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The influence of  ${}^{60}$ Co  $\gamma$ -irradiation at a dose of  $2 \times 10^8$  rad on the low-temperature photoluminescence (PL) spectra of  $Cd_xZn_{1-x}Se/ZnSe$  heterostructures containing a single quantum well (QW) with various cadmium content (x = 0.21, x = 0.23, x = 0.35, and x = 0.43) has been investigated. It has been shown that a partial strain relaxation in the QW and ZnSe layers occurs after irradiation. The mechanism of the strain relaxation in the QW was found to depend on the cadmium content. The strain relaxation in the ZnSe layer and the QW with  $x = 0.21 \div 0.23$ was suggested to occur via the formation (or the transformation) of specifically oriented extended defects, whereas, in the case of QW with  $x = 0.35 \div 0.4$ , this process is due to the diffusion of Cd from the QW into the ZnSe layer enhanced by the deformation and concentration gradients near the QW/ZnSe heterointerface.

### 1. Introduction

The interest in epitaxial CdZnSe/ZnSe heterostructures with quantum wells, which arose at the beginning of the 1990s, was caused by the capability of their application as active elements of lasers and light-emitting diodes in the visible spectral range [1–3]. The demand for such devices was dictated by the quickly developed market of digital compact disks with high density of data recording. Since the data density in such systems quadratically depends on the emission wavelength, researchers aimed at creating blue and blue-green laser diodes. In spite of the substantial progress achieved in the creation of such devices on the basis of CdZnSe, they found no wide application due to their low degradation stability and gave way to commercial GaN-based blue laser light-emitting diodes.

Despite all that, green laser light-emitting diodes on the basis of low-dimensional structures Cd(Zn)Se/ZnSe remain actual up to date, which is associated with the absence of their commercial alternatives, on the one hand, and a high demand for such devices, on the other hand. In particular, environmental analysis or various applications in medicine, as well as in laser video projectors, demand the laser emission just in the green spectral range. Moreover, ZnSe-based laser diodes can become a new kind of light source for plastic optical fibres, in which the minimum of absolute losses at transmittance is about 560 nm [4]. However, in order to obtain the yellow-green emission, the cadmium content in QWs in modern lightemitting diodes on the basis of  $Cd_xZn_{1-x}Se$  QWs has to be  $x = 0.4 \div 0.5$ . Owing to a considerable mismatch between the CdSe and ZnSe lattice constants, the increase of the Cd content in a QW gives rise not only to a growth of the Cd concentration gradient, but also to the growth of strains in the QW. In turn, it can affect both the degradation rate of QWs and the general degradation character of its optical characteristics. Therefore, the elucidation of the details of degradation mechanisms in modern structures on the basis of CdZnSe QWs with various cadmium contents is a challenging task, the solution of which may increase the lifetime of corresponding devices and expand the scope of their possible application.

In this work, the variation of the luminescent characteristics of  $Cd_x Zn_{1-x}Se/ZnSe$  heterostructures with a single QW under irradiation with <sup>60</sup>Co  $\gamma$ -quanta has been studied as a function of the cadmium content in the QW in the range  $x = 0.2 \div 0.4$ . The application of a <sup>60</sup>Co source allowed the Frenkel pairs, as well as electronic excitations, to be generated uniformly over the specimen thickness, thus effectively simulating degradation processes.

# 2. Experimental Specimens and Technique

In the present work, we study  $Cd_xZn_{1-x}Se/ZnSe$  heterostructures with a single  $Cd_xZn_{1-x}Se$  quantum well about 6 nm in thickness and with the cadmium content x = 0.21, 0.23, 0.35, and 0.43 (in different specimens). The thickness of the ZnSe capping layer was about 250 nm, and the thickness of the buffer layer

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was about 1300 nm. Specimens were grown up by the method of molecular beam epitaxy (MBE) on  $n^+$ -GaAs substrates with (100) orientation. The buffer and capping ZnSe layers were grown up at a temperature of 280 °C, provided the existence of a structure of the mixed type ((2×1) and (1×2)), which was determined by the reflection high-energy electron diffraction (RHEED) method. The ratio between the equivalent pressures of molecular beams of the components of groups VI and II was 2:1. When growing the QW, the temperature of a substrate was reduced down to 250 °C. The content and the thickness of  $Cd_xZn_{1-x}$ Se layers were determined from the variation of the reflex intensity oscillation period in the RHEED patterns and monitored by the x-ray diffraction method.

Specimens were studied by the photoluminescence (PL) method. The PL spectra were measured at a temperature of 4.2 K. They were excited by a 337.1-nm line of a pulsed nitrogen laser at a pulse duration of about 7 ns and a pulse-repetition frequency of 100 Hz. The heterostructures were irradiated in air by  $\gamma$ -quanta from a <sup>60</sup>Co source to a dose of  $2 \times 10^8$  rad and at a dose rate of 100 rad/s. In this case, the temperature of specimens did not exceed 60 °C.

## 3. Experimental Results

In Fig. 1, the low-temperature (T = 4.2 K) PL spectra of the heterostructures concerned registered before and after irradiation are depicted. Band  $I_{\rm QW}$  associated with the excitonic radiative transition in the QW dominates in each spectrum. As the content of Cd in the QW increases from x = 0.21 to x = 0.43, the peak position of this band shifts from 2.602 eV (476.5 nm) to 2.398 eV (517 nm), and the half-width grows from 14 to 22 meV (curves 1 and 4, respectively).

Our previous researches of low-temperature PL spectra of those specimens at a low excitation level (by the 325-nm line of a He-Cd laser) [6] showed that band  $I_{\rm QW}$  remains one-component for the specimens with x = 0.35 and 0.43 and transforms into a two-component one in the specimens with x = 0.21 and 0.23. In the latter case, the low-energy component of band  $I_{\rm QW}$  is caused by the emission of excitons bound at shallow donors in the QW, whereas the high-energy one by the emission of excitons localized at fluctuations of the QW potential [6]. In the specimens with x = 0.35 and 0.43, the band  $I_{\rm QW}$  evidently originates from the recombination of excitons localized at fluctuations of the QW potential [6,7]. The increase of the Cd content in the QW leads to the growth of QW composition fluctuations and the en-



Fig. 1. PL spectra of  $\operatorname{Cd}_x \operatorname{Zn}_{1-x} \operatorname{Se}/\operatorname{ZnSe}$  heterostructures with a QW with x = 0.21 (curves 1 and 1'), 0.23 (curves 2 and 2'), 0.35 (curves 3 and 3'), and 0.43 (curves 4 and 4') before (curves 1 to 4) and after (curves 1' to 4') irradiation with  ${}^{60}$ Co  $\gamma$ -quanta to an exposure dose of  $2 \times 10^8$  rad (T = 4.2 K and  $\lambda_{\rm exc} = 337.1$  nm)

hancement of Cd/Zn interdiffusion processes [8]. Both these processes give rise to the growth of QW potential fluctuations and, in turn, to the growth of the QW emission band half-width. This circumstance explains the increase of the band  $I_{\rm QW}$  half-width, which is observed in specimens with single QWs, if the Cd content in the QW becomes higher.

The PL spectra contain also the excitonic emission from ZnSe layers, the intensity of which is two orders of magnitude lower that the band  $I_{\rm QW}$  intensity. Taking into account the magnitude of band-to-band absorption coefficient in ZnSe (of the order of  $10^5 \text{ cm}^{-1}$ ) and a reduction of the diffusion length of nonequilibrium carriers in ZnSe owing to their effective capture and recombination in the QW, we can consider that the observed excitonic emission in ZnSe is mainly excited in the capping layer. In Fig. 2, the low-temperature PL spectra of specimens with x = 0.23 and 0.35 in the region of excitonic emission in ZnSe before and after the irradiation are exhibited. A comparison of these spectra with that of the emission from the ZnSe epitaxial layer, grown by the MBE method on a GaAs substrate [5, 9] demonstrates that they consist of (i) lines of free-exciton emission split by tensile stresses into two components,  $I_{FX}^{hh}$  and  $I_{FX}^{lh}$ , which are connected with the subbands of heavy and light holes, respectively (component  $I_{FX}^{hh}$  manifests itself as a shoulder on the high-energy side of  $I_{FX}$  line); (ii) line  $I_2$  caused by an exciton bound at a neutral donor, and (iii) band  $I_V^0$ , which corresponds to the excitonic emission in the dislocation region. The increase of the



Fig. 2. PL spectra of  $Cd_xZn_{1-x}Se/ZnSe$  structures with a QW with x = 0.23 (a) and 0.35 (b) in the range of excitonic emission by ZnSe before (curves 1) and after (curves 1') irradiation with <sup>60</sup>Co  $\gamma$ -quanta to an exposure dose of  $2 \times 10^8$  rad (T = 4.2 K and  $\lambda_{exc} = 337.1$  nm)

Cd content in the QW gave rise to a shift of excitonic emission lines from ZnSe toward the low-energy spectral range and to the growth of the distance between  $I_{FX}^{lh}$  and  $I_{FX}^{hh}$  lines. Earlier [5], we demonstrated that these effects are caused by the growth of elastic tensile strains in the capping ZnSe layer which appear due to a mismatch between the lattice constants in the QW and ZnSe, as well as by the Cd diffusion from the QW onto the specimen surface during the growth.

After irradiation, the PL spectra from both the QW and ZnSe changed. In the PL spectra of a QW subjected to irradiation, the position of the band  $I_{\rm QW}$  maximum became shifted by about 9 meV toward lower energies for the specimens with x = 0.21 and 0.23 (Fig. 1, curves 1' and 2') and by about 9-13 meV toward higher energies in the specimens with x = 0.35 and 0.43 (Fig. 1, curves 3' and 4'). The integral intensity of the QW emission did not change practically. The band  $I_{\rm QW}$  half-width either did not vary (the specimens with x = 0.21 and 0.43) or grew by 2-3 meV (the specimens with x = 0.23 and 0.43) or grew by 2-3 meV (the specimens with x = 0.23 and 0.35). In the PL spectra from ZnSe layers after irradiation, band  $I_{FX}^{lh}$  shifted by 1.6 - 3.1 meV toward the high-energy spectral range, and the intensity of emission by ZnSe layers decreased a little.

## 4. Discussion

Hence, after the irradiation of CdZnSe/ZnSe QW heterostructures by  $^{60}$ Co  $\gamma$ -quanta to an exposure dose of  $2 \times 10^8$  rad, the shift of the band  $I_{\rm QW}$  maximum of toward either lower or higher energies is observed (the so-called redshift or blueshift, respectively). Similar varia-

tions in the position of the QW emission band were often observed in the course of degradation of blue-green laser diodes on the basis of CdZnSe QWs and also at the simulation of degradation processes in CdZnSe/ZnSe QW heterostructures subjected to the optical and electronic excitations or the thermal annealing. In particular, the researches devoted to the degradation of bluegreen laser diodes on the basis of CdZnSe QWs revealed two degradation processes – "fast" and "slow" [2]. It was demonstrated that the "fast" degradation is caused by the recombination-induced multiplication of dislocations which arise in the vicinity of stacking faults at the ZnSe/GaAs-substrate interface and move into the device active region [2, 10]. The "slow" degradation is caused by the recombination-induced diffusion of pointdefect complexes containing vacancies from nitrogendoped p-layers into the device active region [11, 12]. Both processes are accompanied by the formation of dark-line and dark-spot defects. Cathodoluminescence researches of the degradation of laser diodes on the basis of CdZnSe QWs subjected to the electron-beam irradiation [13, 14] showed that the QW emission band became shifted toward higher energies (the blueshift) in the dark-spot defect region, which was attributed to recombination-induced processes of Cd/Zn interdiffusion across the QW heterointerface. In the dark-line defect region, it shifted toward lower energies (the redshift), which was associated with the relaxation of compressive stresses in the QW at the defect generation. The shift varied from 5 to 50 meV. As a rule, the integral intensity of luminescence was more than an order of magnitude lower in the region of dark-line and dark-spot defects [14], but remained unchanged in some cases [13].

In the majority of works studying the influence of a thermal treatment on the luminescence spectra of a CdZnSe QW, the blueshift of the QW emission band was observed [15–20] and explained by the Cd diffusion from the QW into barrier layers. The fact that the thermal treatment brings about a variation in the spatial distribution of cadmium in the heterostructure was repeatedly confirmed by the secondary ion mass spectrometry (SIMS) method [17–20]. It was also demonstrated that the main mechanism of cadmium diffusion from the QW into the barrier layers is the diffusion via vacancies [18]. After annealing, the magnitude of blueshift could reach 150 meV without an essential (three times at most) decrease of the PL intensity [16]. In this case, the half-width of the QW emission band could either increase or decrease. In some works, the redshift was observed after a thermal treatment [21], which can be explained by a partial relaxation of stresses in the QW.

When analyzing the transformation of PL spectra of specimens under investigation, which were irradiated with  $^{60}$ Co  $\gamma$ -quanta, an analogy can be made with degradation processes described in the literature. Provided the same exposure dose for specimens with a QW, we observe a transition from the redshift to the blueshift with increase of the Cd content in the well. No appreciable drop of the PL intensity is observed in this case. This testifies that new centers of nonradiative recombination are not formed under irradiation. On the basis of the obtained experimental results, we propose the following qualitative explanation for radiation-induced processes occurring in CdZnSe/ZnSe heterostructures with a QW.

Since both the ZnSe layers and the QW in the heterostructures concerned are substantially stressed, the radiation-stimulated relaxation of internal mechanical stresses is possible under irradiation, i.e. the system can transit into a more equilibrium and more stable state, which corresponds to the free energy minimum. The way of the transition into such a state by overcoming the existing energy barriers depends on specific experimental conditions (the type and the dose of radiation, the temperature, and so on), the type of irradiated system, and the presence of the concentration and strain gradients [22, 23].

As was mentioned above, a shift of band  $I_{FX}^{lh}$  into the high-energy spectral region was observed in every specimen subjected to irradiation with  $\gamma$ -quanta. This fact evidences for the relaxation of tensile stresses in the ZnSe layers, which arise due to the mismatch between the lattice constants of a QW and ZnSe. To evaluate the deformation change in the ZnSe layers quantitatively, the following formulas for the energy shifts of free exciton line positions under the action of elastic stresses can be used [9]:

$$\Delta E_{hh} = \left[ -2a \frac{C_{11} - C_{12}}{C_{11}} + b \frac{C_{11} + C_{12}}{C_{11}} \right] \Delta \varepsilon,$$
$$\Delta E_{lh} = \left[ -2a \frac{C_{11} - C_{12}}{C_{11}} - b \frac{C_{11} + 2C_{12}}{C_{11}} \right] \Delta \varepsilon,$$

$$\Delta E_{hh} - \Delta E_{lh} = \left[2b\frac{C_{11} + C_{12}}{C_1 1}\right]\Delta\varepsilon,$$

where  $C_{11} = 8.59 \times 10^{10} \text{ N/m}^2$  and  $C_{12} = 5.06 \times 10^{10} \text{ N/m}^2$  are the elastic shear moduli for ZnSe, a = -5.4 eV and b = -1.20 eV are the hydrostatic and uniaxial, respectively, deformation potentials in ZnSe [24], and  $\varepsilon$  is the amplitude of relative planar elastic deformations ( $\varepsilon > 0$  corresponds to tension and  $\varepsilon < 0$  to compression).

The calculations show that a shift of about 1.6 meVof band  $I_{FX}^{lh}$  toward the high-energy region, which is observed in the specimen with x = 0.23 after irradiation (Fig. 2.a), corresponds to a reduction of relative tensile deformations in the ZnSe capping layer by about  $0.2 \times 10^{-3}$ . In its turn, a shift of about 3 meV of band  $I_{FX}^{lh}$  toward the high-energy region, which is observed in the specimen with x = 0.35 after irradiation (Fig. 2,b), corresponds to a reduction of relative tensile deformations in the ZnSe capping layer by about  $0.4 \times 10^{-3}$ . The formulas presented above for the shift of free exciton line positions in the ZnSe capping layer of initial specimens relative to that in the bulk unstrained ZnSe (2.802 eV [9]) were also used to evaluate the initial tensile strains:  $\varepsilon = 0.92 \times 10^{-3}$  in the specimen with x = 0.21,  $\varepsilon = 1.1 \times 10^{-3}$  for x = 0.23,  $\varepsilon = 1.8 \times 10^{-3}$  for x = 0.35, and  $\varepsilon = 2.3 \times 10^{-3}$  for x = 0.43. One can see that, after the irradiation with  $^{60}$ Co  $\gamma$ -quanta, the relative deformation in the ZnSe capping layer changes by no more than 20%. This fact testifies to only a partial relaxation of stresses in the ZnSe layers under irradiation. We may assume that the stresses relax in the ZnSe layers through the formation (or the transformation) of specifically oriented extended defects (including dislocation loops), owing to the formation of radiation-induced point defects (see below). Such a mechanism of relaxation of internal mechanical stresses is known rather well in the literature [22, 23].

As was indicated above, in the specimens with a QW with x = 0.21 and 0.23 which are subjected to  $\gamma$ irradiation, the redshift of band  $I_{\rm QW}$  is observed. This testifies to a reduction of the compressive stresses in the QW caused by the mismatch between the lattice constants of the QW and ZnSe (Fig. 1, curves 1' and 2'). The formulas given above can be also used to estimate the stress relaxation degree in the QW. The relative compressive strains in the  $Cd_xZn_{1-x}Se$  QWs of initial specimens increase from  $1.5 \times 10^{-2}$  at x = 0.21 to  $3.1 \times 10^{-2}$ at x = 0.43. Such parameters of a  $Cd_x Zn_{1-x}Se$  solid solution, which the QW is composed of, as the lattice constant, the elastic constants, and the deformation potentials were determined by the linear approximation method, taking into account the corresponding values for bulk ZnSe and CdSe (cubic) given in work [24]. In the specimen with x = 0.23, the magnitude of the relative compressive strain in QW,  $\varepsilon \approx 1.66 \times 10^{-2}$ , should result in an increase of the QW bandgap and, respectively, in a shift of the PL band maximum toward the high-energy spectral range by about 21 meV (for a  $Cd_{0.23}Zn_{0.77}Se$ solution, the values  $C_{11} = 7.8943 \times 10^{10} \text{ N/m}^2$ ,  $C_{12} = 4.899 \times 10^{10} \text{ N/m}^2$ , a = -4.95 eV, and b = -1.108 eVwere used). Therefore, if the irradiation gives rise to the redshift of bands  $I_{\rm QW}$  by approximately 9 meV, this means that stresses in the QW relaxed only partially. We can assume that the relaxation of stresses in the specimens with a single QW with x = 0.21 and 0.23 occurs by the same mechanism as that in ZnSe layers, i.e. by the formation or the transformation of specifically oriented extended defects.

The fact that the partial relaxation of stresses in the ZnSe layer occurred in every specimen subjected to irradiation, whereas the red shift of band  $I_{\rm QW}$  was observed only in the specimens with x = 0.21 and 0.23, testifies that another relaxation mechanism dominates in the QWs of specimens with x = 0.35 and 0.43. In particular, the relaxation of stresses in the QW can be partially realized by the Cd/Zn interdiffusion either in the QW plane (the so-called heterointerface smoothing) or across the QW heterointerface. In the latter case, the QW content changes, which gives rise to an increase of the bandgap in a  $Cd_xZn_{1-x}Se$  solid solution. This should lead to the blueshift of QW emission bands, which was really observed in the specimens with x = 0.35 and 0.43. Thus, we observe, for the first time, the change of the strain relaxation mechanism in  $Cd_xZn_{1-x}Se/ZnSe$ QW heterostructures under  $^{6}0$ Co  $\gamma$ -irradiation depending on the composition of the QW solid solution.

For the <sup>60</sup>Co  $\gamma$ -quanta used in this work, the energy of secondary Compton electrons ( $E_{\rm max} \approx 0.6$  MeV) exceeds the collision-induced displacement threshold for Cd, Zn, and Se atoms. As a result, radiation-induced point defects with the concentration  $N_{\rm RID}$   $\approx$  5 ×  $10^{15}$  cm<sup>-3</sup> are formed uniformly over the bulk. Highenergy electrons that arise at  $\gamma$ -irradiation can generate electron-hole pairs. The latter, in turn, can recombine nonradiatively and enhance the migration of initial defects in deformation and concentration fields [25]. Therefore, we may suppose that the irradiation stimulates the formation of new defects and, simultaneously, enhances the migration of intrinsic defects. The migration of existing and radiation-induced defects in the gradients of strain fields in the vicinity of heterointerfaces promotes the enrichment of the latter by defects, including vacancies (vacancies migrate into the compressed region, i.e. into the QW). Most probably, these are vacancies of an element of group II, namely,  $V_{Zn}$ . The generation of  $V_{\rm Zn}$  during irradiation and their radiation-enhanced diffusion into the region of the QW heterointerface should stimulate the process of Cd diffusion from the QW into the surrounding ZnSe layers. The fact that this process is realized in specimens with a single  $Cd_xZn_{1-x}Se$  quantum well with  $x \ge 0.35$  testifies that the main role in the realization of this mechanism belongs to the Cd content in the well, i.e. to the magnitudes of the concentration and strain fields.

The results obtained evidence for a considerable role of elastic deformations at the heterointerface of a QW in degradation processes. Therefore, to enhance the degradation stability of  $Cd_xZn_{1-x}Se/ZnSe$  specimens with a high content of cadmium in the QW, it is necessary to reduce stresses at the QW/barrier heterointerface. For this purpose, the concept of compensation of stresses with different polarities can be used. In particular, in work [26], it was shown that the application of waveguides composed of short-period BeZnSe/ZnSe alternatively strained superlattices in laser diodes with a CdZnSe quantum well as an active region makes it possible not only to provide the effective transport of holes from the  $p^+$ -contact region, but also to redistribute elastic stresses over the waveguide and, in such a way, to reduce the influence of stresses appeared at the QW heterointerface.

### 5. Conclusions

Substantial changes in the recombination characteristics of CdZnSe/ZnSe heterostructures with a single QW under the action of <sup>60</sup>Co  $\gamma$ -quantum irradiation have been experimentally revealed for the first time. It was found that the character of luminescence characteris-

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tic variations depends on the cadmium content in the QW. In particular, in the specimens with x = 0.21 and 0.23, a shift of the QW PL band toward the low-energy range of the spectrum (the redshift) by about 9 meV was observed. At the same time, in the specimens with x = 0.35 and 0.43, a shift of the PL band from the QW toward higher energies (the blueshift) by approximately 9-13 meV was found. We suggest that the redshift is a consequence of the radiation-induced partial relaxation of compressive stresses in the QW, and the blueshift is caused by a reduction of the cadmium content in the QW owing to radiation-stimulated Cd/Zn interdiffusion across the QW heterointerface. At the same time, the PL spectra of ZnSe capping layers after irradiation demonstrate the shift of emission lines of a free exciton connected with the light-hole subband to higher energy by 1.6 - 3.1 meV. This fact evidences for a partial relaxation of tensile stresses in these layers. We suggest that stresses relax in the ZnSe layers and in the QWs with x = 0.21 and 0.23 through the formation (or the transformation) of specifically oriented extended defects, while, in the QWs with  $x \ge 0.35$ , they do due to the diffusion of Cd from the QW into ZnSe layers enhanced by the strain and concentration gradients.

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#### ДЕГРАДАЦІЯ ЛЮМІНЕСЦЕНТНИХ ХАРАКТЕРИСТИК ГЕТЕРОСТРУКТУР CdZnSe/ZnSe I3 КВАНТОВИМИ ЯМАМИ ПІД ДІЄЮ ОПРОМІНЕННЯ γ-КВАНТАМИ

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#### Резюме

У роботі досліджено вплив опромінення  $\gamma$ -квантами <sup>60</sup>Со дозою  $2 \cdot 10^8$  рад на спектри низькотемпературної фотолюмінесценції (ФЛ) гетероструктур  $Cd_x Zn_{1-x}Se/ZnSe$  з одиничними квантовими ямами (КЯ) з різним вмістом кадмію (x = 0, 21; x = 0, 23; x = 0, 35 і x = 0, 43). Показано, що після опромінення відбувається часткова релаксація напружень у КЯ та ZnSe шарах. Виявлено, що механізм релаксації напружень у КЯ залежить від вмісту в ній кадмію. Запропоновано, що релаксація напружень в ZnSe шарі та КЯ з x = 0, 21 - 0, 23 відбувається шляхом утворення (або перетворення) певним чином орієнтованих протяжних дефектів, а в КЯ з x = 0, 35-0, 4 – шляхом дифузії кадмію з КЯ в бар'єрні шари, прискореній деформаційними і концентраційними градієнтами поблизу гетерограниці КЯ/ZnSe.