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Radiative recombination in the InAs/InSb type II broken-gap heterojucntion with quantum dots at the interface

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The electroluminescent properties of narrow-gap type II InAs/InSb/InAs heterostructures containing a single layer of InSb quantum dots placed at the interface of the $p-n$ junction in InAs were studied. The features of the electroluminescence spectra depending on the surface density of nanoobjects at a broken-gap type II heterointerface were investigated both at forward and reverse bias. When applying a reverse bias to the heterostructures under study, the suppression of negative interband luminescence and the dominance of interface recombination transitions at the InSb/InAs type II heterojunction were observed at room temperature. The radiation, which corresponded to recombination transitions involving localized states of InSb quantum dots, was recorded at low temperature.

Keywords: quantum dots, electroluminescence, InAs, InSb, type II heterojunction.

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1. Introduction

Unique properties of nanoheterostructures with quantum dots (QD) are responsible for the wide interest in research and practical use of these materials for various applications of optoelectronics [1,2]. The use of QDs in the active region of lasers allows controlling the wavelength in a wide range, achieving a considerable improvement of quantum efficiency, a noticeable decrease in threshold currents and a significant increase in operating temperature of such lasers [3]. The application of QDs in infrared photodetectors gives the possibility to improve sensitivity and detectivity, provides low dark currents and significantly improves temperature dependencies of key parameters of the above-mentioned photodetectors [4]. To develop optoelectronic devices operating in the middle infrared range with a wavelength of $\lambda > 4\,\mu$m ($h\nu < 0.3$ eV) [5], are very promising nanoheterostructures with QDs in the InAs−InSb system. Energy band diagram of these structures is a type II broken-gap heterojunction at each QD/matrix layer interface (Figure 1, *a*), which allows using the advantages of such junctions as compared to type I heterostructures [6].

Previously, photoluminescent (PL) properties of heterostructures based on InSb QD layers placed in the InAs matrix with *n*-type conductivity have been studied [7]. At low temperatures a long-wavelength radiation has been found in the spectral range of 0.2−0.3 eV caused by the recombination transitions involving QD eigenstates. Also, the electroluminescence (EL) of heterostructures based on a single layer of InSb quantum dots in the indium arsenide matrix with hole-type conductivity has been studied [8]. Under reverse bias, a negative luminescence was observed in EL spectra in a wide range of temperatures: $T = 77-300$ K. This effect was caused by carrier extraction from the lightly doped region of *p*-InAs adjacent to the InAs/InSb broken-gap heterojunction. This study presents results of investigations of electroluminescent properties of heterostructures where a single layer of InSb QDs is located directly at the interface of two matrix layers of indium arsenide of *p*-type and *n*-type, i.e. at the heterointerface of the $p-n$-junction.

2. Structure fabrication technology and experimental technique

Using the combined technology of epitaxial deposition narrow-band heterostructures with a single InSb QD layer in a InAs matrix were formed. The samples were grown on *p*-InAs(001) substrates doped with a Mn acceptor impurity up to a hole concentration of $p_{300} \sim 10^{17}\,\text{cm}^{-3}$, on which a layer of InSb QDs was deposited by the liquid-phase epitaxy (LPE) method. Two types of QD layers with different surface concentrations of nanoobjects were produced. Sample 1 (MP-24/1) contained QD arrays with a density of $n_{QD1} = 0.7 \cdot 10^{10}\,\text{cm}^{-2}$, while sample 2 (MP-1/1) demonstrated a QD surface density of about $n_{QD2} = 2 \cdot 10^{10}\,\text{cm}^{-2}$. The technique of QD formation and their structural properties are described in previous studies [9,10]. Then the produced InSb QD layer was overdeposited by an epitaxial layer of indium arsenide using the method of metalorganic vapour phase epitaxy (MOVPE) at a temperature of 510°C. The overdeposited InAs layer was not doped intentionally in the process of epitaxy and demonstrated *n*-type conductivity with an estimated electron concentration of $n_{300} \sim 6 \cdot 10^{16}\,\text{cm}^{-3}$. The presence of overgrown InSb QD arrays in the InAs matrix was confirmed by transmission electron microscopy studies [10]. Due to the fact that the epitaxial deposition used undoped compounds with the electron type of conductivity, the diffusion of acceptor impurity into the deposited layer through the *p*-InAs substrate/*n*-InAs layer heterointerface can be almost excluded. Thus, a metallurgic $p-n$-junction was formed inside the InAs matrix with a layer of InSb QDs located at its heteroboundary. The *p*-InAs/*n*-InAs structure (without the QD layer) was selected as a reference sample, which was formed simultaneously with samples 1 and 2 in a single MOVPE run.

The samples for EL measurements were made in the form of $500 \times 500\,\mu$m mesa diodes with a continuous contact on the side of substrate and a point contact to the top epitaxial layer. The round mesa made by the standard photolithography and selective wet chemical etching had a diameter of $D \sim 300\,\mu$m and size of the point contact was $d \sim 100\,\mu$m. The mesa diodes under study were mounted on TO-18 holders by their substrates facing the metal housing. EL spectra were studied using an automated setup on the basis of a DK-480 monochromator (CVI Laser Corp.) with a diffraction grating of 150 lines/mm, an InSb photovoltaic detector cooled by liquid nitrogen (Judson Co) and a SR-810 lock-in amplifier (Stanford Research Systems Inc.). The setup was used to record the radiation emerging from the heterostructure through the covering epitaxial layer. The samples were powered by a square wave with a pulse duty factor of $\eta = 50\%$ and a frequency of $f = 512$ Hz for both the forward polarity of external bias and the reversed polarity of external bias. The forward polarity corresponded to the case when a positive potential was applied to the *p*-InAs substrate and a negative potential was applied to the covering epitaxial layer of *n*-type. The amplitude of driving current pulses (i) varied in the range from 20 to 200 mA.

To analyze the obtained EL spectra, preliminary calculations of the energy band diagrams of the experimentally studied heterostructures were performed. The presence of a single InSb QD layer can be represented as a narrow quantum well with a set of single localization levels (see Figure 1, a). It can be seen from Figure 1 that due to the feature of type II broken-gap heterojunction in the InSb/InAs compound system the ground hole level of the InSb QD (μ_h) is located higher in energy than the bottom of the conduction band of the InAs matrix layer.

3. Experimental results and discussion

The EL spectra of the studied samples (for both the *p*-InAs/*n*-InAs homojunction and the *p*-InAs/InSb/*n*-InAs heterostructures with QDs) at room temperature demonstrated similar features under applied forward external bias (Figure 2, a). A typical EL spectrum contained one pronounced emission band with an intensity maximum near $h\nu_{300}(+) = 0.36$ eV and a FWHM = 48 meV, which was correspondent to interband radiative transitions in the bulk of indium arsenide [8]. In addition to the main band of luminescence, one more emission band can be distinguished near $h\nu_{IS} = 0.31$ eV, which was manifested as a shoulder on the low-energy slope of the EL spectrum. As it has been shown in [11], structural crystalline defects in the indium arsenide form acceptor states on the surface of the semiconductor. When a forward bias is applied, due to the bending of energy bands in the indium arsenide (Figure 1, b) electrons on their path from the occupied conduction band of *n*-InAs to the empty conduction band of *p*-InAs under the external electric field meet potential barriers arising in the form of band structure of QDs and interface states (IS) at the interface of matrix layers. Then the additional EL band can be attributed to the radiative transitions involving localized states inside the band gap of InAs with an activation energy of $E_{IS} \sim 50$ meV. No any other radiative recombination transitions involving doping states with an activation energy less than 30 meV were found, which can be explained by their depletion due to the heating at $T = 300$ K.

When a reversed bias was applied, shapes of the EL spectra at room temperature for samples with QDs and without QDs were drastically different from each other (Figure 2, b). The spectrum for the reference structure of *p*-InAs/*n*-InAs demonstrated an emission band near $h\nu_{300}(-) = 0.36$ eV phase-reversed by 180° in relation to the zero level of intensity, i. e. a negative luminescence was observed in the volume of *p*-InAs [8,12]. Together with this band, a weak peak of positive EL was manifested near photon energies of $h\nu_{IS} = 0.31$ eV. In turn, the EL spectra for heterostructures with InSb QDs (*p*-InAs/InSb/*n*-InAs) demonstrated positive luminescence only and contained one emission band with an intensity maximum near $h\nu_{IS} \sim 0.31$ eV and no luminescence was detected in the region of photon energies close to the InAs band gap. The effect of negative luminescence suppression has been observed previously for heterostructures with ultrathin oxide layer that separates surfaces of two contacting semiconductors which form a type II broken-gap heterojunction and keeps levels of localized states on the surface of indium arsenide [13]. Thus, with any direction of the driving current (both forward and reversed bias) radiative recombination transitions involving the interface level take place, which manifests in the kept polarity of the observed emission for the specific EL band. The $h\nu_{IS}$ band observed at room temperature can be attributed to radiative transitions to the surface states of the *p*-InAs/*n*-InAs heterointerface (see Figure 1, b and c).

With a decrease in temperature down to $T = 77$ K, the contribution of radiative recombination transitions involving doping states in *p*-InAs with an activation energy greater than the energy of thermal widening of levels ($kT_{77} \sim 6$ meV) becomes noticeably manifested. The EL spectra for all studied samples under forward external bias contained two clearly distinguished emission bands of $h\nu_{77} = 0.408$ eV and $h\nu_A^{77} = 0.376$ eV (Figure 3). The high-energy band $h\nu_{77}$ was correspondent to interband radiative transitions to the indium arsenide [8]. The photon energy at the emission maximum of low-energy band $h\nu_A^{77}$

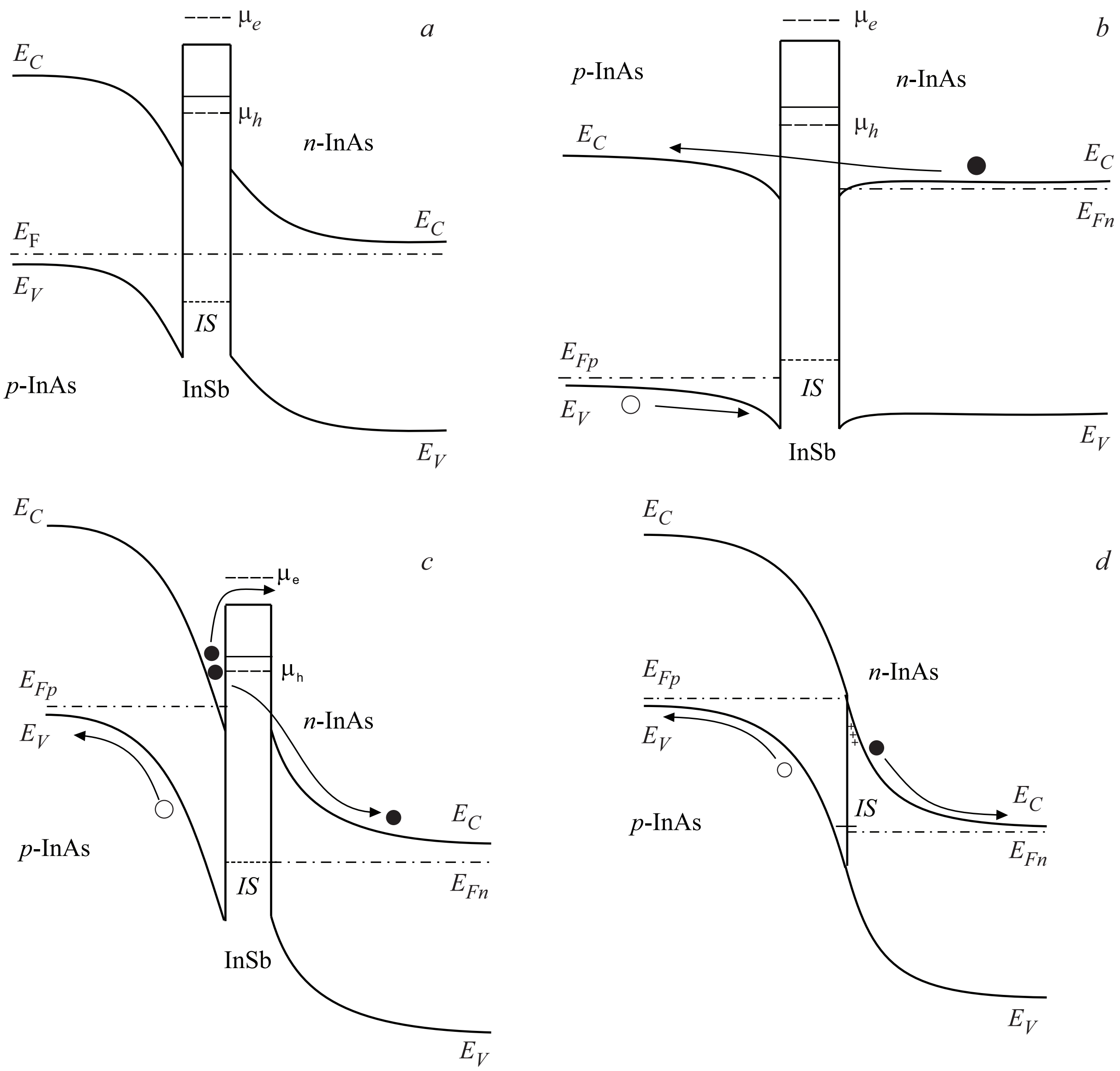

Figure 1. Schematic energy band diagram of InAs/InSb/InAs heterostructures with a InSb QDs layer at the $p-n$-junction of InAs in the thermodynamic equilibrium (a), at forward bias (b) and at reversed bias (c). d — p-InAs/n-InAs structure at reversed bias. IS is the level of surface states at the interface, μ_e and μ_h are ground electron and hole states of InSb QD, respectively.

was 32 meV less than the energy of interband transitions, which was indicative of the involvement in the EL spectrum of the radiative recombination transitions related to the Mn doping acceptor states inside the band gap of InAs [14]. In addition, the presence of one more emission band is assumed with its maximum near 0.35 eV, which was manifested as a shoulder on the low-energy slope of the $h\nu_A^{77}$ band, that may be due to the radiative transitions involving interface states ($\Delta h\nu_{77}(IS) \sim 58$ meV).

During measurements a different behavior of EL band intensities was detected with an increase in the driving current depending on the QDs concentration at the $p-n$-junction interface. For the reference sample (without QDs) and sample 1 (with low density of QDs at the interface) the dominance of emission band $h\nu_A^{77}$ was kept throughout the entire range of driving currents (Figure 3, a). For sample 2 (with high density of QDs) a redistribution of intensities between EL bands was observed, when the dominance of low-energy band $h\nu_A^{77}$ at a low pumping level ($i < 50$ mA) was changed to the dominance of high-energy band $h\nu_{77}$ at higher levels of pumping (Figure 3, b). In other words, an increase in density of nanoobjects at the interface resulted in a decrease in contribution from radiative transitions involving doping acceptor states to the total EL spectrum. According to the energy diagrams shown in Figure 1, the presence of InSb QDs at the interface of indium arsenide matrix layers results in a potential barrier on the path of current flow due to the formation of surface interface states and eigen localized levels in the potential field of QDs. The emergence of a space charge region near the

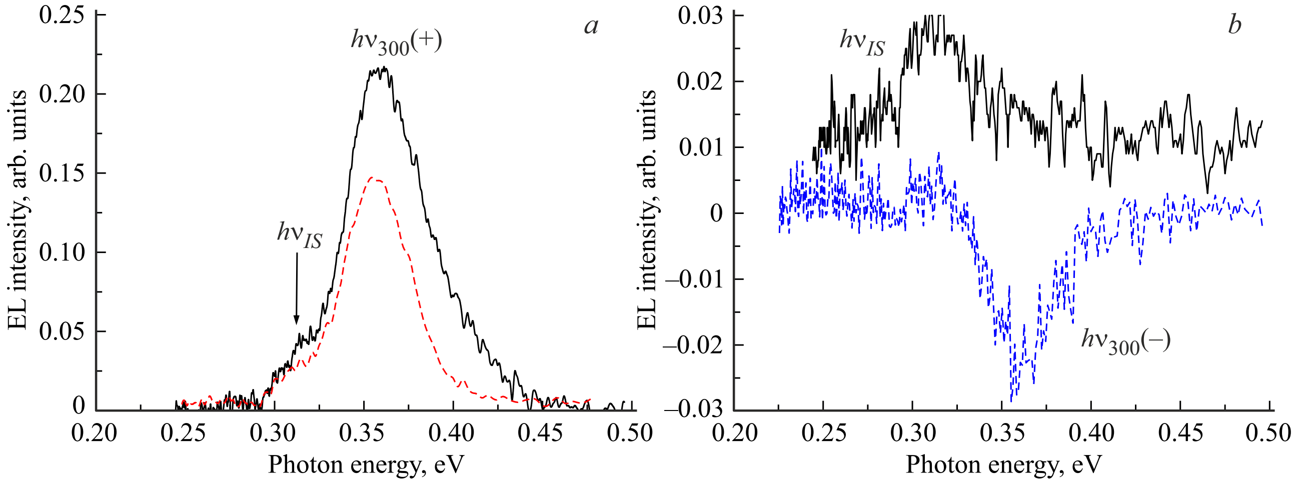

Figure 2. EL spectra obtained for *p*-InAs/*n*-InAs (dashed line) and *p*-InAs/InSb/*n*-InAs heterostructures (solid line). Measurement were carried out at forward (*a*) and reversed (*b*) bias, at a temperature of $T = 300$ K and a driving current of $i = 50$ mA.

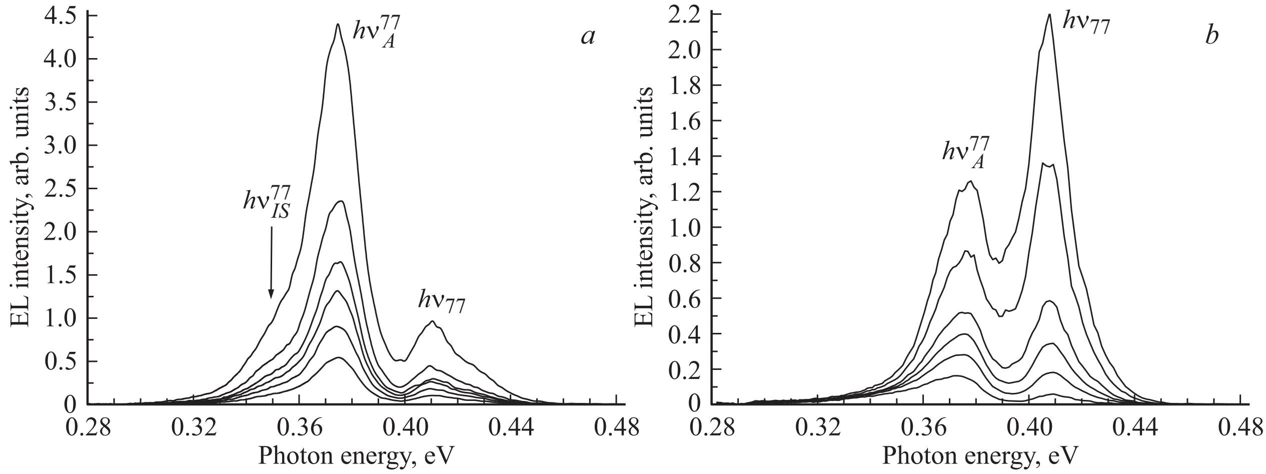

Figure 3. EL spectra under applied forward external bias taken for driving currents $i = 20$, 30, 40, 50, 75, 100 mA (from bottom to top) at a temperature of $T = 77$ K: *a* — for *p*-InAs/*n*-InAs structures and *p*-InAs/InSb/*n*-InAs heterostructures with a QD density less than 10^{10} cm^{-2}; *b* — for *p*-InAs/InSb/*n*-InAs heterostructures with a QD density over 10^{10} cm^{-2}.

heterointerface on the *p*-InAs side (see Figure 1, *b*) prevents the penetration of electrons into the region of flat bands where the emitting recombination involving impurity levels takes place.

With a reversed bias applied to the studied heterostructures, no emission band correspondent to interband transitions in the indium arsenide was observed in EL spectra at $T = 77$ K (Figure 4). The same typical feature of spectra at low temperature has been found previously for heterostructures with a layer of InSb QDs in the InAs matrix with *p*-type conductivity where interband transitions started their manifestation as a negative luminescence only at very high biases [8]. The main emission band in EL spectra under reversed bias (see Figure 4) had an asymmetric shape with a sharp high-energy edge and demonstrated a „blue" shift with an increase in the driving current, i.e. spectral position of the EL band intensity maximum shifted toward higher photon energies (Figure 5). Thus, for the reference sample (without QDs) a linear dependence of $h\nu_{IS+SL}(i)$ was observed in the range of photon energies from 0.345 to 0.358 eV in the current range of $i = 50-150$ mA (Figure 5, dark squares). By comparing the energy band diagram of the reference sample (see Figure 1, *d*) and the energy range where the shift of the main EL band was observed under reversed bias, the energy position of the interface level *IS* at low temperature can be estimated. At the same time, the linear „blue" shift observed in the EL spectra is a result of the radiative transitions from shallow doping levels (SL) formed by natural structural defects in the covering layer of *n*-InAs to the interface level *IS*. Accordingly, an increase in the reversed bias caused gradual depletion of donor levels of the undoped indium arsenide near the heterointerface. According to our estimates, the *IS* level at low temperature ($T = 77$ K) was

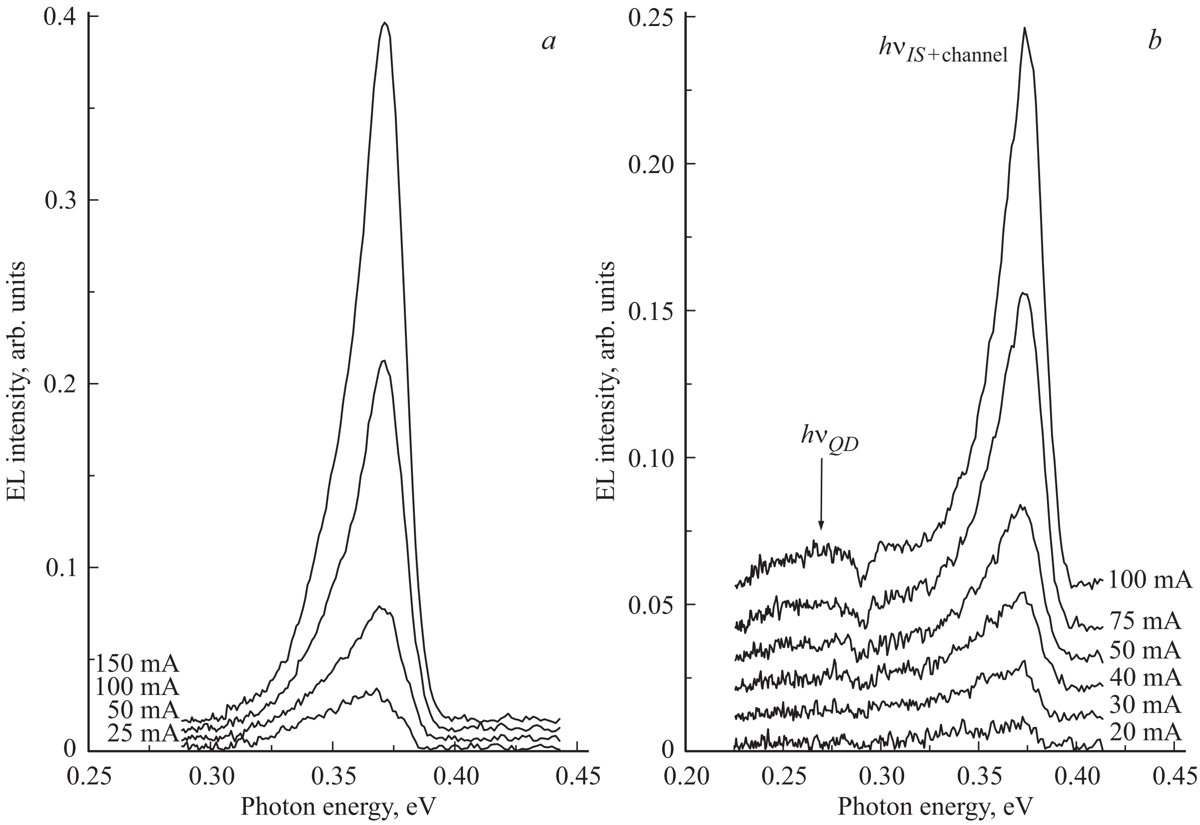

Figure 4. Electroluminescence in p-InAs/InSb/n-InAs structures with InSb QDs (a — sample 1 and b — sample 2) measured under a reversed bias and a temperature of $T = 77$ K.

approximately 50 meV above the valence band top of InAs, which is in a good agreement with the experimental data obtained at room temperature.

It should be emphasized that both $h\nu_{IS+\text{channel}}(i)$ dependencies that describe the „blue" shift for samples with QDs demonstrated saturation at high pumping levels and the specific behavior of the above-mentioned dependencies was determined by the concentration of nanoobjects at the p-InAs/n-InAs heterointerface of the studied structures. For sample 2 with high surface density of QDs at the interface ($n_{QD2} > 10^{10}\,\text{cm}^{-2}$) the luminescence started to be manifested at a photon energy of $h_{IS+\text{channel}} = 0.366$ eV (Figure 5, dark circles). At the same time, the range of motion of the above-mentioned EL peak from the side of high photon energies was limited by the energy of emitting transition of $h\nu_A^{77} = 0.376$ eV (Figure 5, light squares). For sample 1 with low density of QDs at the interface ($n_{QD1} < 10^{10}\,\text{cm}^{-2}$) the $h\nu_{IS+\text{channel}}(i)$ dependence resulted in a superposition of two extreme considered cases (Figure 5, dark triangles). For the given sample the start of luminescence was detected at $h\nu_{IS+\text{channel}} = 0.348$ eV, and the „blue" shift achieved saturation at a photon energy of 0.37 eV.

Now let us consider the nature of emergence and the behavior of the $h\nu_{IS+\text{channel}}$ band. With a reversed bias applied to the investigated heterostructures with InSb QDs

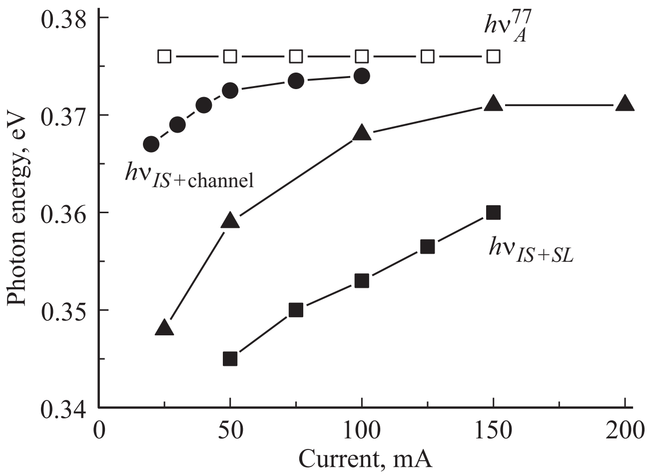

Figure 5. Dependence of the spectral position of the EL band maxima on the driving current at a temperature of $T = 77$ K with the reversed external bias applied (dark symbols). The $h\nu_A^{77}$ dependence with the forward bias applied is presented for comparison (light squares).

(1 and 2) a potential well is formed at the side of p-InAs near the interface as a result of the bend in the conduction band (Figure 1, c). As the external electric field increases, an electron channel is formed at the p-InAs/InSb

heterointerface due to the occupation of the potential well by electrons [15]. In this case there is a probability of radiative recombination transitions from the Fermi quasi-level (E_{Fp}) of the electron channel to the interface level IS. Quantum energy of such transitions can be described by the following relationship:

$$h\nu_{IS+\text{channel}} = E_{IS} + E_{Fp}, \quad (1)$$

where E_{IS} is the energy of interface state level in relation to the bottom of the conduction band of InAs. With further increase in the external bias and, as a consequence, filling of the electron channel, the E_{Fp} level will shift toward higher energies, which will be manifested in the „blue" shift of the EL band correspondent to these transitions. The subsequent saturation of the observed shift with increase in driving current is explained by the fact that the filling of electron channel continues until the electrons start leaking through the potential barrier formed by the band gap of InSb. As a result, an additional channel of current flow involving electron states of QD is formed. Accordingly, samples with different density of QDs at the interface demonstrate different saturation levels of the „blue" shift of EL bands (Figure 5).

The emergence of a new channel of current flow under applied reversed bias in samples with QDs allows occupying eigenstates in the InSb QD. As a result, an emission band is observed in the EL spectra at low temperatures, which is correspondent to recombination transitions between localization levels in QDs. The photon energy of such transition can be estimated as

$$h\nu_{QD} = E_{QD} + \delta E_{QD} + \mu_h + \mu_e, \quad (2)$$

where E_{QD} is a band gap width of the InSb binary compound, δE_{QD} is change in the band gap width of InSb due to internal compression strain in QDs, μ_e and μ_h are energies of ground electron and hole levels of QD, respectively. For sample 2 with a surface density of nanoobjects of $n_{QD2} = 2 \cdot 10^{10}\,\text{cm}^{-2}$ an emission band in the region of $h\nu_{QD} = 0.27\,\text{eV}$ with a half-width of about 60 meV was observed in EL spectra at a reversed bias and a temperature of $T = 77\,\text{K}$ (Figure 4, b). At the same time, a sharp drop on the low-energy side of the EL spectrum near the photon energy of 0.29 eV is related to the absorption of optical emission by molecules of CO_2 ($\lambda = 4.27-4.29\,\mu$m), which is present in the laboratory atmosphere. The spectral position of the $h\nu_{QD}$ band is in a good agreement with the experimental data obtained from the measurement of PL spectra at $T = 77\,\text{K}$ for structures with InSb QDs into the InAs matrix grown by molecular-beam epitaxy [7]. In the cited study a high-intensity PL correspondent to recombination transitions involving localized states of QDs was observed in the spectral range of 0.2–0.3 eV. It is worth to note that the above-mentioned PL band was typical for structures with a surface density of nanoobjects of $n_{QD} > 2 \cdot 10^{10}\,\text{cm}^{-2}$. Due to the fact that the intensity of emission caused by transitions between localization levels in QDs depends on the concentration n_{QD}, no $h\nu_{QD}$ EL band was detected in our experiments for the samples with low density of QDs (Figure 4, a). In EL spectra recorded at a temperature of $T = 300\,\text{K}$ the correspondent emission band was not observed as well (see Figure 2, b). At the same time, it should be noted that in the presented study EL spectra were measured using an InSb photovoltaic detector cooled by liquid nitrogen. This photodetector has a long-wavelength cut-off of sensitivity of $\lambda = 5.3\,\mu$m that does not allow recording an emission with a photon energy less than 0.23 eV. Thus, taking into account the temperature shift of EL spectra toward lower photon energies with increase in temperature, our experimental capabilities were not sufficient to detect the emission that corresponds to recombination transitions involving localized states of QDs at $T = 300\,\text{K}$.

Attempts to obtain EL in heterostructures with InSb QDs in the InAs-based matrix have been taken by other groups of researchers [16]. However, in our opinion, the low surface density of nanoobjects does not allow interpreting the emission band observed in the above study with a photon energy near 0.37 eV as recombination transitions between InSb QD states. The aforementioned EL band most likely is due to the involvement of surface levels at the InAs/InSb heterointerface.

4. Conclusion

The narrow-band heterostructures with a single InSb QDs layer at the p-InAs/n-InAs junction were produced by the combined technology of epitaxial deposition. p-InAs/InSb/n-InAs type-II broken-gap heterostructures under applied forward external bias demonstrated an emitting recombination in the bulk of indium arsenide and an interface luminescence involving surface states at the InAs/InSb heterointerface. The emitting recombination transitions between localization levels in InSb QDs in the InAs-based narrow-band matrix can be observed when the ground electron and hole states of nanoobjects are occupied. For structures with type-II broken-gap InAs/InSb heteroboundaries the given condition has been implemented with the reversed external bias applied, when QD states were filled from the electron channel at the interface.

Conflict of interest

The authors declare that they have no conflict of interest.

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