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THE ROLE OF DEFORMATION AND TEMPERATURE FIELDS IN THE DISTRIBUTION OF DOT DEFECTS AND MODIFICATION OF THE BAND STRUCTURE OF ZnO CAUSED BY PULSED LASER IRRADIATION

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Abstract. The model of self-consistent spatial redistribution of the interstitial zinc and the oxygen vacancies in the ZnO semiconductor under the influence of pulsed laser irradiation in deformation and temperature fields is constructed. The proposed model takes into account the diffusion of dot defects in the nonuniform deformation field created by its own dot defects and temperature gradient, as well as the nonlocal interaction of the interstitial zinc and the oxygen vacancies with atoms of the ZnO crystal lattice. The regularities of spatial redistribution of defects depending on the intensity of laser irradiation, substrate temperature and depth of radiation absorption by the semiconductor are established. Within the proposed model with use of the method of deformation potential the regularities of local shift of edge of the band of conductivity in ZnO under the influence of pulse irradiation are established.

Key words: laser irradiation, diffusion, zinc oxide.

1. INTRODUCTION

Laser-induced periodic surface nanostructures can be generated on almost any material when irradiated with linearly polarized radiation and are formed in the huge range of pulse intervals, ranging from continuous wave irradiation to several femtoseconds [1 – 7]. In experimental work [3] it was shown that their formation occurs when heated to the temperature higher than the melting temperature, and is due to the effect of the long-range of the laser pulse and is explained by the influence of the pressure gradient of the surface acoustic wave. In [2, 4, 7], under the influence of pulsed laser irradiation of Ge, Si and ZnO [7], the nanostructured surface was obtained when it was heated to the temperature lower than the melting temperature. The authors of these works have developed the qualitative model that explains the formation of surface nanostructures due to the emergence of the temperature gradient and, accordingly, diffusion fluxes of interstitial atoms and vacancies in the nonuniform deformation field. Moreover, the interstitial atoms

accumulate near the surface of the irradiated sample, forming the surface superlattice, and vacancies – in the depth of the semiconductor. In works [8, 9] it was shown that nanopores are formed in solids under the influence of pulsed laser irradiation with an intensity greater than some critical value. In particular, in [8] the conditions of laser formation of nanoporous structures with the size of 40 to 50 *nm* were established and it was shown that such clusters of vacancies are more uniform at the distance of several micrometers from the irradiated surface.

Thus, information on the spatial redistribution of dot defects (interstitial atoms and vacancies) under the influence of laser irradiation is important for optimizing the technological process of formation of laser-induced semiconductor nanostructures and predictable control of their physical parameters.

In work [10], in the linear approximation, a theoretical investigation of the redistribution of defects taking into account their nonzero volume based on the stationary equation of diffusion was conducted. The model of "deformation retraction" is proposed, which is based on the flow of defects modified by deformation effects. In the same work it is shown that the deformation flow counteracts the diffusion flow and under certain conditions can exceed it. However, this model does not take into account the effects caused by the nonlocal interaction between defects and matrix atoms. In [11 – 13] the theory of spontaneous nucleation of the surface nanometer lattice was developed, which is caused by instability in the system of adatoms that interact with the self-consistent surface acoustic wave. Within the framework of this theory, which takes into account the nonlocal interaction between defects and matrix atoms and acoustoelectronic interaction [11, 13], the conditions of the formation of nanoclusters on the surface of solids are established and the periods of nanometer lattice as a function of concentration of adatoms, conductivity electrons and temperature are defined. However, the authors of works [10 – 13] do not take into account that under the action of laser irradiation there are deformation gradients due to nonunifirm heating of the material with distance from its irradiated surface. And this factor, as can be seen from the analysis of experimental works [2, 4], often is decisive in the formation of surface nanostructures. In work [14], the model of diffusion in stained solids in the presence of the temperature gradient was developed. However, this theory does not take into account the deformations caused by the defects themselves, which play an important role in their spatial redistribution [10 – 16].

ZnO, due to its unique luminescent, optical and piezoelectric properties, recently has been widely used as the material for bio- and chemosensors, semiconductor lasers of the near ultraviolet and blue ranges, photodetectors [7, 17]. Considerable attention of researchers is paid to laser modification of the morphology of the near-surface ZnO layers, in which the conditions of the formation of nanoclusters are the controlled process. When creating photodetectors and solar cells, the heterojunctions based on *n*-ZnO with the significant discrepancy between the parameters of contacting materials are of particular

interest. Also, the large values of elastic constants indicate that the deformation effects can play an important role in the modification of the near-surface layers.

The ZnO has a complex system of dot defects: interstitial atoms of zinc (Zn_i) and oxygen (O_i), vacancies of zinc (V_{Zn}) and oxygen (V_O), as well as antistructural defects (O_{Zn}, Zn_O) [18, 19]. Under the influence of laser irradiation, pairs of defects are generated in both the oxygen and zinc sublattices. However, the formation energies of vacancy of oxygen and interstitial zinc are the lowest and their concentration is much higher compared to other types of defects [20]. In the case of the significant difference in the ionic radii of elements of the crystal (0,06 *nm* for Zn^{2+} and 0,138 *nm* for O^{2-}), the probability of the formation of Frenkel defects is higher for ions with the smaller radius, i.e. for Zn [21]. On the contrary, the formation of Schottky defects is more energetically advantageous for the oxygen lattice. This explains the presence of the significant concentration of V_O, compared with the concentration of other defects. In [22], the values of self-diffusion coefficients in ZnO were analyzed and it was shown that the diffusion coefficient of vacancies V_{Zn} is much smaller than Zn_i. In addition, another fact that indicates the dominant role of Zn_i and V_O defects in ZnO is that the intrinsic ZnO is an *n*-type semiconductor before and after irradiation [7, 18, 19]. It is known that Zn_i and V_O are donors, and V_{Zn} and O_i are acceptors. Therefore, in the future we will consider only two types of defects: interstitial atoms Zn_i and oxygen vacancies V_O.

In this work, we have developed the model of self-consistent redistribution of dot defects (Zn_i and V_O) in ZnO under the influence of pulsed laser irradiation, which takes into account the diffusion of defects in the nonuniformly deformed field (created both by the presence of defects themselves and by the temperature gradient) and the nonlocal interaction between defects and matrix atoms.

2. THE MODEL

Consider the semiconductor whose surface is exposed to pulsed laser irradiation (fig. 1a). Under the action of laser irradiation, the dot defects (interstitial atoms and vacancies) with an average concentration $N_{0i} = G_{di}\tau_{di}$ (G_{di} is the rate of generation of defects; τ_{di} is the defect lifetime) are generated in the near-surface layer. Nonuniform heating of the matrix to the depth of the crystal is also observed (fig. 1b).

In [23, 24] the spatial distribution of temperature in solids when irradiated with femtosecond laser was investigated.

This distribution is well approximated by the function:

$$T(z) = (T_{\max} - T_{\min})e^{-z^2/l_T^2} + T_{\min}, \quad (1)$$

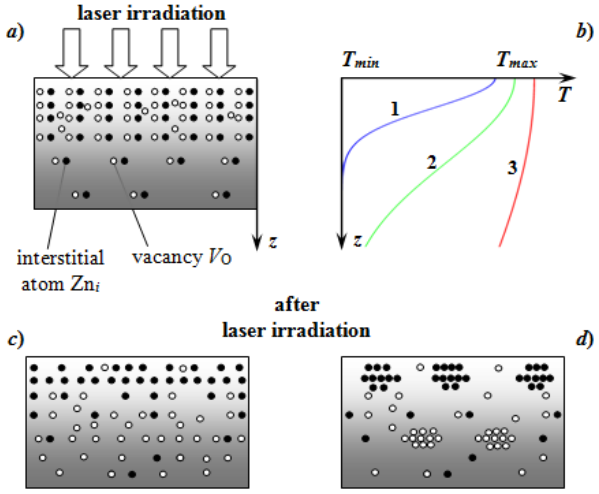

Fig. 1 – The geometric model of ZnO semiconductor exposed to laser irradiation (a), and the spatial distribution of the temperature in the depth of the crystal (b); the geometric model of semiconductor after laser irradiation (the clusters of defects are not formed (c), the clusters of defects are formed (d).

where T_{max} is the the temperature of the irradiated surface (determined by the intensity of laser irradiation and the pulse duration); T_{min} is the temperature of the substrate (in the experiment it is usually room temperature); l_T is the depth of radiation absorption, which is determined by the duration of the laser pulse (curves 1, 2, 3 in fig. 1b) [23]. The small value of the parameter l_T (curve 1 in fig. 1b) corresponds to the shorter duration of the laser pulse and, accordingly, the larger values of the parameter l_T (curves 2 and 3 in fig. 1b correspond to the longer duration of the laser pulse.

Due to the nonuniform heating of the semiconductor, there is the nonuniform deformation field, which creates deformation-diffusion fluxes of defects. Moreover, defects that are centers of stretching (interstitial atoms) move in the region of relative stretching (to the crystal surface), and defects that are centers of compression (vacancies) move in the opposite direction [15] (fig. 1c).

Due to this redistribution near the surface of the crystal increases the concentration of interstitial atoms, and at the certain distance from the surface increases the concentration of vacancies. In works [25, 26] it was shown that in such defect-deformation systems when exceeding some critical value of the concentration of defects $N_{ci} = E \cdot k_B T / \theta_i^2$ due to nonlinear interaction, the separate clusters of defects and their periodic formations are formed (fig. 1d). Here E is the Young's modulus; k_B is the Boltzmann constant; $\theta_i = K\Delta\Omega_i$ is the deformation potential of defects; K is the module of comprehensive compression; $i = 1$ corresponds to interstitial atoms; $i = 2$ corresponds to vacancies.

It is clear that over time, the thermodynamic system will return to equilibrium state (the temperature will equalize). Therefore, the above-described processes can occur when the time of return of the system to equilibrium state is greater than the time during which the diffusion-deformation redistribution of defects occurs. This model uses the adiabatic approximation, which allows us to limit ourselves to considering only the stationary state.

Considering that the deformation is determined by two factors: defects and thermal expansion, the flow of defects can be written as [25, 27]:

$$j_i = -D_i \frac{dN_i}{dz} + N_i \frac{D_i \theta_i}{k_B T}\left(\sum_i \frac{\theta_i}{K}\left(\frac{dN_i}{dz} + \frac{d^3 N_i}{dz^3} r_{da}^2\right) + \alpha\left(\frac{dT}{dz} + \frac{d^3 T}{dz^3} r_{da}^2\right)\right). \quad (2)$$

where α is the temperature coefficient of volume expansion, D_i is the diffusion coefficient; ε is the deformation; r_{da} is the characteristic distance of the interaction of defects with matrix atoms. The second term is the flow of defects in the nonuniform deformation field created by the defects themselves. Here takes into account that $\varepsilon = \sum_i \theta_i N_i / K$. The third term is the flow of defects in the nonuniform deformation field, which arises due to the presence of the temperature gradient.

Then the stationary diffusion equation will take the form:

$$\frac{d}{dz}\left(D_i \frac{dN_i}{dz} - \frac{N_i D_i \theta_i}{k_B T}\left(\sum_i \frac{\theta_i}{K}\left(\frac{dN_i}{dz} + \frac{d^3 N_i}{dz^3} r_{da}^2\right) + \alpha\left(\frac{dT}{dz} + \frac{d^3 T}{dz^3} r_{da}^2\right)\right)\right) + \upsilon \frac{dN_i}{dz} = 0. \quad (3)$$

The last term in (3) expresses the fact that the surface of the semiconductor shifts with velocity υ due to the release of the interstitial atoms to the surface [10].

Given that the deformation potential for vacancies is much smaller than for interstitial atoms ($\theta_2 << \theta_1$), the system of equations (3) can be written as:

$$\frac{d}{dz}\left(D_1 \frac{dN_1}{dz} - \frac{N_1 D_1 \theta_1}{k_B T}\left(\frac{\theta_1}{K}\left(\frac{dN_1}{dz} + \frac{d^3 N_1}{dz^3} r_{da}^2\right) + \alpha\left(\frac{dT}{dz} + \frac{d^3 T}{dz^3} r_{da}^2\right)\right)\right) + \upsilon \frac{dN_1}{dz} = 0. \quad (4)$$

$$\frac{\partial}{\partial z}\left(D_2 \frac{\partial N_2}{\partial z} - \frac{N_2 D_2 \theta_2}{k_B T}\left(\frac{\theta_1}{K}\left(\frac{\partial N_1}{\partial z} + \frac{\partial^3 N_1}{\partial z^3} r_{da}^2\right) + \alpha\left(\frac{\partial T}{\partial z} + \frac{\partial^3 T}{\partial z^3} r_{da}^2\right)\right)\right) + \upsilon \frac{\partial N_2}{\partial z} = 0. \quad (5)$$

Integrating the equations (4) and (5) with taking into account that $dN_i(z \to \infty)/dz = 0$, we obtain:

$$N_i(z) = C_i T^{b_i} \exp\left(\int\left(\frac{T_{\min}}{T N_{ci}}\left(\frac{dN_1(z)}{dz} + \frac{d^3 N_1(z)}{dz^3} r_{da}^2\right) + \frac{b_i}{T}\frac{d^3 T(z)}{dz^3} r_{da}^2 - \frac{\upsilon}{D_i(z)}\right) dz\right). \quad (6)$$

where $b_i = \alpha\theta_i / k_B$ and $N_{ci} = K \cdot k_B T_{\min} / \theta_i \theta_1$. The constants of integration are determined from the condition of preserving the number of defects:

$$\int_0^\infty (N_i - N_{0i}) dz = 0. \quad (7)$$

3. NUMERICAL CALCULATIONS AND DISCUSSION OF RESULTS

Calculations of the spatial redistribution of the concentration of interstitial zinc atoms and oxygen vacancies were performed for ZnO at the following values of parameters: the diffusion coefficient Zn_i: $D_i = 170 \exp(-3{,}3 eV / k_B T)$ cm^2/s [28]; the coefficient of oxygen vacancies in ZnO is presented in work [29]; the temperature coefficient of volume expansion of zinc oxide is presented in work [30]; $K = (c_{11} + 2c_{12})/3 = 0{,}77 Mbar$ [31]; θ_1 = 15 *eV*; θ_2 = -7 *eV*.

In fig. 2 and 3 shows the spatial distribution of interstitial atoms and vacancies at different values of their average concentration and the characteristic distance of interaction of defects with the atoms of the matrix at T_{max} = 800 *K*, T_{min} = 300 *K*, l_T = 5 *nm*. The critical concentration (dashed line), at exceeding of which the processes of self-organization of defects occur, was calculated by the formula [25, 26]:

$$N_{ci} = E \cdot k_B T / \theta_i^2 . \quad (8)$$

As you can see from fig. 2, the interstitial atoms Zn accumulate near the irradiated surface (up to 5 *nm*). Moreover, at an average concentration of interstitial atoms $N_0 = 10^{19}$ cm^{-3} (fig. 2a), their concentration exceeds the critical value only in the near-surface layer with thickness of ~ 2,5 *nm* (at r_d = 0) or in the layer with thickness of ~ 1,5 *nm* (at r_d = 4 *nm*). In these regions of the semiconductor substrate under these conditions can be formed surface defect-deformation structures of interstitial atoms. The parameter r_d depends on the crystal structure, the concentration of defects and the temperature and can be determined from the condition of the minimum of free energy of the defect system [16]. With increasing concentration of interstitial atoms (increasing intensity of laser irradiation, fig. 2b), the surface nanostructures of interstitial atoms can be formed in the near-surface layer with thickness of ~ 6 *nm* at any value of the parameter r_d.

As you can see from fig. 3, due to deformation fluxes, the oxygen vacancies move away from the irradiated surface and accumulate at the distance of (4 – 10) *nm*. Although in the case of strong nonlocal interaction ($r_d \geq 9$ *nm*) the vacancies can also accumulate near the surface of the substrate (curve 3, fig. 3). At the lower intensity of laser irradiation ($N_0 = 10^{19}$ cm^{-3}, fig. 3a), the formation of self-organized nanostructures of vacancies is impossible. With increasing intensity

of laser irradiation ($N_0 = 5 \cdot 10^{19}$ cm^{-3}, fig. 3b), the concentration of vacancies at the distance of (4 – 10) *nm* exceeds the critical value, which leads to the formation of their self-organized structures, such as pores. These results are in good agreement with the experimental results of the works [4, 7].

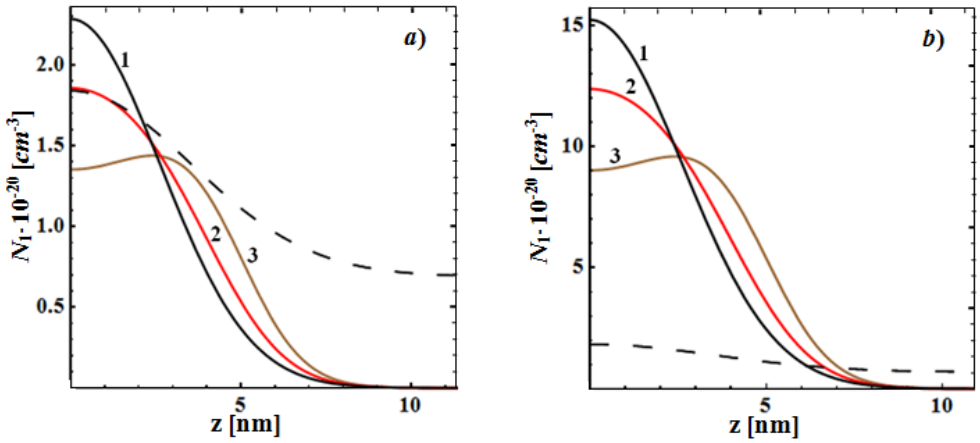

Fig. 2 – The spatial distribution of interstitial atoms of zinc at different values of their average concentration and the characteristic distance of interaction of defects with the atoms of the matrix: 1 – $r_d = 0$; 2 – $r_d = 4$ *nm*; 3 – $r_d = 9$ *nm*; *a*) $N_0 = 10^{19}$ cm^{-3}; *b*) $N_0 = 5 \cdot 10^{19}$ cm^{-3}. The dashed line corresponds to the critical concentration of interstitial atoms.

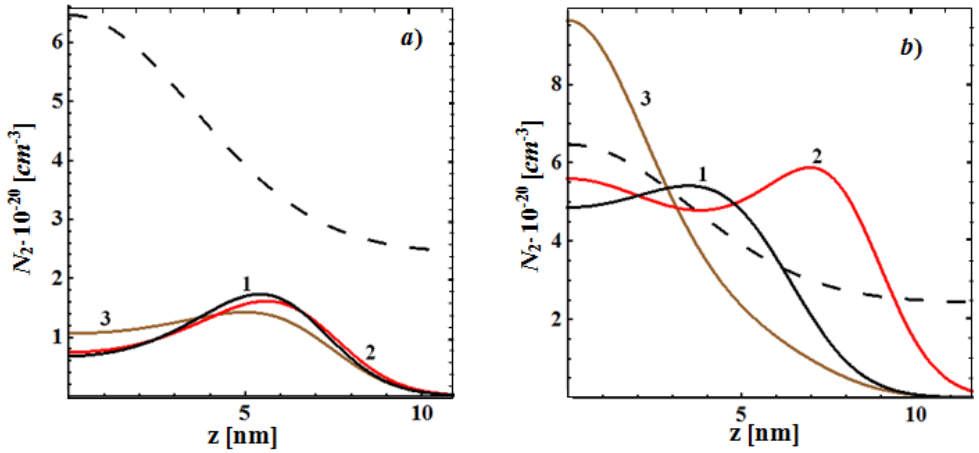

Fig. 3 – The spatial distribution of oxygen vacancies at different values of their average concentration and the characteristic distance of interaction of defects with the atoms of the matrix: 1 – $r_d = 0$; 2 – $r_d = 4$ *nm*; 3 – $r_d = 9$ *nm*; *a*) $N_0 = 10^{19}$ cm^{-3}; *b*) $N_0 = 5 \cdot 10^{19}$ cm^{-3}. The dashed line corresponds to the critical concentration of vacancies.

In fig. 4 and 5 shows the spatial distribution of Zn_i and V_O at different values of substrate temperature and depth of radiation absorption at the average concentration of defects $N_0 = 10^{19}$ cm^{-3}. In the cases of the large depth of radiation absorption (fig. 4a), the concentration of Zn_i exceeds the critical value only near the surface at any substrate temperature (T_{min} = 100 – 300 *K*). When the depth of radiation absorption l_T decreases (determined by the duration of the laser pulse), the near-surface region, where self-organization processes are possible, first narrows to 1-2 *nm*, and at l_T < 4 nm shifts from the surface (fig. 4b). In this case, the formation of clusters is possible at any substrate temperature at the distance (1,5 – 4) *nm*.

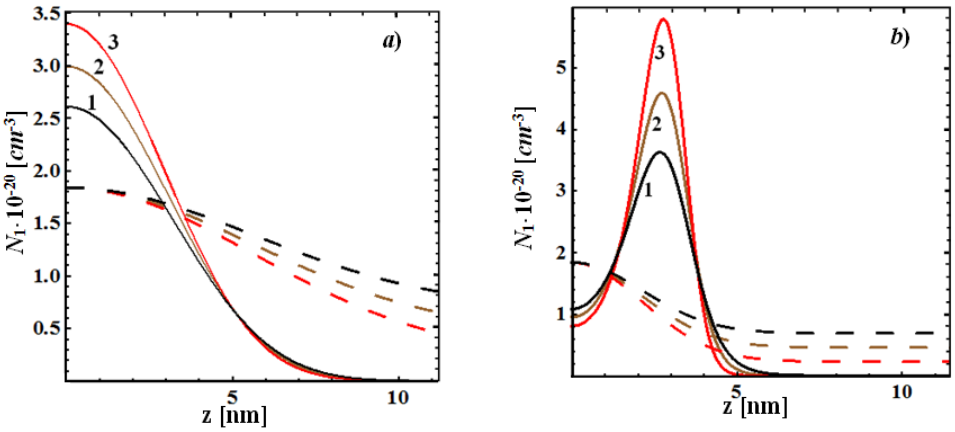

Fig. 4 – The spatial distribution of interstitial atoms of zinc ($N_0 = 10^{19}$ cm^{-3}) at different values of the depth of radiation absorption and substrate temperature:
1 – T_{min} = 300 *K*; 2 – T_{min} = 200 *K*; 3 – T_{min} = 100 *K*; *a*) l_T = 8 *nm*; *b*) l_T = 3 *nm*.

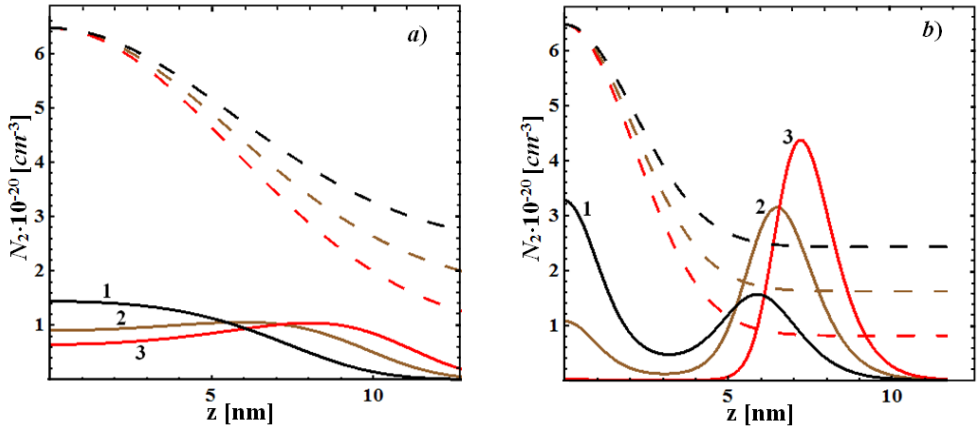

Fig. 5 – The spatial distribution of oxygen vacancies ($N_0 = 10^{19}$ cm^{-3}) at different values of the depth of radiation absorption and substrate temperature:
1 – T_{min} = 300 *K*; 2 – T_{min} = 200 *K*; 3 – T_{min} = 100 *K*; *a*) l_T = 8 *nm*; *b*) l_T = 3 *nm*.

In the case of vacancies (fig.5), depending on the depth of radiation absorption, the defects accumulate at the distance (5 – 15) *nm* from the irradiated surface. However, at $l_T > 7\ nm$ (fig.5a) the processes of self-organization of vacancies are impossible at any temperature. When the characteristic distance of action of the laser beam is reduced to 5 *nm*, the formation of self-organized structures of vacancies is possible only at the low substrate temperature $T_{min} < 120\ K$. When the characteristic distance of action of the laser beam is reduced to 3 nm, the formation of self-organized structures of vacancies is possible at the substrate temperature $T_{min} < 260\ K$ (fig.5b).

4. THE MODIFICATION OF THE BAND STRUCTURE OF ZnO UNDER THE INFLUENCE OF LASER IRRADIATION

Since the defects are the centers of deformation (the interstitial atoms are the centers of deformation of stretching, and the vacancies are the centers of deformation of compression), near the surface there is the nonuniform deformation, which is determined by the formula:

$$\varepsilon = \sum_i \theta_i N_i / K + \alpha \Delta T \,. \tag{9}$$

This deformation leads to the local displacement of the bottom of the conduction band and the top of the valence band:

$$\Delta E_{c(\upsilon)} = a_{c(\upsilon)} \varepsilon \,. \tag{10}$$

and, accordingly, to the localization in different regions of the semiconductor the carriers of different types and the formation of barrier structures (a_c is the constant of hydrostatic deformation potential of the conduction band; a_υ is the constant of hydrostatic deformation potential of the valence band) [1].

The electron subsystem in the ZnO semiconductor is sensitive to deformation. Thus, in work [32] it was shown that comprehensive compression of ZnO leads to the expansion of the band gap and to the displacement of the edge of the conduction band by 20 *meV/GPa* (a_c = -2,15 *eV*). Unalloyed ZnO is the *n*-type semiconductor due to the deviation of the composition from stoichiometric. The presence of a significant quantity of interstitial atoms of Zn and oxygen vacancies due to laser irradiation only reinforces this fact (because the latter are donors). Therefore, we consider the influence of the deformation effects only on the energy displacement of the edge of the conduction band.

In fig. 6 show the coordinate dependence of the bottom of the conduction band (to the depth of irradiated ZnO) at different values of the temperature, the depth of absorption of radiation l_T and the characteristic distance of interaction of defects with the atoms of the matrix r_d.

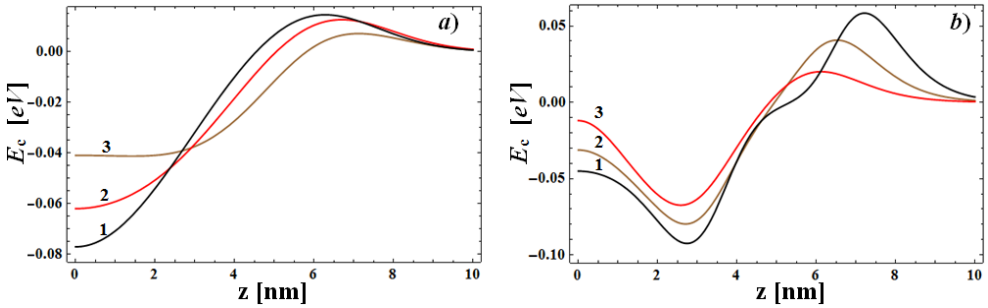

Fig. 6 – The coordinate dependence of the bottom of the conduction band at $N_0 = 5 \cdot 10^{19}$ cm^{-3}
a) T_{min} = 300 K, l_T = 5 nm and at different values of the characteristic distance of interaction of defects with the atoms of the matrix:1 – r_d = 0; 2 – r_d = 4 nm; 3 – r_d = 9 nm
b) r_d = 4 nm, l_T = 3 nm and at different values of the substrate temperature:
1 – T_{min} = 300 K; 2 – T_{min} = 200 K; 3 – T_{min} = 100 K
(the energy is counted from the bottom of the conduction band of undeformed ZnO).

Under the influence of pulsed laser irradiation, due to the self-consistent redistribution of defects and deformation of ZnO material, near the irradiated surface, the lowering of the bottom of the conduction band is observed and, accordingly, the quantum well appears for conduction electrons. Therefore, it is expected that the localization of the negative charge will be observed in the near-surface layer (up to 5 nm) and, conversely, at the distance of (5 – 9) nm from the surface there will be the shortage of negative charge compared to the average value. This effect is better manifested at the lower temperature (fig. 6b) and the smaller values of the characteristic distance of interaction of defects with the atoms of the lattice (fig. 6a). Also, when the parameter l_T decreases (the laser pulse duration decreases), the coordinate of the point of minimum of the conduction band is shifted to the depth of the crystal.

The upward shift of the conduction band in the region (5 – 9) nm from the surface of the crystal is explained by the occurrence of compression deformation in this region due to the localization of oxygen vacancies.

5. CONCLUSIONS

The model of self-consistent deformation-diffusion redistribution of the interstitial atoms of zinc and the oxygen vacancies in the ZnO semiconductor under the influence of pulsed laser irradiation has been developed, which takes into account the diffusion of defects in the nonuniformly deformed field (created both by the presence of the defects themselves and by the temperature gradient) and the nonlocal interaction between defects and matrix atoms.

Within the framework of the developed model, the concentration profiles of the interstitial atoms of zinc and the oxygen vacancies in ZnO at different values of their average concentration (parameter that is determined by the power of laser radiation), substrate temperature and depth of radiation absorption (parameter that is determined by the pulse duration) were investigated.

It is shown that the interstitial atoms of zinc accumulate near the irradiated surface of the semiconductor, and the oxygen vacancies accumulate at the distance $(4 – 10)$ *nm* from it. This result is qualitatively consistent with the experimental data of the work [7].

It is shown that pulsed laser irradiation leads to nonuniform local shift of the edges of the allowed bands and, accordingly, to localization in different regions of the semiconductor of carriers of different type.

Based on the comparison of the concentration profiles of interstitial atoms and vacancies with the critical values of concentrations, when exceeding which the self-organization processes occur, the possibility of formation of the nanoclusters of defects in the ZnO semiconductor is analyzed.

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Conflict of Interest
The authors declare that they have no competing interests.
Competing financial interests: The authors declare no competing financial interests.

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