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FORMATION OF PERIODIC STRUCTURES UNDER THE INFLUENCE OF AN ACOUSTIC WAVE IN SEMICONDUCTORS WITH A TWO-COMPONENT DEFECT SUBSYSTEM

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A deformation-diffusion model describing the formation of periodic structures in semiconductors with a two-component defect subsystem by means of an acoustic wave has been developed. The theory makes allowance for the deformation created by the acoustic wave and point defects. In the framework of this model, a possibility of the ultrasound-stimulated hydrogen passivation of electrically active Cl centers in the CdTe semiconductor and the size dispersion reduction of strained InAs/GaAs quantum dots doped with an isovalent impurity are analyzed.

K e y w o r d s: point defects, acoustic wave, diffusion, deformation.

1. Introduction

The optical and electric properties of semiconductors are known to substantially depend on both the lattice deformation and the spatial distribution of point defects. Such defects can penetrate from outside or arise in the course of crystal growth. Diffusion processes associated with the introduction of impurities into the semiconductor structure also play an important role in the manufacture technology of optoelectronic devices. The character of a deformation of the crystal lattice is governed by both the presence of point defects and the influence of external factors, e.g., an acoustic wave.

It was shown in works [1–3] that an ultrasonic wave can be used to control the transport properties of semiconductors and change their defect structure. This possibility is associated with the diffusion processes of impurity atoms, the formation and decay of complexes, and the clustering of impurity atoms and intrinsic defects in periodic deformation fields. Therefore, the development of a theoretical model that would self-consistently describe diffusion-deformation phenomena in semiconductors with intrinsic point defects and impurities, which are controllable by means of an acoustic wave, is a challenging task.

In works [4, 5], the corresponding theory was developed for the case of one-component defect subsystem, i.e. for the interaction of an acoustic wave with defects of one kind only. However, for a number of practical problems (the passivation of electrically active centers by hydrogen, the creation of an array of homogeneous quantum dots with impurities, the enhancement of the diffusion coefficient, and so forth), the deformation field can be created by both an ultrasonic wave and defects of several kinds. In the case of multicomponent defect subsystem, the flux of intrinsic defects and impurities depends not only on the intrinsic gradient field, but also on the field associated with the gradients of other defect concentrations [6].

In this work, a model of self-consistent deformation-diffusion redistribution of point defects under the influence of ultrasound in semiconductors with a two-component defect subsystem will be developed.

2. Model

Let a semiconductor with point defects of two kinds with the average concentrations N_{d01} and N_{d02} undergo the action of an acoustic wave. The non-uniform deformation of this semiconductor material results from two factors: the acoustic wave action and the non-uniform redistribution of point defects. The expression for the crystal lattice deformation can be presented in the form

$$\varepsilon(x,t) = \varepsilon_0 \cos\left(\omega t - \frac{\omega}{c_l}x\right) + \frac{\theta_{d1}}{K}N_{d1}(x,t) + \\ + \frac{\theta_{d2}}{K}N_{d2}(x,t), \quad (1)$$

where ε_0 is the amplitude of the deformation induced by an acoustic wave with frequency ω, c_l the longitudinal sound velocity, $\theta_{di} = K\Delta\Omega_i$ the deformation potential, K the elasticity modulus, and $\Delta\Omega$ the crystal volume change per i-th ($i = 1, 2$) defect. For interstitial atoms and substitutional impurities with the ionic radius exceeding the ionic radius of matrix atoms, the deformation potential $\theta_{di} > 0$, whereas, for vacancies and substitutional impurities with ionic radius smaller than the ionic radius of matrix atoms, $\theta_{di} < 0$.

Knowing the energy of interaction between a point defect and the elastic continuum,

$$W_{di}(x,t) = -\theta_{di}\varepsilon(x,t), \tag{2}$$

the force acting on the point defect can be determined:

$$F_i = -\frac{\partial W_{di}(x,t)}{\partial x} = \theta_{di}\frac{\partial\varepsilon(x,t)}{\partial x}. \tag{3}$$

Under the action of this force, besides the ordinary diffusion-driven flux of defects, there arises an additional deformation-induced flux

$$j_{di} = \upsilon_i N_{di}, \tag{4}$$

where $\upsilon_i = \frac{D_i\theta_{di}}{kT}\frac{\partial\varepsilon(x,t)}{\partial x}$ is the defect velocity emerging as a result of the strain gradient in the semiconductor, D_i is the diffusion coefficient, and k the Boltzmann constant. In view of Eq. (4), the system of equations for diffusing defects acquires the form

$$\frac{\partial N_{di}(x,t)}{\partial t} = D_i\frac{\partial^2 N_{di}(x,t)}{\partial x^2} - \frac{D_i\theta_{di}}{kT}\frac{\partial}{\partial x}\left(N_{di}(x,t)\frac{\partial\varepsilon(x,t)}{\partial x}\right) + G_{di} - \frac{N_{di}(x,t)}{\tau_{di}}, \tag{5}$$

where G_{di} is the rate of point defect generation, which depends on the ultrasound intensity and the energy of defect formation in the semiconductor; and τ_{di} is the defect lifetime, which is determined by the relation $\tau_{di} = \nu_i^{-1}\exp(\Delta E_i/kT)$, where $\nu_i = = 10^{12}\div10^{13}$ Hz is the frequency of thermal defect vibrations, and $E_i = 1\div4$ eV is the potential barrier height [6].

Let us write the defect concentration in the form

$$N_{di}(x,t) = N_{d0i} + N_d^{(i)}(x,t), \tag{6}$$

where $N_d^{(i)}(x,t)$ is its spatially non-uniform component. Taking Eq. (1) into account and making the approximation $N_d^{(i)} \ll N_{d0i}$, we obtain the following system of equations for $N_d^i(x,t)$:

$$\frac{\partial N_d^{(1)}(x,t)}{\partial t} = D_1\frac{\partial^2 N_d^{(1)}(x,t)}{\partial x^2} - D_1\frac{N_{d01}}{N_{dc1}}\left(\frac{\partial^2 N_d^{(1)}(x,t)}{\partial x^2} + \frac{\theta_{d2}}{\theta_{d1}}\frac{\partial^2 N_d^{(2)}(x,t)}{\partial x^2} - \frac{K\varepsilon_0}{\theta_{d1}}\left(\frac{\omega}{c_l}\right)^2\cos\left(\omega t - \frac{\omega}{c_l}x\right)\right) + G'_{d1} - \frac{N_d^{(1)}(x,t)}{\tau_{d1}}, \tag{7}$$

$$\frac{\partial N_d^{(2)}(x,t)}{\partial t} = D_2\frac{\partial^2 N_d^{(2)}(x,t)}{\partial x^2} - D_2\frac{N_{d02}}{N_{dc2}}\left(\frac{\partial^2 N_d^{(2)}(x,t)}{\partial x^2} + \frac{\theta_{d1}}{\theta_{d2}}\frac{\partial^2 N_d^{(1)}(x,t)}{\partial x^2} - \frac{K}{\theta_{d2}}\varepsilon_0\left(\frac{\omega}{c_l}\right)^2\cos\left(\omega t - \frac{\omega}{c_l}x\right)\right) + G'_{d2} - \frac{N_d^{(2)}(x,t)}{\tau_{d2}}, \tag{8}$$

where $N_{dci} = \frac{K\cdot kT}{\theta_{di}^2}$ and $G'_{di} = G_{di} - \frac{N_{d0i}}{\tau_{di}}$.

In order to find a solution of the system of differential equations (7) and (8), let us apply the integral Laplace transformation

$$X_i(x,p) = \int_0^\infty N_d^{(i)}(x,t)e^{-pt}dt. \tag{9}$$

Then the system of equations (7) and (8) reads

$$pX_1 - N_d^{(1)}(x,0) = D_1\left(1 - \frac{N_{d01}}{N_{dc1}}\right)\frac{\partial^2 X_1}{\partial x^2} + \frac{G'_{d1}}{p} - \frac{X_1}{\tau_{d1}} - D_1\frac{N_{d01}}{N_{dc1}}\frac{\theta_{d2}}{\theta_{d1}}\frac{\partial^2 X_2}{\partial x^2} + D_1\frac{N_{d01}}{N_{dc1}}\frac{K}{\theta_{d1}}\varepsilon_0\left(\frac{\omega}{c_l}\right)^2\times\left(\frac{p}{p^2+\omega^2}\cos\left(\frac{\omega}{c_l}x\right) + \frac{\omega}{p^2+\omega^2}\sin\left(\frac{\omega}{c_l}x\right)\right), \tag{10}$$

$$pX_2 - N_d^{(2)}(x,0) = D_2\left(1 - \frac{N_{d02}}{N_{dc2}}\right)\frac{\partial^2 X_2}{\partial x^2} + \frac{G'_{d2}}{p} - \frac{X_2}{\tau_{d2}} - D_2\frac{N_{d02}}{N_{dc2}}\frac{\theta_{d1}}{\theta_{d2}}\frac{\partial^2 X_1}{\partial x^2} + D_2\frac{N_{d02}}{N_{dc2}}\frac{K}{\theta_{d2}}\varepsilon_0\left(\frac{\omega}{c_l}\right)^2\times\left(\frac{p}{p^2+\omega^2}\cos\left(\frac{\omega}{c_l}x\right) + \frac{\omega}{p^2+\omega^2}\sin\left(\frac{\omega}{c_l}x\right)\right), \tag{11}$$

where $N_d^{(i)}(x,0)$ is the spatially non-uniform component of the initial distribution of defect concentration.

By finding the solution of the system of differential equations (10) and (11), which is finite as $x \to \pm\infty$ and satisfies the initial condition $N_d^{(i)}(x,0) = 0$, and applying the inverse Laplace transformation, we obtain the space-time redistribution of point defects of both kinds, $N_{d1}(x,t)$ and $N_{d2}(x,t)$, under the influence of an acoustic wave and the strain distribution $\varepsilon(x,t)$ created by both ultrasound and point defects.

3. Results of Numerical Calculations and Their Discussion

1. In Fig. 1, the spatial redistributions of Cl impurities (curve *1*) ($N_{d0\mathrm{Cl}} = 5 \times 10^{19}$ cm^{-3}) and hydrogen (curves *2* to *4*) in the CdTe semiconductor under the influence of an acoustic wave are presented for various hydrogen concentrations and at various temperatures. The numerical calculations were carried out with the following parameters: $\omega = 10^8$ s^{-1}, $\varepsilon_0 = 10^{-5}$, $c_l = 3570$ m/s, $\theta_{d\mathrm{Cl}} = 13$ eV, $D_{\mathrm{Cl}} = 1.7 \times 10^5 \exp\left(-\frac{0.56}{kT}\right)$ cm^2/s [6], and $D_{\mathrm{H}} = 1.5 \times 10^{-4} \exp\left(-\frac{0.12}{kT}\right)$ cm^2/s [7]. At those values, the condition $1/\tau_{di} \ll \frac{D_i \theta_{di} \varepsilon}{kT} \left(\frac{\omega}{c_l}\right)^2$ is satisfied. In this case, the last term in Eq. (5) can be neglected ($\tau_{di}^{-1} = 0$). In the calculations, we also supposed no defect generation under the influence of ultrasound ($G_{di} = 0$). The calculations were carried out for the point defect concentration in the interval $N_{d0i} < N_{dci}$, because, at higher concentrations, the nonlinear interaction of point defects with the field of elastic deformations [4, 8, 9] becomes essential, which was not taken into account in the model concerned.

Under the action of an external periodic nonuniform deformation in the semiconductor material, there arises, besides the gradient flux, an additional deformation-induced flux of defects. It leads to the formation of periodic defect-deformation structures with the period equal to the ultrasound wavelength. As one can see from Fig. 1, the oscillation amplitude for the hydrogen impurity concentration increases with its average concentration. At the same time, despite that the average concentration of the hydrogen impurity changes, the spatial distribution of the chlorine impurity practically remains constant. The temperature variation results in the amplitude change of defect concentration oscillations and the shift of the defect structures created by the hydrogen and chlorine impurities with respect to each other. The temperature reduction gives rise to the increase of the oscillation amplitude for the defect concentrations. This result follows from the fact that, as the temperature decreases, the velocity of defects in the deformation field increases. Simultaneously, the temperature change leads to the variation in the diffusion coefficients of the chlorine and hydrogen impurities and, accordingly, to the spatial shift of the positions of their concentration maxima. The calculations were carried out for the time moment $t \gg \frac{1}{D_i} \frac{N_{dci}}{N_{dci} - N_{d0i}} \left(\frac{c_l}{\omega}\right)^2$. In this case, the curves for $N_d^{(i)}$ shift in time, but their relative spatial shift remains invariable.

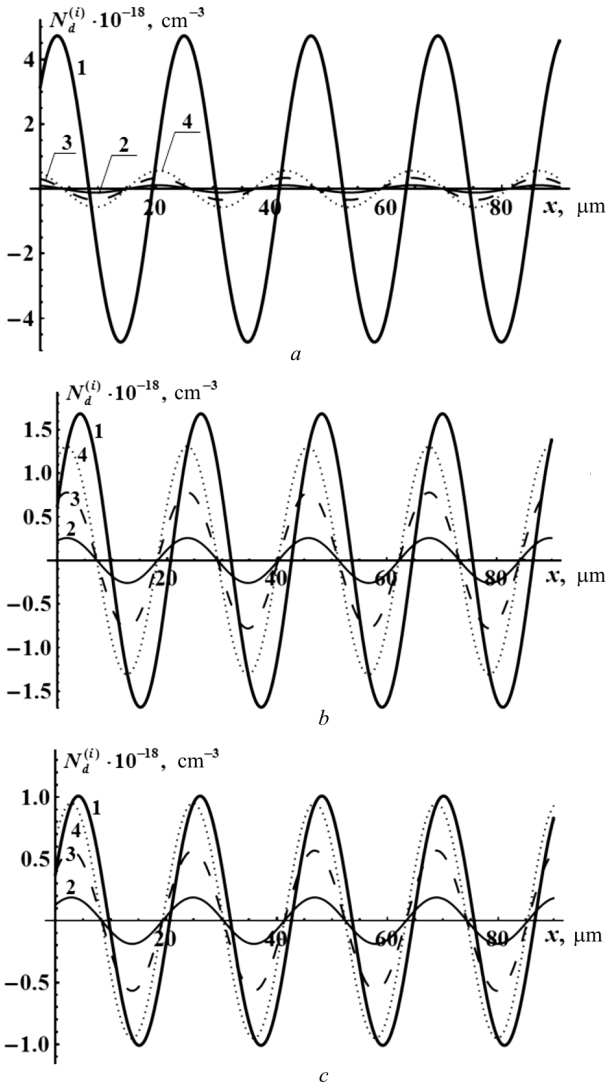

Fig. 1. Spatial redistributions of the chlorine (curve *1*) and hydrogen (curves *2* to *4*) impurities under the influence of an acoustic wave for various values of their average concentration $N_{d0\mathrm{H}} = 10^{20}$ (2), 3×10^{20}, and 5×10^{20} cm^{-3} (*4*) and at various temperatures $T = 300$ (*a*), 600 (*b*), and 900 K (*c*)

In the framework of the model concerned, let us analyze a possibility to stimulate the passivation of electrically active Cl centers by hydrogen in the CdTe semiconductor with the help of ultrasound. The hydrogenation of heterostructures and bulk semiconductors gives rise to the photoluminescence intensity

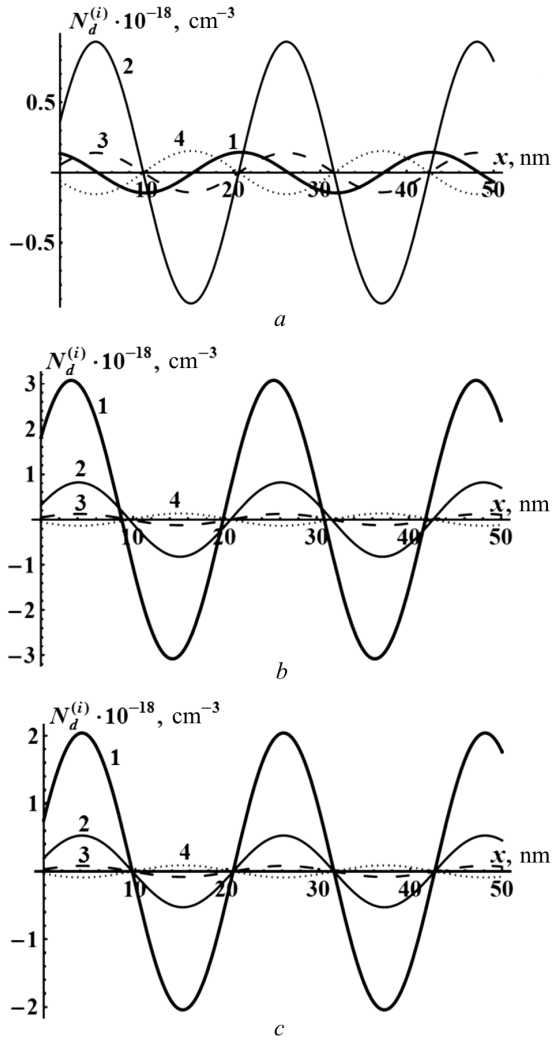

Fig. 2. Spatial redistributions of In (*1*), Bi (*2*), Sb (*3*), and P (*4*) at various temperatures $T = 450$ (*a*), 600 (*b*), and 900 K (*c*)

growth, which can be explained by the hydrogen-induced passivation of non-radiative recombination centers [1, 10–13]. However, in heterostructures with stressed quantum wells, the passivation degree of defects is much lower [10, 13]. It is so because the stressed layers can slow down the diffusion of defects and impurities. A considerable difference between the diffusion coefficients of defects and hydrogen atoms also promotes their accumulation in different regions of the material and, accordingly, a reduction of the passivation degree.

The creation of an additional strain gradient under the influence of ultrasound should promote the emergence of directed defect and impurity fluxes. On the other hand, the optimum choice of parameters (the temperature and the concentration of hydrogen atoms) should allow the impurities and defects to redistribute in such a manner that the effect of defect passivation would be maximum.

In particular, at the temperature $T = 900$ K and the hydrogen concentration $N_{d0\mathrm{H}} = 5 \times 10^{20}$ cm^{-3}, the accumulation regions for the chlorine and hydrogen impurities almost coincide (Fig. 1, *c*; curves *1* and *4*). Hence, at the given parameters, the effect of the hydrogen-induced passivation of electrically active centers (the chlorine impurities) is maximum. The obtained results qualitatively agree with the experimental data of work [1], where it was found that the ultrasonic treatment increases the intensity of photoluminescence in Si, if the latter was treated in hydrogen plasma. In this case, at higher temperatures of the ultrasonic treatment, the degree of defect passivation by hydrogen increases.

2. On the basis of the developed model, let us analyze the possibility to reduce the size dispersion of strained InAs/GaAs quantum dots (QDs) doped with an isovalent impurity with the help of ultrasound.

Recently, a new approach has been developed to control the properties of semiconductor heterostructures with QDs. It is based on the introduction of a single impurity atom into the QD [14–17]. One of the methods to form the ordered arrays of QDs is the self-organization of QDs on a crystal surface. The epitaxial growth kinetics and the distribution of deformations in the QD-matrix of a system affect the size, shape, and arrangement of QDs in the matrix. The exclusive properties of the structures with QDs reveal themselves only in the case where the QDs are uniform enough by their shapes and dimensions. Therefore, the main task at the QD growing is the control over their morphology: the average size, density, uniformity, and so on. In works [14, 17], it was found experimentally that the introduction of silicon [17] and bismuth [14] impurities considerably improves the optical properties of InAs/GaAs QDs. In work [16], the theory of deformation was developed for QDs doped with an isovalent impurity. The theory explained experimental data on the creation of an array of uniform InAs QDs doped with Bi [14]. Bismuth is not an electrically active impurity, i.e. it does not increase the charge carrier concentration. However, it substantially changes the conditions of QD formation

owing to the induced diffusion-deformation flux. The direction of an atomic flux is governed by signs of the deformation potential and the strain gradient (formula (4)). In other words, defects that play the role of stretching centers ($\theta_{di} > 0$) are accumulated in the region with a relative stretching deformation of the materal, whereas defects that are squeezing centers ($\theta_{di} < 0$) are accumulated in the region with the relative squeezing deformation. Since the covalent radius of In is larger than that of Ga, In atoms can be regarded as stretching centers. The Bi impurity is also a stretching center. Therefore, the deformation-induced flux of In atoms promotes their localization in the QDs, which, accordingly, leads to the restriction of their migration mobility and to a higher uniformity of QD sizes.

Evidently, the creation of a non-uniform deformation by an ultrasonic wave at the growth of InAs/GaAs QD with an impurity induces additional deformation fluxes of In atoms, which would result in their higher localization in the QDs, provided a proper choice of corresponding parameters.

Let us consider a semiconductor structure with an InAs/GaAs QD doped with an isovalent impurity. The latter is an As-atom substitutional defect. Depending on impurity's ionic radius r_i, the increase of the QD volume amounts to $\Delta\Omega_i = \frac{4}{3}\pi(r_i^3 - r_{\mathrm{As}}^3)$. The ionic radii of In, Ga, and As, as well as isovalent impurities Bi, Sb, and P, are listed in Table.

Numerical calculations were carried out for the following parameters: $\omega = 10^{11}$ s^{-1}, $\varepsilon_0 = 0.00001$, $K = 6500$ eV/nm^3, $c_l = 3500$ m/s, $D_{\mathrm{In}} = 6 \times 10^5 \exp\left(-\frac{4}{kT}\right)$ cm^2/s [6], and $D_{\mathrm{P}} = 1.26 \times 10^2 \exp\left(-\frac{2.7}{kT}\right)$ cm^2/s [7]. There are no data in the scientific literature concerning the temperature dependences of the diffusion coefficients for Bi and Sb in InAs. Therefore, in calculations, we used the same values as for P.

Ion	Ionic radius, r_i, nm
Ga^{3+}	0.062 [6]
In^{3+}	0.092 [6]
As^{3+}	0.069 [6]
Bi^{3+}	0.12 [6]
Sb^{3+}	0.082 [6]
P^{3+}	0.044 [18]

In Fig. 2, the spatial redistributions of In, which is a substitutional defect, with the concentration $N_{d0\mathrm{In}} = 10^{20}$ cm^{-3} (curve *1*) and the isovalent impurities Bi, Sb, and P (curves *2, 3*, and *4*, respectively) in the GaAs matrix under the action of an acoustic wave at various temperatures are shown. In the temperature interval 600–900 K (Figs. 2, *b* and *c*), the ultrasonic wave favors the accumulation of In and Bi (Sb) in the same spatial regions. This fact should promote the size uniformity enhancement over the QD array. The strain gradient is much larger if Bi impurities are available, which is associated with a larger difference between the ionic radii of Bi^{3+} and As^{3+} in comparison with that between Sb^{3+} and As^{3+}. On the contrary, In and P are accumulated in different spatial regions, which should result in a larger dispersion of the QD sizes. This conclusion follows from the fact that the ionic radius of P^{3+} is smaller than that of As^{3+}, so that the P impurities are localized at the strain minima.

However, by lowering the temperature ($T < 500$ K), it is possible to obtain a partial overlapping of the regions, where the P and In impurities are accumulated (Fig. 2, *a*). In this case, on the contrary, a delocalization of In atoms around the Sb and Bi impurities is observed. The largest shift of the In concentration maximum with respect to impurities amounts to a quarter-wavelength.

The obtained results qualitatively agree with the experimental data of work [19]. Namely, it was found that the ultrasonic treatment of semiconductor quantum dots for 5–90 min in the course of their synthesis allows one to obtain nanoclusters that are more uniform by dimensions.

4. Conclusions

A theory describing the formation of periodic defect-deformation structures under the influence of acoustic waves in semiconductors with a two-component defect subsystem has been developed. It is found that, under the influence of an acoustic wave with the optimal choice of parameters (hydrogen concentration, temperature, and frequency and amplitude of ultrasound), the effect of the defect passivation by hydrogen becomes stronger. It is shown that the ultrasonic treatment of structures with quantum dots should enhance the procedure efficiency, while creating an array of uniform quantum dots, by using the doping with the Bi isovalent impurity.

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ФОРМУВАННЯ ПЕРІОДИЧНИХ СТРУКТУР ПІД ВПЛИВОМ АКУСТИЧНОЇ ХВИЛІ У НАПІВПРОВІДНИКАХ З ДВОКОМПОНЕНТНОЮ ДЕФЕКТНОЮ ПІДСИСТЕМОЮ

Р е з ю м е

Побудовано деформаційно-дифузійну модель формування періодичних структур під впливом акустичної хвилі у напівпровідниках з двокомпонентною дефектною підсистемою. Запропонована теорія враховує деформацію, створену акустичною хвилею та точковими дефектами. У межах даної моделі проаналізовано можливість стимулювання ультразвуком пасивації електрично активних центрів Cl воднем у напівпровіднику CdTe та зменшення дисперсії розмірів напружених квантових точок InAs/GaAs, легованих ізовалентною домішкою.