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The influence of deformations on single electron states in a molecule formed from three quantum dots of the heterosystem InAs/GaAs

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A molecule consisting of three quantum dots, whose centers form a triangle of the InAs/GaAs heterosystem, was studied. A numerical calculation of the energy spectrum of an electron in a molecule formed from three spherical quantum dots was carried out. The influence of deformations on the electron energy depending on the size of the nanocrystals, the distance between them, and the symmetry of the quantum molecule were investigated. The case of symmetry of an equilateral and isosceles triangle is considered.

Keywords: three quantum dots, quantum molecule, deformation.

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Introduction

Deformation effects significantly impact the energy states of particles in nanosystems. In particular, on the baric coefficient value, the charge carriers' effective mass, and degradation effects. Quantum dots (QDs) are formed under the influence of the field of internal elastic deformation. Therefore, deformation's influence must be considered when determining the energy spectrum of quasiparticles in nanosystems formed from several QDs.

Coherent electron tunneling transitions occur in closely spaced nanocrystals. As a result, configurations are formed that are connected by common electrons. They were called artificial molecules. The simplest example of an artificial molecule is a tunnel-bonded KT pair. It can be used to implement a qubit. For this, you can use the spin [1] or the charge [2] of quasiparticles of the nanosystem.

Nanosystems consisting of three tunnel-bonded quantum dots or "triatomic" artificial molecules allow studying interesting phenomena related to electrostatics and molecular states of the triple quantum molecule (QM). Experimentally, molecules from quantum dots were intensively studied [3-8]. There may be two [3], three [4-6], four [7], or more nanocrystals in the QM. If we consider the geometry of the molecule, it can be linear [6-7] or triangular [4-5]. In turn, the first can be symmetrical, when the distances between QDs are the same, and not symmetrical.

The changes in the bottom of the conduction band (the top of the valence band) at the quantum dot-matrix heterojunction is due to the difference in the band gap width of the QD and matrix materials. This forms a profile of the quantization potential, which has the character of a three-dimensional potential well. In works [9-11], the profile of a three-dimensional potential well created by QD was modeled as rectangular without taking into account the deformation potential. The depth was determined only due to the different widths of forbidden bands of the material of the matrix and QD.

In the case of heterosystems with stressed QDs in the presence of elastic stress fields around them, the depth of the quantization potential is also determined by the nature of the deformation of the matrix and QDs. In particular, the stresses arising in the InAs/GaAs nanoheterosystem with an array of InAs QDs significantly affect the structure of the allowed zones and their discontinuity [12]. For an isolated QD, deformations were taken into account in works [13-14].

The proposed work is devoted to the study of the influence of deformations on the electron energy in a quantum molecule formed from three spherical quantum dots of the InAs/GaAs heterosystem.

I. Formulation and solution of the problem

Consider a quantum molecule consisting of three spherical InAs/GaAs QDs that are tunnel-connected to each other. We will choose the coordinate system so that the centers of two QDs lie on the OZ axis, and the point O is in the middle between these QDs. The third QD is placed on the OY axis at the distance *d* from the origin (Fig. 1). In particular, the stresses arising in the InAs/GaAs nanoheterosystem with an array of InAs QDs significantly affect the structure of the allowed zones and their discontinuity [7]. Since the lattice constant of indium arsenide is a_{InAs} = 6.08 *Å*, and that of gallium arsenide is a_{GaAs} = 5.65 Å, it follows that the QD material is subjected to all-round compression deformation, and the matrix is tensile.

One QD can be considered as an elastic sphere of radius R, which is placed in the spherical cavity of the matrix. The difference between the volumes of the sphere and the cavity of the matrix is $\Delta V=4\pi R^3 f$, where $f = (a_{InAs} - a_{GaAs})/a_{InAs} = 0.07$ is the mismatch parameter of the fixed lattices. This deformation leads to a change in the relative depth of the potential well of one quantum dot:

$$\Delta U = \left|\alpha_c^{(1)}\varepsilon^{(1)}\right| + \left|\alpha_c^{(2)}\varepsilon^{(2)}\right|,$$

where $\varepsilon^{(1)} = Sp\ \varepsilon^{(1)}$, $\varepsilon^{(2)} = Sp\ \varepsilon^{(2)}, \boldsymbol{\varepsilon}$ – is the strain tensor defined in works [13-14]. $\alpha_c^{(1)} = -5.08eB$; $\alpha_c^{(2)} = -7.17eB$ – are constants of the hydrostatic deformation potential of the conduction zone.

Let there is one electron in the heterosystem. Then the Hamiltonian of this system in the effective mass approximation has the form

$$\hat{H} = -\frac{\hbar^2}{2}\nabla\frac{1}{m(\boldsymbol{r})}\nabla + U(\boldsymbol{r}), \qquad (1)$$

Where

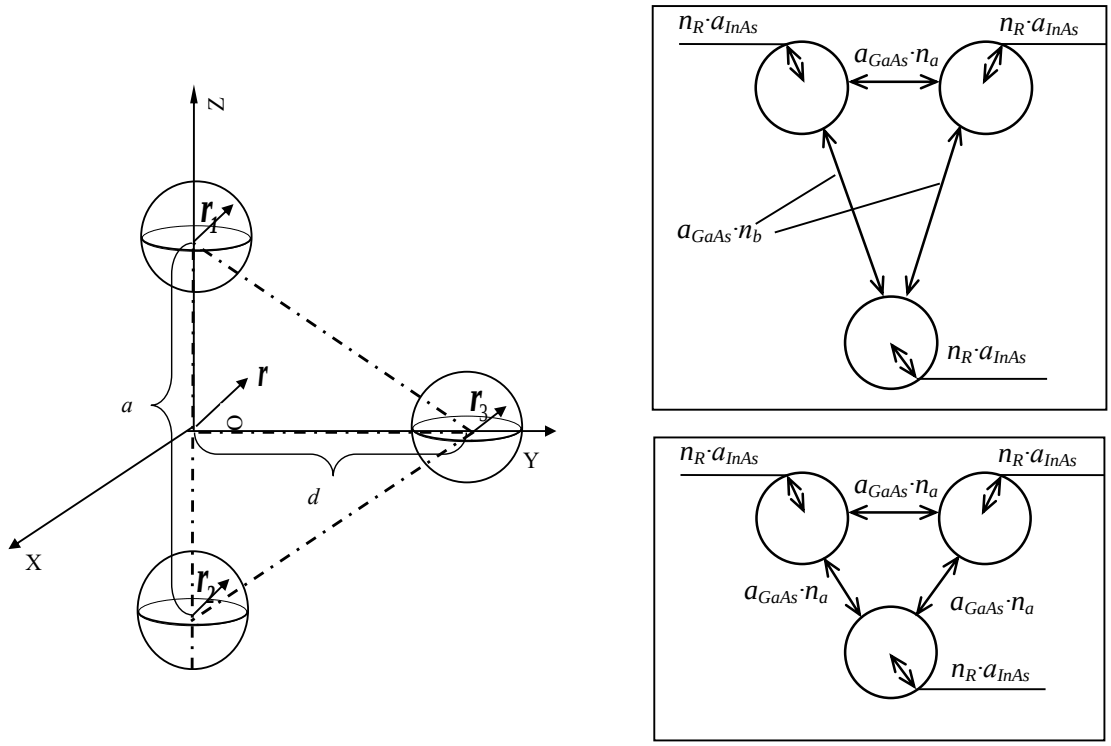

Fig. 1. Model of the studied nanosystem. The insets show the change in the parameters of the heterostructure (the upper inset shows an equilateral triangle, and the lower one shows an isosceles triangle).

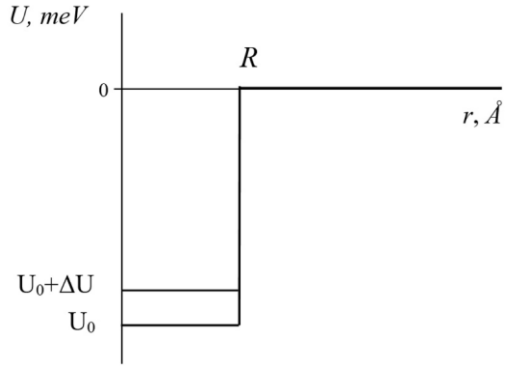

Fig. 2. Electron energy in one isolated QD, taking into account deformations.

$$U(\boldsymbol{r}) = \begin{cases} U_0 + \Delta U, & if\ \boldsymbol{r}\ are\ in\ any\ QD \\ 0, & if\ \boldsymbol{r}\ runs\ over\ the\ matrix \end{cases}, \qquad (2)$$

$U_0 = \chi_2 - \chi_1 < 0$, $\chi_1 = 4.9eB$; $\chi_2 = 4.07eB$ (Fig. 2.), $m(\boldsymbol{r})$ is electron effective mass for the corresponding region,

$$m(r) = \begin{cases} m_1, & if\ \boldsymbol{r}\ are\ in\ any\ QD, \\ m_2, & if\ \boldsymbol{r}\ are\ not\ in\ the\ QD, \end{cases}$$

To solve the problem, we will use the approximation of a linear combination of quantum well orbitals. For this, the wave function is represented as a linear combination of the electron wave functions of individual QDs:

$$\psi(\boldsymbol{r}) = C_1\psi_1(\boldsymbol{r}) + C_2\psi_2(\boldsymbol{r}) + C_3\psi_3(\boldsymbol{r}), \qquad (3)$$

where $\psi_i(\boldsymbol{r})$ is the wave function of the ground state of the i-th QD, which in the Cartesian coordinate system has the form:

$$\psi_1(x,y,z) = \begin{cases} A_1 \frac{\sin k_1\sqrt{x^2+y^2+(z-\alpha)^2}}{\sqrt{x^2+y^2+(z-\alpha)^2}}, \text{for } x^2+y^2+(z-\alpha)^2 \le R_1^2 \\ B_1 \frac{\exp(-\chi_1\sqrt{x^2+y^2+(z-\alpha)^2})}{\sqrt{x^2+y^2+(z-\alpha)^2}}, \text{for } x^2+y^2+(z-\alpha)^2 > R_1^2 \end{cases},$$

$$\psi_2(x,y,z) = \begin{cases} A_2 \frac{\sin k_2\sqrt{x^2+y^2+(z-\alpha)^2}}{\sqrt{x^2+y^2+(z-\alpha)^2}}, \text{for } x^2+y^2+(z-\alpha)^2 \le R_2^2 \\ B_2 \frac{\exp(-\chi_2\sqrt{x^2+y^2+(z-\alpha)^2})}{\sqrt{x^2+y^2+(z-\alpha)^2}}, \text{for } x^2+y^2+(z-\alpha)^2 > R_2^2 \end{cases}, \qquad (4)$$

$$\psi_3(x,y,z) = \begin{cases} A_3 \frac{\sin k_3\sqrt{x^2+y^2+(z-\alpha)^2}}{\sqrt{x^2+y^2+(z-\alpha)^2}}, \text{for } x^2+y^2+(z-\alpha)^2 \le R_3^2 \\ B_3 \frac{\exp(-\chi_3\sqrt{x^2+y^2+(z-\alpha)^2})}{\sqrt{x^2+y^2+(z-\alpha)^2}}, \text{for } x^2+y^2+(z-\alpha)^2 > R_3^2 \end{cases},$$

$$A_i = \frac{1}{\sqrt{\frac{R_i}{2} - \frac{\sin(k_iR_i)}{4k_i} + \frac{\sin^2(k_iR_i)}{2\chi_i}}},$$

$$B_i = \frac{\sin(k_iR_i)}{\exp(-\chi_iR_i)},$$

$$\alpha > R_1,\ \ \alpha > R_2,\ \ d > R_3;$$

$$k_i = \sqrt{\frac{2m_1}{h^2}\left|U_0 - E_i^{one}\right|}, \qquad \chi_i = \sqrt{\frac{2m_2}{h^2}\left|E_i^{one}\right|},$$

a is the distance between the centers of QDs lying on the OZ axis, d is the distance from another QD to the OZ axis, R_i is the radius of the i-th QD, and E_i^{one} is the electron energy in the i-th isolated QD.

Taking into account (3) and (4), the Schrödinger equation can be reduced to a system of three algebraic equations:

$$\begin{cases} C_1(H_{11} - E) + C_2(H_{12} - ES_{12}) + C_3(H_{13} - ES_{13}) = 0 \\ C_1(H_{21} - ES_{21}) + C_2(H_{22} - E) + C_3(H_{23} - ES_{23}) = 0, \\ C_1(H_{31} - ES_{31}) + C_2(H_{32} - ES_{32}) + C_3(H_{33} - E) = 0 \end{cases} \qquad (5)$$

where, $H_{ij} = \int \varphi_i^*(r)\hat{H}\varphi_j(r)d\tau$, $S_{ij} = \int \varphi_i^*(r)\varphi_j(r)d\tau$ $i,j = 1,2,3$.

The system of homogeneous equations (5) has a nonzero solution when the determinant of this system is equal to zero:

$$\begin{vmatrix} (H_{11} - E) & (H_{12} - ES_{12}) & (H_{13} - ES_{13}) \\ (H_{21} - ES_{21}) & (H_{22} - E) & (H_{23} - ES_{23}) \\ (H_{31} - ES_{31}) & (H_{32} - ES_{32}) & (H_{33} - E) \end{vmatrix} = 0. \qquad (6)$$

Equation (6) is the dispersion equation for the electron in three QDs molecules.

II. Analysis of the effect of deformation on the energy spectrum of an electron in a molecule with three QDs

Numerical calculations were performed for *InAs* QDs placed in a *GaAs* matrix. The effective mass of an electron in these materials is $m_1 = m_{AlAs} = 0{,}15m_0$ and $m_2 = m_{GaAs} = 0{,}063m_0$, m_0 is a free electron mass. The band gap in the heterosystem, which determines the depth of the potential well, without taking into account deformations, is equal to 830 meV. Let's consider QDs of such sizes that there is one bound electronic state in them (18 Å $\le R_i \le$ 30 Å). Modeling of CM from three quantum dots was carried out for:

a) symmetrical arrangement (QD centers were at the vertices of an equilateral triangle);

b) unsymmetrical (QD centers were at the vertices of an isosceles triangle).

In all these cases, the radii of the three CTs were chosen to be the same $R_1=R_2=R_3=R$.

If we consider the symmetrical placement of three QDs (option a) forming the QM (Fig. 3 and Fig. 4), then the energy spectrum of the studied nanosystem is affected by two parameters: the distance between the boundaries of the single crystals $\alpha = 2 \cdot R + n_\alpha \cdot \alpha_{GaAs}$ and their size $R = n_R \cdot \alpha_{InAs}$. In fig. 3 shows the dependence of the energy spectrum of the CM formed from three QDs on the distance between the boundaries of the nanocrystals. As can be seen from fig. 3, taking deformations into account significantly changes the electron energy for all states. Thus, with a distance between the boundaries equal to two permanent GaAs lattices, the ground state increases by 84.2 meV, and the first excited state increases by 90 meV. The size of the splitting also changes, for this distance, it increases by 5.9 meV. This can be explained by the reduction of the potential well, which is a consequence of taking deformations into account. As a result, tunneling of particles occurs at greater distances than without considering deformations. For non-stressed QDs at a distance between QD boundaries of six permanent GaAs lattices, the splitting is about 2 meV, and taking into account deformations at a distance of seven permanent GaAs lattices – 2.2 meV.

When analyzing the influence of the QD size on the electron energy in a QM of three nanocrystals located at the vertices of an equilateral triangle (case a), it should be noted that as the size of the nanocrystals increases, the difference between the electronic states with strained and unstrained QDs increases significantly. If $R = 3 \cdot \alpha_{InAs}$, then the difference between the energy levels of the

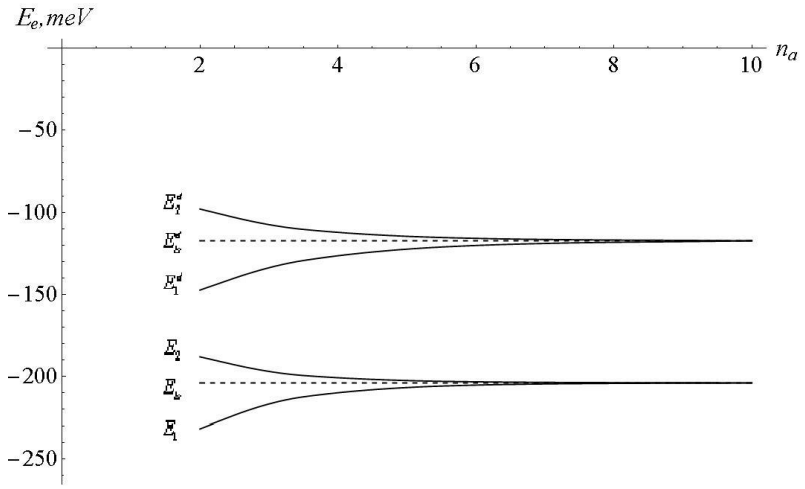

Fig. 3. Electron energy in the QM as a function of the distance between the boundaries of the nanocrystals at the symmetry of an equilateral triangle with (E_{is}^d , E_1^d , and E_2^d) and without taking into account deformations (E_{is} , E_1 and E_2) (the distance between QDs in a stable GaAs lattice, and their radii are equal to 4 of a stable InAs lattice). E_{is} , E_{is}^d is the energy of one isolated QD without deformation and with deformation, respectively.

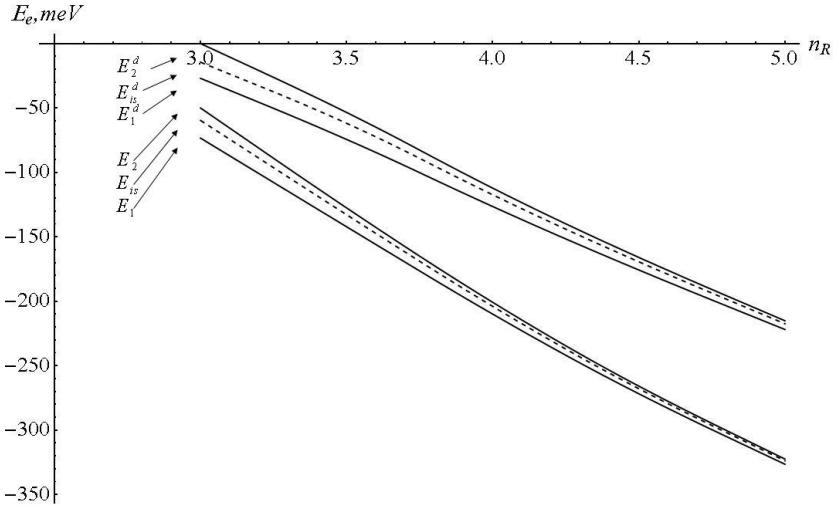

Fig. 4. Electron energy in the QM as a function of the size of the nanocrystal at the symmetry of an equilateral triangle with (E_{is}^d , E_1^d and E_2^d) and without taking into account deformations (E_{is} , E_1 and E_2) (the distance between QDs is equal to 4 constant GaAs lattices, and their radii are equal to constant InAs lattices). E_{is} , E_{is}^d is the energy of one isolated QD without deformation and with deformation, respectively.

electron for the ground state is 46.4 meV, for the first excited state - 59 meV, and the splitting value has increased by 4 meV. If $R = 5 \cdot \alpha_{InAs}$, then the difference will be 104.5 meV for the ground state, 107.3 meV for the first excited state, and the splitting value will increase by only 2 meV.

The non-symmetric arrangement of QDs in the QM of three nanocrystals (case b) removes the degeneracy of the first excited state [11], so the heterosystem will be characterized by three energy levels. Two options for changing the geometry of the heterosystem are considered:

b1) the distance between the boundaries of two QDs changes when the third one is at the same distance to the axis passing through the centers of these QDs (Fig. 5);

b2) the centers of two QDs are fixed, and the third is shifted relative to the axis on which the fixed nanocrystals are located (Fig. 6).

As in the case of a symmetric QD arrangement (option a), taking into account the deformations increases all electron energy levels in a given CM and the case of an asymmetric arrangement (option b). In the case of b1)

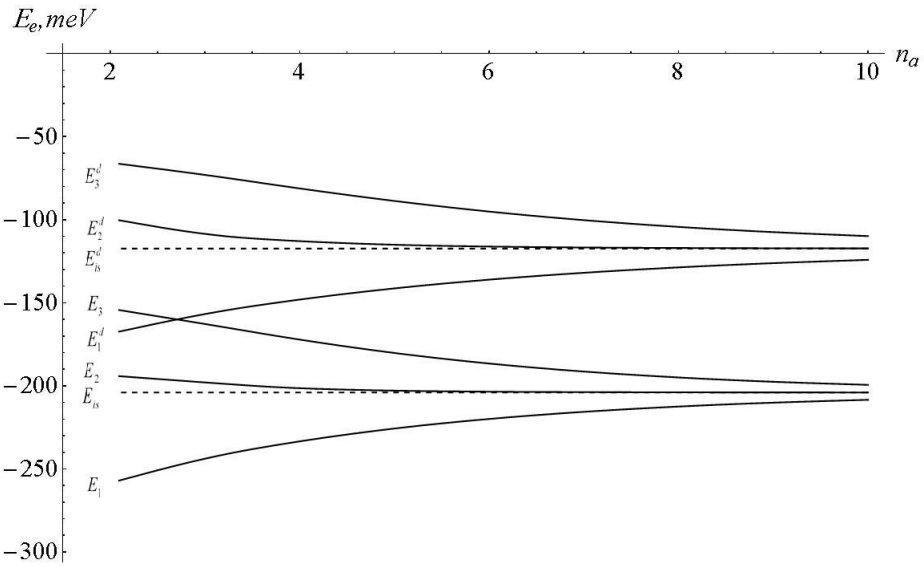

Fig. 5. Electron energy in the QM as a function of the distance between the boundaries of two QDs when the third one is at the same distance to the axis passing through the centers of these QDs with the symmetry of an isosceles triangle taking into account (E_{is}^d , E_1^d , E_2^d and E_3^d) and without taking into account deformations (E_{is} , E_1 , E_2 and E_3) (the distance of the third QD to the axis is 4 permanent GaAs lattices, and the radii of all are equal to 4 permanent InAs lattices). E_{is} , E_{is}^d is the energy of one isolated QD without deformation and with deformation, respectively.

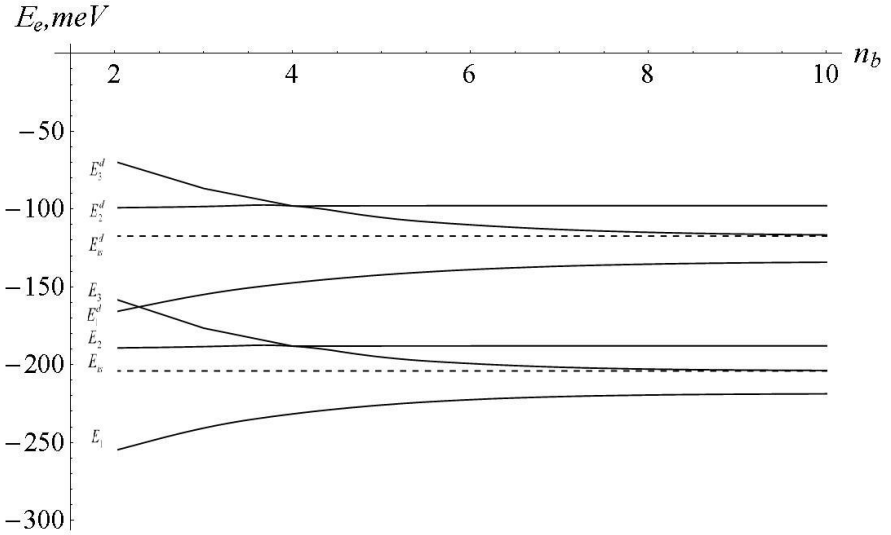

Fig. 6. The energy of an electron in a QM as a function of the distance between the boundaries of one QD and two others whose centers are fixed, with the symmetry of an isosceles triangle taking into account (E_{is}^d , E_1^d , E_2^d and E_3^d) and without taking into account deformations (E_{is} , E_1 , E_2 and E_3) (the distance between two QDs is equal to 4 constant GaAs lattices, and the radii of all are equal to 4 constant InAs lattices). E_{is} , E_{is}^d is the energy of one isolated QD without deformation and with deformation, respectively.

arrangement of nanocrystals in the CM, at a distance between the boundaries of two QDs, three stable GaAs lattices, the ground state becomes higher by 87 meV, the first excited by 89.4 meV, and the second excited by 89.7 meV. The difference between the levels increases, and between the main and the second excited becomes larger by 2.4 meV. Between the excited states, the electron energy differences for stressed and strained QDs are almost the same. In case b2), when the distance from the boundary of the third QD to the boundaries of two fixed ones is three constant GaAs lattices, the ground state differs by 85.8 meV, the first excited state by 90.1 meV, and the second excited state by 89.8 meV. The difference between the ground and the second excited state increases by 4 meV.

Conclusions

A molecule of three quantum dots, whose centers form a triangle of the InAs/GaAs heterosystem, taking into account deformation effects, was studied. It is shown that in the case of QDs located at the vertices of an equilateral triangle, uniform convergence of QDs causes splitting of energy levels with the formation of two (one non-degenerate and one doubly degenerate). This result was obtained both with and without deformation. However, taking the strain into account increases the splitting energy. A similar result was obtained for the case of a QM consisting of QDs located at the vertices of an isosceles triangle. However, in this case, the number of levels is three (all non-degenerate). This is explained by the lower symmetry of an isosceles triangle compared to an equilateral triangle.

Therefore, taking into account the deformation of the QD matrix in the QM causes a change in the value of the energy level splitting. These results should be taken into account when analyzing the interlevel and interband absorption of electromagnetic waves of different ranges, as well as the luminescence of heterosystems with quantum molecules. Also, the proposed model can be expanded to multilayer QDs in applied fields[15] which form the QM. These studies will be implemented in our next works.

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Вплив деформацій на одноелектронні стани в молекулі, утвореній із трьох квантових точок гетеросистеми InAs/GaAs

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Досліджено молекулу з трьох квантових точок, що своїми центрами утворюють трикутник гетеросистеми InAs/GaAs. Проведено чисельний розрахунок енергетичного спектра електрона в молекулі, що утворена з трьох квантових точок сферичної форми. Досліджено вплив деформацій на енергію електрона в залежності від величини нанокристалів, відстані між ними та симетрії квантової молекули. Розглянуто випадок симетрії рівностороннього та рівнобедреного трикутника.

Ключові слова: три квантові точки, квантова молекула, деформація.