 40 meV for structure A and ~ 38 meV for structure B at $T = 4.2$ K. In addition, the position of the spectral maximum of EL bands shifted toward higher photon energies with increasing temperature in the $30 < T < 100$ K interval. The „blue" shift of EL spectra here was $\sim 1.3 \cdot 10^{-4}$ eV/K. Within the $100 < T < 200$ K temperature interval, changes in the position of EL peaks followed the temperature narrowing of the band gap of $InAs_{1-y}Sb_y$ (with a certain deviation toward lower energies with respect to calculated dependence $E_g(T)$). The magnitude of this deviation was 15 meV at $T = 100$ K and near-zero at $T > 200$ K. The value of 15 meV is close to the activation energy of a shallow Zn acceptor in indium arsenide and related solid solutions [16]. Thus, it is fair to assume that spontaneous luminescence in this temperature interval was driven by radiative recombination involving acceptor Zn states in the bulk of the active region, which form due to the diffusion of zinc from the barrier InAsSbP layer to the epitaxial InAsSb layer. The photon energy at the emission band maximum at higher temperatures ($200 < T < 300$ K) matched the band gap of the ternary solid solution. This is indicative of the fact that interband radiative recombination in the active region of heterostructures produces the dominant contribution to EL spectra. In view of this, it seems probable that several channels of radiative recombination exist in the studied heterostructures: (1) interband transitions in the active region at near-room temperatures ($T \sim 300$ K), (2) radiative transitions to impurity states in the band gap of the ternary solid solution at lower temperatures around 100 K, and (3) radiative recombination transitions with a very weak temperature dependence in the low-temperature range ($T < 35$ K). Thus, the $35 < T < 100$ K temperature range may be identified as a transition region where a superposition of contributions of the last two recombination channels, which is manifested in the emergence of a „blue" shift in the EL spectra at higher temperatures (see Fig. 1), may be observed.

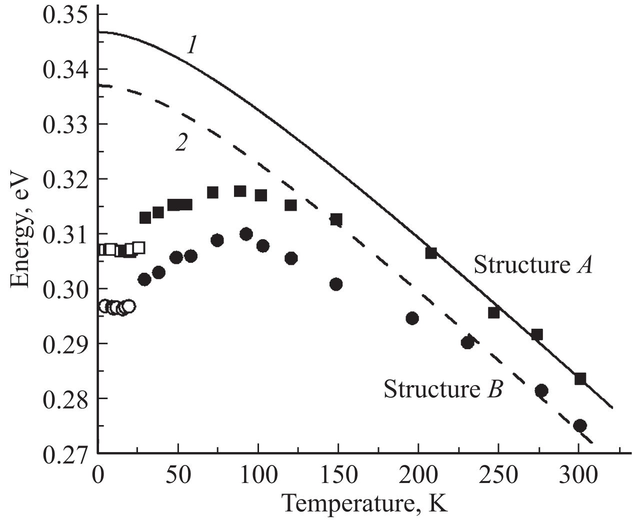

Figure 1. Calculated temperature dependences of the band gap (E_g) of the active region for heterostructures A (curve *1*) and B (curve *2*) and experimental values of the photon energy at the maxima of bands of stimulated (open symbols) and spontaneous (filled symbols) emission.

Figure 2, a shows the schematic band diagram of the InAs/InAsSb/InAsSbP asymmetric heterostructure. It is evident that the confinement for holes is established by a dominant energy jump in the valence band at the InAs/InAsSb heterointerface (as against the conduction band). The confinement for electrons is provided by a potential barrier, which is formed by a layer of the InAsSbP quaternary solid solution, in the conduction band at the InAsSb/InAsSbP heterointerface. Thus, asymmetric electron confinement for carriers in the InAsSb active region is established in the studied structure. It has been demonstrated earlier in [10] that a type II heterojunction InAsSb/InAsSbP

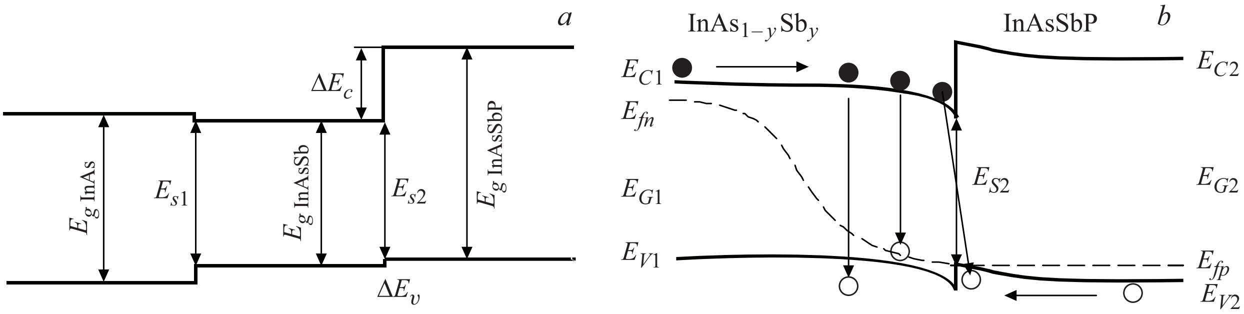

Figure 2. Schematic band diagrams of the InAs/InAsSb/InAsSbP asymmetric heterostructure in general (*a*) and type II staggered heterojunction *n*-InAsSb/*p*-InAsSbP under forward bias applied to the heterostructure (*b*).

may be formed in compositions $y > 0.09$ of narrow-gap solid solution $InAs_{1-y}Sb_y$. An increase in the molar fraction of indium antimonide in $InAs_{1-y}Sb_y$ translated into a more pronounced discontinuity in the valence band (ΔE_V) at the InAsSb/InAsSbP heterointerface. Since the band discontinuities at a type II heterointerface are staggered, potential wells for electrons and holes form on opposite sides of the InAsSb/InAsSbP interface if forward bias is applied (Fig. 2, *b*). The energy distance between electrons and holes (E_{S2}) involved in recombination may be smaller than E_g of the solid solution with the narrowest gap (among the solutions forming the considered heterojunction) and may decrease with increasing molar fraction of InSb in solid solution InAsSb [17].

Thus, radiative transitions observed experimentally at $T = 4.2$ K, which have a photon energy that is considerably lower than the value of E_g of the active region, may be attributed to the recombination of carriers near the InAsSb/InAsSbP heterointerface. As was demonstrated earlier for an individual type II heterostructure *n*-InGaAsSb/*p*-GaInAsSb, localization of electrons and holes in potential wells at a type II heterointerface may well allow for lasing driven by quasi-indirect (tunnel) radiative transitions through the interface [18]. Owing to this carrier localization, the energy positioning of EL spectra has almost no dependence on temperature.

The studied heterostructures exhibited stimulated emission at temperatures $T = 4.2-30$ K. Figure 3 presents the EL spectra obtained at $T = 4.2$ K and different pumping currents. At low excitation levels ($i < 0.2$ A), the EL spectra contained one marked band of spontaneous luminescence with FWHM $\sim 20-23$ meV. The observed rapid low-energy decay in the spontaneous EL spectra around photon energy $h\nu \sim 0.29$ eV is attributable to the absorption of generated emission by carbon dioxide gas molecules (CO_2) from the atmosphere (Fig. 3, *b*). When the pumping current was raised, a narrow band of stimulated emission with FWHM ~ 2 meV (~ 30 nm) emerged. Single-mode generation was observed at $h\nu = 0.304$ eV (structure *A*) and $h\nu = 0.297$ eV (structure *B*), which corresponded to the spectral positions of the spontaneous luminescence maximum of the studied heterostructures at the given temperature. At high pumping levels (in the current range of 0.8−2.4 A), the multimodality of the EL spectrum was manifested, and the spectrum shifted toward higher photon energies (see Figs. 3, *a* and *b*). One may also note that the intensity gets redistributed between modes as the excitation level increases. An average intermode distance on the order of $\Delta\lambda \sim 1.2$ nm corresponded to cavity length $L = 170\,\mu$m, which was governed by the InAs substrate thickness.

Figure 4, *a* presents the typical multimodal structure of the stimulated emission spectrum. Approximating this spectrum with a set of curves based on the Lorentz distribution, one may isolate four modes. The dependences of the spectral position of each mode on the pumping current (Fig. 4, *b*) were plotted by combining the EL spectra obtained at different pumping levels (see Fig. 3). An overall shift of the spectrum of stimulated emission toward higher photon energies and an increase in the intermode distance were observed at higher pumping currents. The latter fact may be attributed a change in the shape of potential wells at type II heterointerface InAsSb/InAsSbP occurring when the external bias increases. Thus, it is fair to say that stimulated emission observed in the present study at low temperatures is attributable to interfacial radiative recombination of holes near the Fermi level with electrons localized in a potential well. The observed shift of the EL spectra toward higher photon energies at higher injection levels is associated with a shift of the localization level for electrons (Fig. 4, *b*).

As was demonstrated below, the temperature of transition from stimulated emission to spontaneous one was ~ 30 K. This value is lower than the one reported in [9] for a similar heterostructure with an active region made from InAsSb with a lower molar fraction of InSb. The crystalline perfection of the structure is known to be associated closely with the lattice mismatch of InAsSb solid solutions and substrates made from binary compound InAs that are used to grow them. This mismatch for the compositions of epitaxial layer $InAs_{1-y}Sb_y$ considered in the present study ($y = 0.095$ and $y = 0.110$) does not exceed the critical value for thin layers (1%). The generation of mismatch dislocations (accompanied by an increase in the density of extended and point defects) is possible if the magnitude of mismatch between the InAsSb layer and the InAs substrate

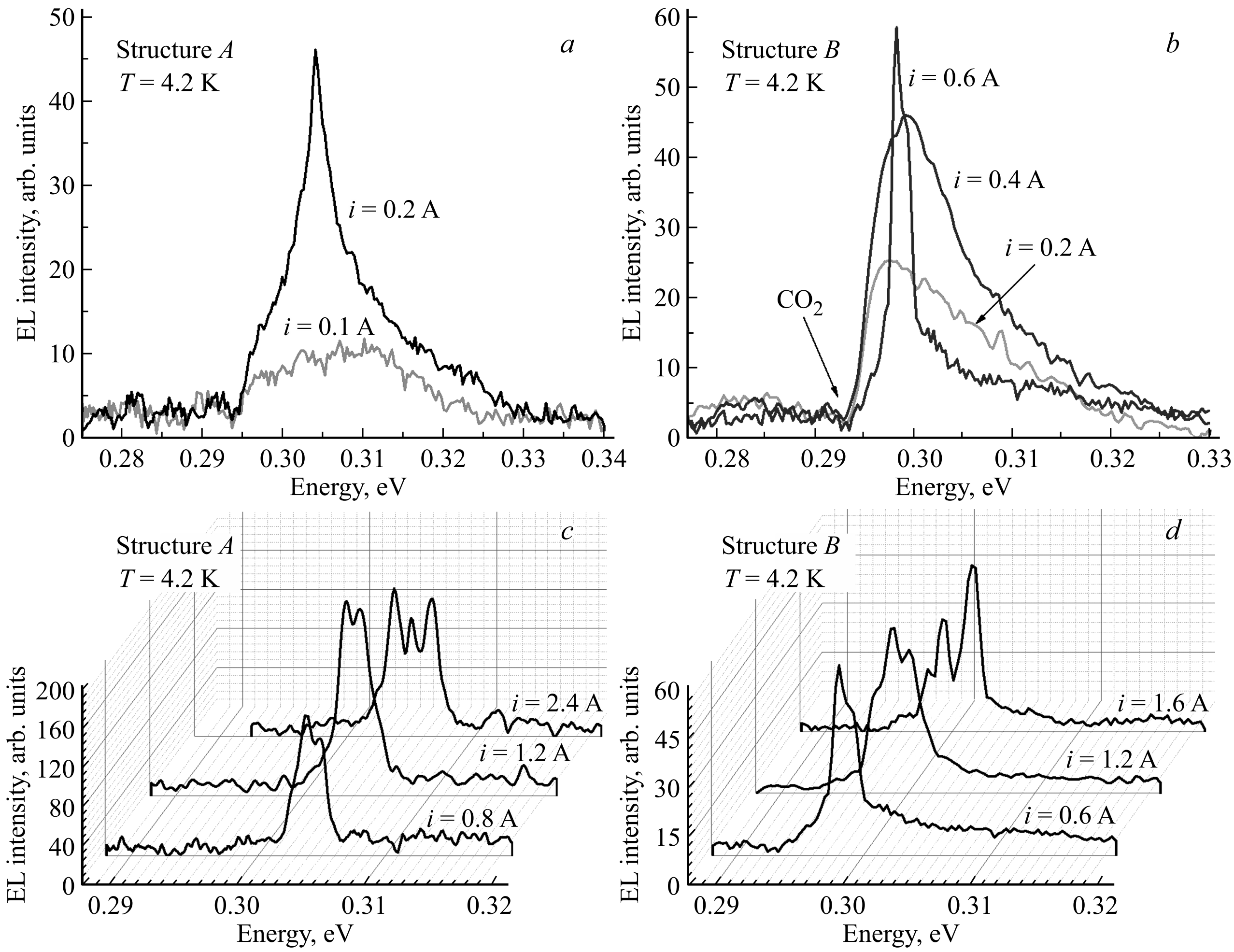

Figure 3. EL spectra for heterostructures A (a, c) and B (b, d) obtained at $T = 4.2$ K and different pumping currents.

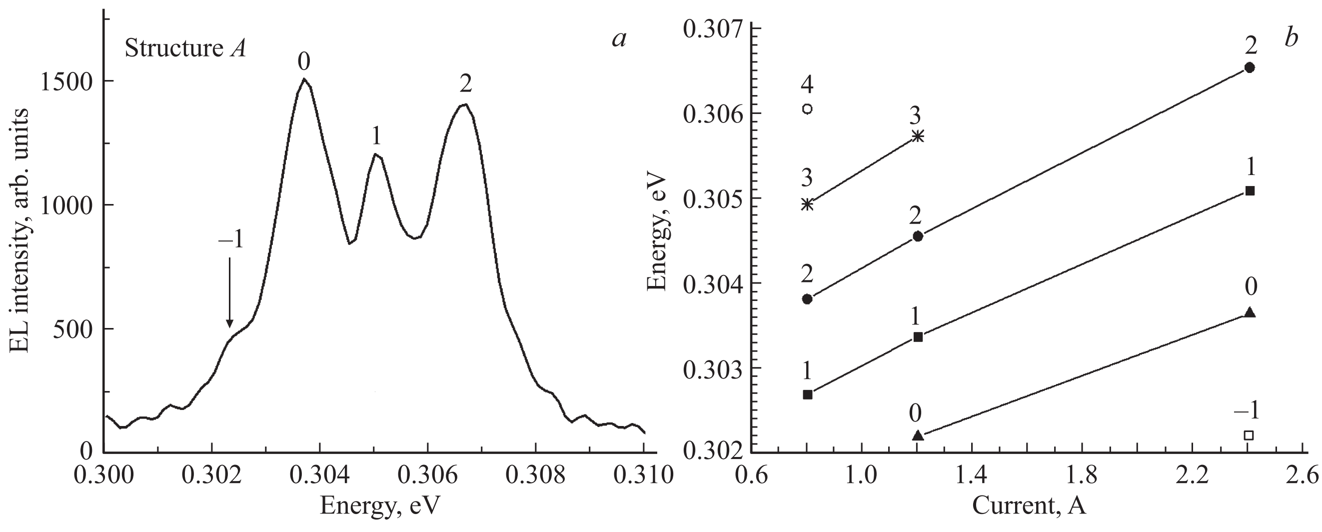

Figure 4. EL spectrum for heterostructure A at $i = 2.4$ A and $T = 4.2$ K (a); dependence of the spectral positioning of stimulated emission modes on the pumping current for this heterostructure (b).

increases. Experimental EL data for similar narrow-gap asymmetric heterostructures did not reveal any stimulated emission in samples with an elevated InSb concentration in an InAsSb active region [11].

4. Conclusion

The results of examination of electroluminescence of asymmetric LED n-InAs/InAs$_{1-y}$Sb$_y$/p-InAsSbP heterostructures with molar fraction $y = 0.095$ and 0.110 of InSb in the active layer at temperatures $T = 4.2-300$ K were reported. Stimulated emission was observed in the luminescence spectra at low temperatures within the $4.2-30$ K range. The redistribution of intensity of spectral modes with increasing injection current at $T = 4.2$ K was demonstrated. The contribution of three different channels to radiative recombination, which become dominant at different temperatures, was identified. At low temperatures $T < 100$ K, spontaneous and stimulated emission spectra were shaped by interfacial transitions at type II heterointerface InAsSb/InAsSbP. When the temperature increased to $T > 100$ K, only spontaneous luminescence was observed; transitions to acceptor zinc levels in the active region became the dominant recombination channel. At high temperatures $T > 200$ K, EL was governed by interband radiative recombination transitions of carriers in the bulk of the active InAsSb layer. The observed intense interfacial EL at the InAsSb/InAsSbP heterointerface demonstrates that these heterostructures have potential for application in IR emitters with $\lambda > 4\,\mu$m.

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] S. Adachi. *Properties of Semiconductor Alloys: Group-IV, III-V, and II-VI Semiconductors* (John Wiley & Sons, Ltd., Chichester West Sussex, 2009).
[2] A. Rogalski, P. Martyniuk, M. Kopytko, P. Madejczyk, S. Krishna. Sensors, **20**, 7047 (2020).
[3] A.P. Ongstad, R. Kaspi, G.C. Dente, M.L. Tilton, R. Barresi, J.R. Chavez. Appl. Phys. Lett., **92**, 141106 (2008).
[4] D. Ting, A. Soibel, A. Khoshakhlagh, S. Keo, B. Rafol, A. Fisher, B. Pepper, E. Luong, C. Hill, S. Guhapala. Infr. Phys. Technol., **97**, 210 (2019).
[5] D. Jung, S. Bank, M.L. Lee, D. Wasserman. J. Opt., **19**, 123001 (2017).
[6] Q. Lu, Y. Zhou, J. Chen, L. He, A. Krier. Proc. SPIE, Smart Photonic and Optoelectronic Integrated Circuits XXII, **11284**, 39 (2020).
[7] F.A. Al-Saymari, A.P. Craig, Q. Lu, A.R.J. Marshall, P.J. Carrington, A. Krier. Opt. Express, **28**, 23338 (2020).
[8] D.M. Kabanau, Y.V. Lebiadok, Yu.P. Yakovlev. J. Appl. Spectrosc., **84**, 843 (2017).
[9] K.D. Mynbaev, N.L. Bazhenov, A.A. Semakova, A.V. Chernyaev, S.S. Kizhaev, N.D. Stoyanov, V.E. Bougrov, H. Lipsanen, Kh.M. Salikhov. Inf. Phys. Technol., **85**, 246 (2017).
[10] K.D. Moiseev, V.V. Romanov. Phys. Solid State, **63**, 595 (2021).
[11] A.A. Semakova, V.V. Romanov, N.L. Bazhenov, K.D. Mynbaev, K.D. Moiseev. Semiconductors, **55**, 354 (2021).
[12] V.V. Romanov, E.V. Ivanov, K.D. Moiseev. Phys. Solid State, **61**, 1699 (2019).
[13] V.V. Romanov, E.V. Ivanov, A.A. Pivovarova, K.D. Moiseev, Yu.P. Yakovlev. Semiconductors, **54**, 253 (2020).
[14] Y.P. Varshni. Physica, **4**, 149 (1967).
[15] J.A. Van Vechten, T.K. Bergstresser. Phys. Rev. B, **1**, 3351 (1970).
[16] Landolt-Börnstein. *Handbook, Numerical Data*, Ser. III, v. 17a [ed. by O. Madelung (Springer, Berlin–Heidelberg, 1982)].
[17] V.V. Romanov, E.V. Ivanov, K.D. Moiseev. Phys. Solid State, **62**, 2039 (2020).
[18] K.D. Moiseev, M.P. Mikhailova, Yu.P. Yakovlev. Semiconductors, **37**, 985 (2003).