ENERGY SPECTRUM OF A CHARGE OF QUANTUM DOTS WITH DIFFERENT SHAPES

V.I. BOICHUK, I.V. BILYNSKY, I.O. SHAKLEINA

Ivan Franko Drohobych State Pedagogical University
(24, Ivan Franko Str., Drohobych 82100, Ukraine; e-mail: ioshak@mail.ru)

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The article presents a theoretical analysis of the splitting of the energy levels of a charge (an electron, a hole) by group theory for quantum dots of cubic, hexagonal, and tetrahedral shapes in the cases where a particle spin equals 0 or 1/2. The energies and the wave functions of states for quantum dots with different shapes are evaluated by perturbation theory. As a basis, a quantum dot with spherical symmetry is chosen, and an exact value of the energy of a charge in it is found. The specific calculations are performed for the GaAs/AlAs and InAs/GaSb heterostructures.

1. Introduction

Significant progress in nanotechnologies has stimulated the beginning and intensive development of nanoelectronics as a constituent part of the new field of physical researches, namely the physics of low-dimensional structures. Therefore, a great number of reports focus in recent years on the growth and investigation of nanoscale heterosystems of quantum dots (QDs) and quantum wires (QWs) of different shapes [1–10].

The attention was paid to QDs and QWs with surfaces in the form of a sphere [4–6], cylinder [7,8], cube [12], ellipsoid [13–15], regular six-facet prism [16,17], and pyramid [18–21].

Experimental researches prove that the shape of a quantum dot essentially influences the energy spectra of an electron, hole, exciton, and phonon. In turn, the optical and energetic properties of heterosystems depend on the energy spectrum.

Theoretical studies which aim to treat the experimental data consider real heterostructure models. In the framework of these models, certain assumptions were made concerning the shapes of QDs or QWs, their size distribution, and the interaction with a matrix and with each other. Even within the model of noninteracting identical quantum dots (wires), complex numerical calculations are frequently required, because an analytical solution of the Schrödinger equation is not available or is critically simplified. In addition, the fact that the above-mentioned QDs and QWs are characterized by a certain symmetry of the shape is left beyond attention.

The objective of the present investigation is to examine the influence of a QD shape on the splitting of energy states of charged quasiparticles by using group theory and to determine the value of splitting of these states in QDs with different shapes by perturbation theory. A quantum dot with spherical symmetry is taken as a basis, for which an exact value of the energy of an electron or a hole is found as a solution of the Schrödinger equation.

2. Analysis of the Schrödinger Equation of a QD Charge by Group Theory

First, we consider a quantum dot located in the domain G, whose sizes in various directions do not differ essentially. The Hamiltonian of a particle is

$$\hat{H} = \hat{H}_p + U(\mathbf{r}). \tag{1}$$

For an electron in the approximations of effective mass and parabolic conduction band, we have

$$\hat{H}_p = -\frac{\hbar^2}{2}\nabla\frac{1}{m(r)}\nabla. \tag{2}$$

For a hole, the form of $\hat{H}_p$ depends on the number of considered bands [10, 22–24]. In the six-band approximation, provided one neglects the corrugation of the energy surface,

$$\hat{H}_p = \frac{1}{m_0} \begin{vmatrix} P+Q & L & M & 0 & i\sqrt{1/2}L & -i\sqrt{2}M \\ L^* & P-Q & 0 & M & -i\sqrt{2}Q & -i\sqrt{3/2}L \\ M^* & 0 & P-Q & -L & -i\sqrt{3/2}L^* & -i\sqrt{2}Q \\ 0 & M^* & -L^* & P+Q & -i\sqrt{2}M^* & i\sqrt{1/2}L^* \\ -i\sqrt{1/2}L^* & i\sqrt{2}Q & i\sqrt{3/2}L & i\sqrt{2}M & P-\Delta & 0 \\ i\sqrt{2}M^* & -i\sqrt{3/2}L^* & i\sqrt{2}Q & i\sqrt{1/2}L & 0 & P-\Delta \end{vmatrix}. \tag{3}$$

The potential energy of a particle is

$$U(\mathbf{r}) = \begin{cases} -U_0, & x,y,z \in G \\ 0, & x,y,z \notin G. \end{cases} \tag{4}$$

By introducing a virtual spherical QD with radius R of the same "volume", we can write Hamiltonian (1) as

$$\hat{H} = \hat{H}_p + U_{\rm sph}(\mathbf{r}) + (U(\vec{r}) - U_{\rm sph}(\mathbf{r})) \equiv$$

$$\equiv \hat{H}_p + U_{\rm sph}(\mathbf{r}) + W(\mathbf{r}) = \hat{H}_0(\mathbf{r}) + W(\mathbf{r}), \tag{5}$$

where $W(\mathbf{r})$ is the correction and

$$U_{\rm sph}(\mathbf{r}) = \begin{cases} -U_0, & x^2+y^2+z^2 < R, \\ 0, & x^2+y^2+z^2 \geq R. \end{cases}$$

For the Hamiltonian $\hat{H}_0(\mathbf{r})$, the Schrödinger equation

$$\hat{H}_0(\mathbf{r})\,\Psi^0(\mathbf{r}) = E^0\,\Psi^0(\mathbf{r}) \tag{6}$$

possesses the exact solution, and the wave functions of states $\Psi^0(\mathbf{r})$ are represented by products of the radial and spherical functions $R_{n\,j}(r)\,Y_{j\,m}(\vartheta,\varphi)$.

The functions $Y_{j\,m}(\vartheta,\varphi)$ can be chosen as the basis of a spherical symmetry group representation [25]. Since the characters of irreducible representations of elements of the same class are identical, it is sufficient to consider a rotation around one of the axes, the z-axis. With a turn around the z-axis by an angle φ, the wave functions Y_{jm} are multiplied by $e^{im\,\varphi}$, and the matrix of the representation looks as

$$D_j(\varphi) = \begin{pmatrix} e^{i\,j\,\varphi} & 0 & 0 & ... & 0 \\ 0 & e^{i(j-1)\,\varphi} & 0 & ... & 0 \\ ..... & & & & \\ 0 & 0 & 0 & ... & e^{-i\,j\,\varphi} \end{pmatrix}. \tag{7}$$

It is seen from (7) that the character $\chi_j(\varphi)$ of the representation D_j is defined as

$$\chi_j(\varphi) = \sum_m D_{j,\,mm}(\varphi) = \sum_{m=-j}^{j} e^{im\varphi} = \frac{\sin(j+\frac{1}{2})\,\varphi}{\sin\frac{\varphi}{2}}. \tag{8}$$

For small angles of rotation φ, the character $\chi_j = 2\,j+1$ is equal to the dimension number of the representation D_j.

First, we neglect a particle spin, which is completely justified in a number of cases. If a particle spin is zero, then $j = l$, l = 0,1,2,3 ... We perform the symmetry analysis of the Schrödinger equation for the Hamiltonians with different symmetries. First, we consider a QD with cubic shape. If one compares the characters of representations of spherical and cubic symmetry groups, it is seen that the states with l =0,1 (s- and p-states) of the Hamiltonian $\hat{H}_0(\mathbf{r})$ with regard for a perturbation W do not split. The account for the cubic symmetry of a quantum dot changes only the energy value of every state under consideration. For states with $l > 1$, we observe the splitting of levels, because the corresponding representations become reducible if one takes into account the cubic symmetry correction (W). By expanding each representation D_l into irreducible parts, one can define the required splitting.

The calculations show that $D_0 = A_1$, $D_1 = F_1$, $D_2 = E+F_2$, $D_3 = A_2+F_1+F_2$, where A_1, A_2, E, F_1, F_2 are the representations of the cube group.

Thus, in a cubic QD, d-states are split into two ones: one twofold and one threefold degenerate, and f-states are split into three ones: one twofold and two threefold degenerate.

Consider the quantum dots with surfaces in the form of a regular six-facet prism. Let the QD sizes differ slightly in different directions. In this case, the Hamiltonian of an electron can again be chosen in form (5), where $\hat{H}_0(\mathbf{r})$ is characterized by spherical symmetry.

The whole Hamiltonian of the system is characterized with D_6, whose symmetry is set by six irreducible representations (four one-dimensional and two two-dimensional ones).

The calculation of the splitting of states for a QD under consideration shows that $D_1 = A_1 + E_2$, $D_2 = A_1 + E_1 + E_2$, $D_3 = A_2 + A_3 + A_4 + E_1 + E_2$.

The consideration of the hexagonal lattice structure of a quantum dot leads to the splitting of p-states into two ones: one non-degenerate and one twofold degenerate. States with $l = 2$ are also split, but into three states: two twofold degenerate and one non-degenerate. Meanwhile, f-states are split into five ones: three non-degenerate and two twofold degenerate.

As is known, heterosystems of quantum dots can be obtained by different technologies. In particular, in the heterosystems fabricated by the Stranski–Krastanow technique, QD are assumed to have the form of a pyramid or a truncated pyramid [18–21]. Consider the transformation of spherical symmetry group states at the transition to the tetrahedron symmetry group.

The calculations carried out for a QD of the considered shape show that $D_1 = F$, $D_2 = E_1 + F$, $D_3 = A + 2F$. Therefore, the "inclusion" of a tetragonal symmetry perturbation does not split degenerate p-states; d-states are split into two non-degenerate states and one threefold degenerate, and f-states are split into one non-degenerate and two threefold degenerate ones.

We now study how the splitting of energy levels in QDs with different shapes changes under the condition that a particle spin is $s = 1/2$. The characters $\chi_j(\varphi)$ of the corresponding representations $D_j(j = l + s, |l - s|)$ are also defined by (8).

First, we examine a QD with cubic shape. Similarly to the case where $s = 0$, we get that the states with $j = 1/2, 3/2$ are not split: $D_{1/2} = E_1'$, $D_{3/2} = G'$. For the states with $j = 5/2, 7/2$, we get the following splitting of levels: $D_{5/2} = E_2' + G'$, $D_{7/2} = E_1' + E_2' + G'$. Thus, if a particle spin $s = 1/2$, then the states with $j = 7/2$ are split into two ones: twofold degenerate and fourfold degenerate; whereas the states with $j = 1/2$ are split into three ones: two twofold degenerate and one fourfold degenerate.

Consider a QD with its surface in the form of a regular six-facet prism. We obtain that, in the QD under study, the state with $j = 1/2$ is not split: $D_{1/2} = E_1'$, i.e., taking the hexagonal lattice structure into account changes only the energy of the state. For the states with $j > 1/2$, the following splitting is observed: $D_{3/2} = E_1' + E_3'$, $D_{5/2} = E_1' + E_2' + E_3'$, $D_{7/2} = E_1' + 2E_2' + E_3'$. Thus, the states with $j = 3/2$ are split into two twofold degenerate; the states with $j = 5/2$ are split into three twofold degenerate, and the states with $j = 7/2$ in a hexagonal QD are split into four twofold degenerate states.

For a pyramidal QD, the analysis of the splitting of states by group theory results in the following: the state $D_{1/2}$ is not split ($D_{1/2} = E_1'$), the states with $j = 3/2$ are split into two twofold degenerate states ($D_{3/2} = G_1' + G_2'$), the states with $j = 5/2$ are split into three twofold degenerate ones ($D_{5/2} = E' + G_1' + G_2'$); and the states with $j = 7/2$ are split into four twofold degenerate states ($D_{7/2} = 2E' + G_1' + G_2'$).

3. Energy of Electron States in QDs with Different Shapes

The present general analysis can be specified by calculating the charged particle state energies in QDs of different surfaces. With regard for the above consideration concerning the "potential" W, it is possible to use perturbation theory. In order to specify further calculations, we will consider the electron energy spectrum.

We calculate the electron state energy in QDs of different shapes for two heterosystems: GaAs/AlAs and InAs/GaSb. The Hamiltonian of the problem is given by (5), and $\hat{H}_0$ has form (2). In this case, we can neglect the electron spin. The solution of Eq.(6) is a function

$$\Psi^0(\mathbf{r}) = R_{n\,l}(r)\, Y_{l\,m}(\vartheta, \varphi). \tag{9}$$

The radial wave function is written differently depending on the region: $r < R, r \geq R$. For the bound states, if $E^0 < 0$, we have

$$R_l(r) = A\, j_l(kr), \quad r \leq R,$$

$$R_l(r) = B\, h_l^{(1)}(xr), \quad r > R, \tag{10}$$

where $k = \sqrt{\frac{2m_1}{\hbar^2}(E^0 + V_0)}$, $x = \sqrt{\frac{2m_1}{\hbar^2}E^0}$, $E^0 < 0$, $j_l(x)$, $h_l^{(1)}(x)$ are Bessel and Hankel functions [26].

The energy of quantum states is found from the matching conditions for the wave function

$$A j_l(k\,R) = B\, h_l^{(1)}(x\,R),$$

$$\frac{1}{m_1} A j_l'(k\,R) = \frac{1}{m_2} B\, h^{(1)\prime}_l(x\,R). \tag{11}$$

To determine the electron energy, we use perturbation theory. We define the value of the first correction to the energy caused by a perturbation W for QDs of cubic,

tetrahedral, and hexagonal shapes. The formation of the correction value is based on two competitive factors. First, as the analysis shows, the function $W = W(x, y, z)$ is a complicated function of coordinates which takes both positive and negative values in a small region at the interface of the media. Outside this interval, $W = 0$. A reduction of the QD volume leads to the effective increase in the "capacity" of the potential W. Second, the volume reduction is accompanied by an increase in the energy of a charge and, hence, by a decrease of the particle location probability density in the actual region of space where $W \neq 0$.

Consider first a QD with cubic shape. Note that states with l =1,2 (p- and d-states) are degenerate. Therefore, in order to calculate the first correction to the energy ΔE_l, it is necessary to find the matrix elements of $(W - \Delta E'_l)$ on the corresponding wave functions of the Hamiltonian $\hat{H}_0$ and to equate the determinant of the matrix obtained to zero.

According to group theory [25], the spherical functions are transformed as partners in the bases of irreducible representations of the cube group O. In particular, the functions $Y_{1,-1}, Y_{1,0}, -Y_{1,1}$ form the basis of the representation F_1; the representation E is attributed to the basis: $\frac{1}{\sqrt{2}}(Y_{2,2} + Y_{2,-2})$, $Y_{2,0}$, and the functions $\frac{1}{\sqrt{2}}(Y_{2,2} - Y_{2,-2})$, $Y_{2,1}$, $Y_{2,-1}$ form the basis of the representation F_2.

The calculation of matrix elements made for the heterostructures GaAs/AlAs and InAs/GaSb has shown that, according to group theory, only the diagonal matrix elements differ from zero on the considered functions. This result shows that the numerical calculations of integrals are performed with a necessary degree of accuracy.

In case $l = 2$, the matrix 5×5, as one would expect, is also diagonal and contains two elements of one magnitude (E-state) and three elements of other magnitude (F_2-state). Thus, we obtain two energy correction values, and the difference between them defines the splitting of d-states in a cubic QD.

Figure 1 illustrates the results of calculations of the dependence $\Delta E'_l$ on the volume of a QD for both heterostructures and for the states with l = 0,1,2. The calculations show that the corrections are small in comparison with the energy level values and the difference between energy levels of the initial ("zero") problem. In particular, for the heterostructures InAs/GaSb, we have ${E^0}_{n_r=0,\,l=0} = -0.5$ eV, ${E^0}_{n_r=0,\,l=1} = -0.176$ eV, $\Delta E_{l=1} = 0.04$ eV, $\Delta E_{l=0} = 0.035$ eV if $V = 600$ nm^3; ${E^0}_{n_r=0,\,l=0} = -0.66$ eV, ${E^0}_{n_r=0,\,l=1} = -0.49$ eV, $\Delta E_{l=1} = 0.05$ eV, $\Delta E_{l=0} = 0.033$ eV if $V = 2050$ nm^3. For the heterostructures GaAs/AlAs, we obtain ${E^0}_{n_r=0,\,l=0} = -0.188$ eV, ${E^0}_{n_r=0,\,l=1} = -0.083$ eV, $\Delta E_{l=1} = 0.017$ eV, $\Delta E_{l=0} = 0.015$ eV if $V = 600$ nm^3; ${E^0}_{n_r=0,\,l=0} = -0.23$ eV, ${E^0}_{n_r=0,\,l=1} = -0.178$ eV, $\Delta E_{l=1} = 0.022$ eV, $\Delta E_{l=0} = 0.014$ eV if $V = 2050$ nm^3. It is seen that, in the case of large sizes of a QD, the corrections to the energy of both s- and p-states are small. A reduction in the volume leads to an increase in ΔE_l ($l = 0, 1$), which is caused by increase in W. The value of ΔE_l decreases slightly with the further reduction of the volume because the matrix element given by corrections decreases due to the electron tunneling. The energies ${\Delta E_{l=3}}^{(1)}$, ${\Delta E_{l=3}}^{(2)}$ not only essentially differ in value (at $V = 7 \times 10^3$ nm^3 of a QD of the heterosystem InAs/GaSb ${\Delta E_{l=3}}^{(2)} - {\Delta E_{l=3}}^{(1)} = 110$ meV), but change differently with change in the volume. The reduction of V leads to an increase of the splitting of the d-level, but, at $V < V_0$ (V_0 for the present heterostructures is different), the splitting decreases with V. If we compare the values of the splitting of the d-level for two different types of QDs, it is seen that the splitting is smaller in the GaAs/AlAs system than that in InAs/GaSb. This effect, as well as the correction values for s- and p-states, is explained by the difference of electron masses in QDs of the respective nanoheterosystems. For the InAs/GaSb heterostructure, the mass of a particle inside a QD is smaller than that in the GaAs/AlAs structure.

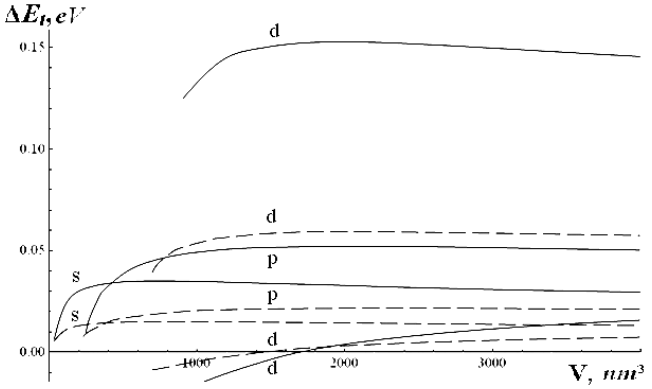

Fig. 1. Dependence $\Delta E_l = \Delta E_l(V)$ for a QD with cubic shape. Solid curves stand for the InAs/GaSb heterosystem, and dashed curves correspond to GaAs/AlAs

Analogous calculations were done for a QD with surface in the form of a tetrahedron. For the states with l = 0,1,2, the first correction to the energy ΔE_l is calculated for the heterostructures InAs/GaSb and GaAs/AlAs. The states with l = 1,2 are degenerate like

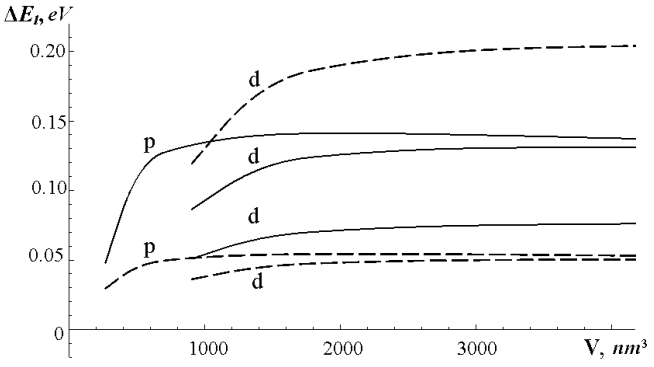

Fig. 2. Dependence $\Delta E_l = \Delta E_l(V))$ for the GaAs/AlAs heterosystem (solid curves) and the InAs/GaSb heterosystem (dashed curves) for a QD with pyramidal shape

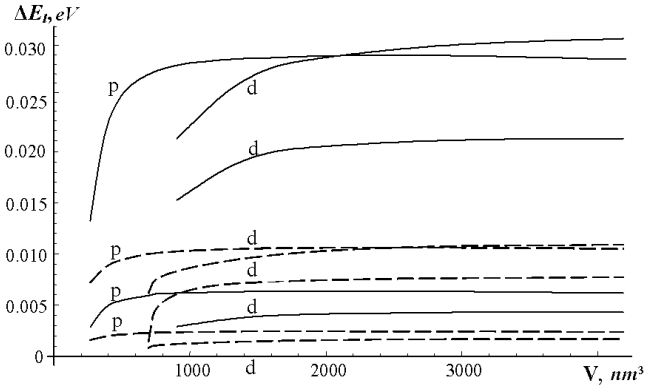

Fig. 3. Dependence $\Delta E_l = \Delta E_l(V))$ for the GaAs/AlAs heterosystem (solid curves) and the InAs/GaSb heterosystem (dashed curves) for a QD with hexagonal form

the case of a cubic QD. Therefore, in order to calculate the correction to the energy, the matrix elements of $(W - \Delta E_l')$ have been found on the corresponding wave functions of the Hamiltonian $\hat{H}_0$.

Figure 2 presents the dependence of the first correction to the energy on the volume of a QD with the surface in the form of a regular triangular pyramid. The qualitative dependences are analogs to the corresponding data on a QD with cubic shape. The obtained splitting of energy levels completely agrees with the results obtained within group theory. Thus, a change of the QD shape in this case leads only to a change of the value of respective corrections to the energy ΔE_l.

A slightly different picture is got for a QD with the surface in the form of a regular six-facet prism. For the present quantum dot, the splitting of energy levels begins with the state $l = 1$, which is split into two energy levels (E_1 and F-state). In both heterosystems, the level with $l = 2$ is split into three levels (A-state and two F-states), which perfectly agrees with group theory.

The results of calculations of the dependence of the correction to the energy $\Delta E_l'$ on the volume of a QD under consideration are given in Fig. 3. Since, in this case, there are a lot of energy levels and they are located close to one another, the state with $l = 0$ for both heterostructures is not presented. The figure shows that, for a hexagonal quantum dot, the correction value also increases with decrease in the QD volume. It is worth to note that, for the heterostructure InAs/GaSb, the corrections for the states with $l = 1$ and $l = 2$ at large QD volumes are close. At $V = 2.1 \times 10^3$ nm^3, the values of $\Delta E_l'$ for these states practically coincide.

The higher the energy state, the larger the energy correction for each heterostructure, the splitting of both p- and d-levels in the GaAs/AlAs heterosystem being smaller than that in InAs/GaSb, which was discussed above.

Thus, we have investigated the influence of the symmetry of a QD shape on the splitting of energy states of charges, by using group theory. In particular, the splitting of states for cubic, hexagonal and tetrahedral QDs is analyzed in the cases where a particle spin is zero or different from zero.

By perturbation theory, the energies and the wave functions of states for QDs with surfaces in the form of a cube, pyramid, and six-facet prism are determined. By using perturbation theory and symmetry analysis, the charge energy corrections have been calculated for the s-, p-, and d-states. The specific calculations have been performed for the GaAs/AlAs and InAs/GaSb heterostructures.

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ЕНЕРГЕТИЧНИЙ СПЕКТР ЗАРЯДУ КВАНТОВИХ ТОЧОК РІЗНОЇ ФОРМИ

В.І. Бойчук, І.В. Білинський, І.О. Шаклеіна

Р е з ю м е

Засобами теорії груп детально проаналізовано розщеплення енергетичних рівнів заряду (електрона, дірки) для квантової точки (КТ) кубічної, гексагональної та тетраедральної форм. Проведено аналіз розщеплення рівнів для випадків, коли спін частинки дорівнює нулю та 1/2. За допомогою теорії збурень знайдено енергії та хвильові функції станів КТ різної форми. За основу взято квантову точку сферичної симетрії, для якої енергія заряду знаходиться точно. Конкретні обчислення проведено для гетероструктур GaAs/AlAs та InAs/GaSb.