

Hole, impurity and exciton states in a spherical quantum dot

V.I. Boichuk, I.V. Bilynskyi, R.Ya. Leshko*

Department of Theoretical Physics, Ivan Franko Drohobych State Pedagogical University, 3 Stryiska Str., 82100, Drohobych, Lviv region, Ukraine

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The $3 \times 3 \vec{k}\vec{p}$ hole Hamiltonian for the wave-function envelopes (effective mass Hamiltonian) was used for calculation of discrete states of the hole and acceptor hydrogenic impurity in a spherical Si/SiO₂ nanoheterostructure as a function of the quantum dot radius by neglecting the corrugation of constant-energy surfaces. A study was conducted in the case of finite potential well at the separation boundary of the nanoheterosystem. The dependence of the hole energy spectrum on polarization charges, which arise at the separation boundary of the media, and on the dielectric permittivity, was defined. Using the exact electron and hole solutions, the exciton wave-function was constructed and the exciton ground-state energy was defined. The theoretical results have been compared with experimental data.

Key words: *impurity, multiband model, polarization charges*

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1. Introduction

Recently a study of properties of electrons and holes has been given a lot of attention in quantum dots (QD's). Since the conductive band of most semiconductors considered in this study can be described by the parabolic dispersion relation, the effective masses were introduced and the Schrödinger equation has been obtained. The use of such an approximation and the dielectric continuum model yields good theoretical results that conform to experimental data. The presence of impurities in QD's can significantly change the localization states. First theoretical researches of impurity donor states in QD's were reported in [1–6], where exact solutions of the Schrödinger equation with the Coulomb potential interaction between particles were obtained. It is shown in work [6] that the exact solutions of the Poisson and Schrödinger equations for hydrogenic donor impurity being taken into account changes to some extent the electron spectrum as compared with results [1–5].

The valence band in many semiconductors is degenerate. Work [7] is one of the first works, where the general spherically symmetrical solutions of the hole with a total angular momentum f were obtained in a multiband model. The study of shallow acceptors in bulk semiconductors was conducted in [8]. Using [9, 10], the hole Hamiltonian was derived there in the spherical approximation, and acceptor states were calculated in the cases of strong and weak spin-orbit interaction.

Electron-hole pairs were theoretically studied in the multiband effective-mass approximation with the infinite potential well at the heteroboundary [11]. Single-particle states in spherical Si/SiO₂ QD's were studied in [12], where the finiteness of crystals band discontinuity and exact boundary conditions were already taken into account.

The presence of donor and acceptor impurities in QD's can significantly change the localized states. Although the calculations of acceptor states were performed, for example [8, 13] – in bulk crystals, [14] – in thin films, today the acceptor states and their effect on the QD's properties have not been studied well enough. On this account, the aim of the present work is to study the effect of QD size, matrix dielectric permittivity and polarization charges on the energy spectrum

*E-mail: leshkoroman@meta.ua

of the hole, acceptor impurity and exciton. Specific calculations are performed for the Si/SiO₂ nanoheterostructure. Theoretical results have been compared with experimental data.

2. Formulation of the problem and its solution

2.1. Hole Hamiltonian 3x3 and wave functions

We consider a spherical QD heterostructure. The radius of the QD is a . There is an acceptor impurity in the center of the QD. Let the heterosystem be constructed of crystals with a large band gap E_g and weak spin-orbit interaction Δ [8]. Therefore, the conduction band can be neglected and one can assume, that $\Delta = 0$. In order to simplify formulas, the system of units ($m_0 = 1$, $\hbar = 1$, $e = 1$) is used, and the energy is counted downward. Taking into account these remarks and neglecting the corrugation of constant-energy surfaces (the so-called spherical approximation), we write the kp -Hamiltonian for envelope functions in the form of [15]

$$\mathbf{H} = - \left[(A + 2B) \mathbf{p}^2 - 3B \left(\tilde{\mathbf{p}} \tilde{\mathbf{J}} \right)^2 \right] + \Pi(r), \quad (1)$$

where

$$A = -\frac{\gamma_1}{2}, \quad B = -\gamma, \quad \mu = \frac{2\gamma}{\gamma_1}, \quad \gamma = \frac{1}{5} (3\gamma_3 + 2\gamma_2),$$

$\gamma_1, \gamma_2, \gamma_3$ are the Luttinger parameters, which for different regions of the heterostructure are denoted as follows:

$$(\gamma_1 \quad \gamma_2 \quad \gamma_3) = \begin{cases} (\gamma_1^{\text{in}} & \gamma_2^{\text{in}} & \gamma_3^{\text{in}}), & r \leq a, \\ (\gamma_1^{\text{out}} & \gamma_2^{\text{out}} & \gamma_3^{\text{out}}), & r > a. \end{cases}$$

Further all notations with the index *in* will correspond to a QD, and those with the index *out* – to the matrix. The operator $\tilde{\mathbf{J}}$ of the angular momentum corresponds to unity spin. The hole potential energy is given by

$$\Pi(r) = U(r) + V_c(r) + V_p(r), \quad (2)$$

where

$$U(r) = \begin{cases} 0, & r \leq a, \\ U_0 & r > a \end{cases} \quad (3)$$

is the potential energy caused by the band offset. Using the exact solution of the Poisson equation, the potential energy of the hole-acceptor ion coupling is given by

$$V_c(r) = - \begin{cases} 1/(\varepsilon^{\text{in}} r) + (\varepsilon^{\text{in}} - \varepsilon^{\text{out}}) / (\varepsilon^{\text{in}} \varepsilon^{\text{out}} a), & r \leq a, \\ 1/(\varepsilon^{\text{out}} r), & r > a. \end{cases} \quad (4)$$

A charged particle will induce polarization charges at the separation boundary of the media. The corresponding energy of the electron-polarization charges interaction will be determined like in [16]:

$$\begin{aligned} V_p(r) &= \frac{\omega}{4\varepsilon(r)} \int_0^\infty dt \left[\text{th} \left(\frac{t-a}{r_0} \right) + \frac{t}{r_0} \text{sech}^2 \left(\frac{t-a}{r_0} \right) \right] \frac{1}{t^2 - r^2}, \\ \varepsilon(r) &= \frac{\varepsilon^{\text{in}} + \varepsilon^{\text{out}}}{2} \left[1 - \omega \cdot \text{th} \left(\frac{r-a}{r_0} \right) \right], \quad \omega = \frac{\varepsilon^{\text{in}} - \varepsilon^{\text{out}}}{\varepsilon^{\text{in}} + \varepsilon^{\text{out}}}, \end{aligned} \quad (5)$$

where r_0 is the thickness of the transition layer at the interface where the dielectric permittivity depends on coordinates.

The wave function, which is the eigenfunction of the Hamiltonian (1), can be written as a product of the radial function and the eigenfunction of the square of the total angular momentum $\mathbf{F}^2 = (\tilde{\mathbf{J}} + \tilde{\mathbf{L}})^2$. It is

$$\Phi_{f,M}^l(\theta, \varphi) = \sum_{m=-l}^l \sum_{m_j=-1}^1 C_{l,m;1,m_j}^{f,M} Y_{l,m}(\theta, \varphi) \chi_{m_j}, \quad (6)$$

where $f(f+1)$, $l(l+1)$, M , m , m_j are the quantum numbers, which determine the eigenvalues of the operators \mathbf{F}^2 , \mathbf{L}^2 , \mathbf{F}_z , \mathbf{L}_z , \mathbf{J}_z respectively. $C_{l,m;1,m_j}^{f,M}$ are the Clebsch-Gordan coefficients, χ_{m_j} are the spin functions, $Y_{l,m}$ are the spherical harmonics, being the eigenfunctions of \mathbf{L}^2 . According to the common rules of addition of the angular momenta, we write the most general expression of solutions of the Schrödinger equation with the Hamiltonian (1) for three types of states [8, 12]:

$$\psi_{f,M}^{f-1,f+1}(\vec{r}) = R_f^{f-1}(r) \Phi_{f,M}^{f-1}(\theta, \varphi) + R_f^{f+1}(r) \Phi_{f,M}^{f+1}(\theta, \varphi), \quad (f \geq 1; l = f-1, f+1), \quad (7)$$

$$\psi_{f,M}^f(\vec{r}) = R_f^f(r) \Phi_{f,M}^f(\theta, \varphi), \quad (f \geq 1; \quad l = f), \quad (8)$$

$$\psi_{0,0}^1(\vec{r}) = R_0^1(r) \Phi_{0,0}^1(\theta, \varphi), \quad (f = 0; \quad l = 1). \quad (9)$$

Inserting functions (7)–(9) into the Schrödinger equation with the Hamiltonian (1), which is then multiplied by the corresponding conjugated spinors $(\Phi_{f,M}^l(\theta, \varphi))^+$, we get the following equations for the radial functions of the above-mentioned states:

$$A \begin{pmatrix} C_1 \mathbf{B}_{f-2}^+ \mathbf{B}_{f-1}^- & C_2 \mathbf{B}_f^- \mathbf{B}_{f+1}^- \\ C_2 \mathbf{B}_f^+ \mathbf{B}_{f-1}^+ & C_3 \mathbf{B}_{f+2}^- \mathbf{B}_{f+1}^+ \end{pmatrix} \begin{pmatrix} R_f^{f-1} \\ R_f^{f+1} \end{pmatrix} + \Pi(r) \begin{pmatrix} R_f^{f-1} \\ R_f^{f+1} \end{pmatrix} - E \begin{pmatrix} R_f^{f-1} \\ R_f^{f+1} \end{pmatrix} = 0, \quad (10)$$

$$A(1-\mu) (\Delta_f R_f^f) + \Pi(r) R_f^f - E R_f^f = 0, \quad (11)$$

$$A(1+2\mu) (\Delta_1 R_0^1) + \Pi(r) R_0^1 - E R_0^1 = 0. \quad (12)$$

In equations (10)–(12) the following notations are introduced:

$$\Delta_l = \frac{\partial^2}{\partial r^2} + \frac{2}{r} \frac{\partial}{\partial r} - \frac{l(l+1)}{r^2}, \quad \mathbf{B}_l^+ = \frac{d}{dr} - \frac{l}{r}, \quad \mathbf{B}_l^- = \frac{d}{dr} + \frac{(l+1)}{r},$$

$$C_1 = 1 + \frac{f-1}{2f+1} \mu, \quad C_2 = -\frac{3\sqrt{f(f+1)}}{2f+1} \mu, \quad C_3 = 1 + \frac{f+2}{2f+1} \mu.$$

From the equations for the radial functions one can find the hole energies and wave functions.

2.2. Boundary conditions

In order to get the boundary conditions for the spherical QD, two conditions are used. They are as follows: the continuity of the radial wave-function and the continuity of the normal component of the probability density flux at the boundary of the QD. The continuity of wave function gives

$$\left(\begin{pmatrix} (R_f^{f-1})^{\text{in}} \\ (R_f^{f+1})^{\text{in}} \end{pmatrix} \right) \Big|_{r=a} = \left(\begin{pmatrix} (R_f^{f-1})^{\text{out}} \\ (R_f^{f+1})^{\text{out}} \end{pmatrix} \right) \Big|_{r=a}, \quad (13)$$

$$(R_f^f)^{\text{in}} \Big|_{r=a} = (R_f^f)^{\text{out}} \Big|_{r=a}, \quad (R_0^1)^{\text{in}} \Big|_{r=a} = (R_0^1)^{\text{out}} \Big|_{r=a}.$$

In order to determine the normal component of the probability density flux, the normal component of the velocity operator $\mathbf{V}_r = \vec{r}/r (\partial \mathbf{H} / \partial \vec{\mathbf{p}})$, which is proportional to the normal component of the probability density flux, is defined. Then, the operator was written in the spinor representation for three types of states (7)–(9). Using the explicit form of the operators, three more conditions are found (see Appendix)

$$\begin{pmatrix} \mathbf{T}_{11}^{\text{in}} & \mathbf{T}_{12}^{\text{in}} \\ \mathbf{T}_{21}^{\text{in}} & \mathbf{T}_{22}^{\text{in}} \end{pmatrix} \begin{pmatrix} (R_f^{f-1})^{\text{in}} \\ (R_f^{f+1})^{\text{in}} \end{pmatrix} \Big|_{r=a} = \begin{pmatrix} \mathbf{T}_{11}^{\text{out}} & \mathbf{T}_{12}^{\text{out}} \\ \mathbf{T}_{21}^{\text{out}} & \mathbf{T}_{22}^{\text{out}} \end{pmatrix} \begin{pmatrix} (R_f^{f-1})^{\text{out}} \\ (R_f^{f+1})^{\text{out}} \end{pmatrix} \Big|_{r=a}, \quad (14)$$

$$A^{\text{in}} (1 - \mu^{\text{in}}) \frac{d}{dr} (R_f^f)^{\text{in}} \Big|_{r=a} = A^{\text{out}} (1 - \mu^{\text{out}}) \frac{d}{dr} (R_f^f)^{\text{out}} \Big|_{r=a}, \quad (15)$$

$$A^{\text{in}} (1 + 2\mu^{\text{in}}) \frac{d}{dr} (R_0^1)^{\text{in}} \Big|_{r=a} = A^{\text{out}} (1 + 2\mu^{\text{out}}) \frac{d}{dr} (R_0^1)^{\text{out}} \Big|_{r=a}. \quad (16)$$

We use the following notations in equations (14)–(16):

$$\begin{aligned} \mathbf{T}_{11}^{\{\text{in}\}_{\text{out}}\}} &= A^{\{\text{in}\}_{\text{out}}\}} \left[\left(1 + \frac{f-1}{2f+1} \mu^{\{\text{in}\}_{\text{out}}\}} \right) \frac{d}{dr} + \frac{3(f-1)}{2(2f+1)} \frac{\mu^{\{\text{in}\}_{\text{out}}\}}}{r} \right], \\ \mathbf{T}_{12}^{\{\text{in}\}_{\text{out}}\}} &= \frac{3\sqrt{f(f+1)}}{2f+1} B^{\{\text{in}\}_{\text{out}}\}} \left(\frac{d}{dr} + \frac{f+2}{r} \right), \\ \mathbf{T}_{21}^{\{\text{in}\}_{\text{out}}\}} &= \frac{3\sqrt{f(f+1)}}{2f+1} B^{\{\text{in}\}_{\text{out}}\}} \left(\frac{d}{dr} - \frac{f-1}{r} \right), \\ \mathbf{T}_{22}^{\{\text{in}\}_{\text{out}}\}} &= A^{\{\text{in}\}_{\text{out}}\}} \left[\left(1 + \frac{f+2}{2f+1} \mu^{\{\text{in}\}_{\text{out}}\}} \right) \frac{d}{dr} + \frac{3}{2} \frac{f+2}{2f+1} \frac{\mu^{\{\text{in}\}_{\text{out}}\}}}{r} \right]. \end{aligned}$$

The hole energy spectrum can be determined using the solution of equations (10)–(12), when the boundary conditions (13)–(16) are satisfied. If we denote the number of solution of the corresponding equation system through n ($n = 1, 2, \dots$), then apart from the above-mentioned quantum numbers, the energies and wave-functions will depend on one more quantum number – n . Therefore, functions (7)–(9) may be redenoted as $\psi_{f,M}^{f-1,f+1} \equiv \psi_{n,f,M}^{f-1,f+1}$; $\psi_{f,M}^f \equiv \psi_{n,f,M}^f$; $\psi_{0,0}^1 \equiv \psi_{n,0,0}^1$. It will be used to analyze figure 1 and figure 2.

2.3. Hole energy spectrum without impurity

We assume $\Pi(r) = U(r) + V_p(r)$. This is equivalent to the study of the hole states in a spherical QD without acceptor impurity. The potential energy $V_p(r)$ of the interaction of a hole with polarization charges at the interface can be considered as perturbation, the effect of which will be accounted for in first-order perturbation theory [16]. For the unperturbed system $\Pi(r) = U(r)$. In this case equations (10)–(12) have exact solutions given by the spherical Bessel functions of the first kind and the modified spherical Bessel functions of the second kind

$$\begin{aligned} (R_f^{f-1})^{\text{in}} &= D_1^{\text{in}} j_{f-1}(\lambda^{\text{in}} r/a) + D_2^{\text{in}} j_{f-1}(\lambda^{\text{in}} \beta^{\text{in}} r/a), \\ (R_f^{f+1})^{\text{in}} &= -D_1^{\text{in}} \sqrt{\frac{f}{f+1}} j_{f+1}(\lambda^{\text{in}} r/a) + D_2^{\text{in}} \sqrt{\frac{f+1}{f}} j_{f+1}(\lambda^{\text{in}} \beta^{\text{in}} r/a), \\ (R_f^f)^{\text{in}} &= D_{f,f}^{\text{in}} j_f(\lambda^{\text{in}} r/a), \\ (R_0^1)^{\text{in}} &= D_{1,0}^{\text{in}} j_1(\lambda^{\text{in}} \beta^{\text{in}} r/a), \quad r \leq a, \end{aligned} \quad (17)$$

$$\begin{aligned} (R_f^{f-1})^{\text{out}} &= D_1^{\text{out}} k_{f-1}(\lambda^{\text{out}} r/a) + D_2^{\text{out}} k_{f-1}(\lambda^{\text{out}} \beta^{\text{out}} r/a), \\ (R_f^{f+1})^{\text{out}} &= D_1^{\text{out}} \sqrt{\frac{f}{f+1}} k_{f+1}(\lambda^{\text{out}} r/a) - D_2^{\text{out}} \sqrt{\frac{f+1}{f}} k_{f+1}(\lambda^{\text{out}} \beta^{\text{out}} r/a), \\ (R_f^f)^{\text{out}} &= D_{f,f}^{\text{out}} k_f(\lambda^{\text{out}} r/a), \\ (R_0^1)^{\text{out}} &= D_{1,0}^{\text{out}} k_1(\lambda^{\text{out}} \beta^{\text{out}} r/a), \quad r > a, \end{aligned} \quad (18)$$

where

$$\lambda^{\text{in}} = \sqrt{\frac{-Ea^2}{A^{\text{in}}(1-\mu^{\text{in}})}}, \quad \lambda^{\text{out}} = \sqrt{\frac{-(-E+U_0)a^2}{A^{\text{out}}(1-\mu^{\text{out}})}}, \quad \beta^{\left\{\begin{smallmatrix} \text{in} \\ \text{out} \end{smallmatrix}\right\}} = \sqrt{\frac{1-\mu^{\left\{\begin{smallmatrix} \text{in} \\ \text{out} \end{smallmatrix}\right\}}}{1+2\mu^{\left\{\begin{smallmatrix} \text{in} \\ \text{out} \end{smallmatrix}\right\}}}}.$$

Applying the boundary conditions to solutions (17)–(18), we determine the eigenvalues. The energy, caused by the interaction of electron-polarization charges, is given by

$$E_p = \int d\vec{r} (\psi_{f,M}^l)^+ V_p(r) \psi_{f,M}^l. \quad (19)$$

2.4. Acceptor energy spectrum

The total potential energy of the hole in the spherical QD with an acceptor impurity has the form (2). If the term $V_p(r)$ is neglected in zero-order approximation, then equations (11)–(12) (second and third types of states) have exact solutions, and the effect of $V_p(r)$, like in the previous section, can be taken into account in first-order perturbation theory. Those two equations are written for two regions.

If $r \leq a$, then

$$-\frac{1}{2b^{\text{in}}} (\Delta_n R^{\text{in}}) - \frac{1}{\varepsilon^{\text{in}} r} R^{\text{in}} - \tilde{E}^{\text{in}} R^{\text{in}} = 0. \quad (20)$$

For the second type of states $b^{\text{in}} = 1/(\gamma_1^{\text{in}}(1-\mu^{\text{in}}))$, $\Delta_n = \Delta_f$, $R^{\text{in}} = (R_f^f)^{\text{in}}$, and for the third type of states $b^{\text{in}} = 1/(\gamma_1^{\text{in}}(1+2\mu^{\text{in}}))$, $\Delta_n = \Delta_1$, $R^{\text{in}} = (R_0^1)^{\text{in}}$. In both cases $\tilde{E}^{\text{in}} = E + U^*(a)$, $U^*(a) = (\varepsilon^{\text{in}} - \varepsilon^{\text{out}})/(\varepsilon^{\text{in}}\varepsilon^{\text{out}}a)$. Equation (20) is considered in two energy ranges: $\tilde{E}^{\text{in}} < 0$, $\tilde{E}^{\text{in}} > 0$.

If $\tilde{E}^{\text{in}} < 0$, we introduce the following notations: $\xi^{\text{in}} = \alpha^{\text{in}}r$, $(\alpha^{\text{in}})^2 = -8b^{\text{in}}\tilde{E}^{\text{in}}$, $\lambda^{\text{in}} = 2b^{\text{in}}/(\varepsilon^{\text{in}}\alpha^{\text{in}})$, $R^{\text{in}}(\xi^{\text{in}}) = g^{\text{in}}(\xi^{\text{in}})/\xi^{\text{in}}$. After performing some simple transformations, we obtain the Whittaker equation

$$\frac{\partial^2}{(\partial\xi^{\text{in}})^2} g^{\text{in}} + \left[-\frac{1}{4} + \frac{\lambda^{\text{in}}}{\xi^{\text{in}}} - \frac{n(n+1)}{(\xi^{\text{in}})^2} \right] g^{\text{in}} = 0. \quad (21)$$

The solution of equation (21) is represented by

$$g^{\text{in}}(\xi^{\text{in}}) = D^{\text{in}} e^{-\xi^{\text{in}}/2} (\xi^{\text{in}})^{n+1} M(n+1-\lambda^{\text{in}}, 2n+2, \xi^{\text{in}}), \quad (22)$$

where $M(a, b, x)$ is the confluent hypergeometric function of the first kind [17].

Let us consider the other energy range: $\tilde{E}^{\text{in}} > 0$. By defining dimensionless values $\xi^{\text{in}} = \beta^{\text{in}}r$, $(\beta^{\text{in}})^2 = 2b^{\text{in}}\tilde{E}^{\text{in}}$, $\delta^{\text{in}} = -b^{\text{in}}/(\varepsilon^{\text{in}}\beta^{\text{in}})$, $R^{\text{in}}(\xi^{\text{in}}) = g^{\text{in}}(\xi^{\text{in}})/\xi^{\text{in}}$ equation (20) is transformed into the Coulomb equation

$$\frac{\partial^2}{(\partial\xi^{\text{in}})^2} g^{\text{in}} + \left[1 - \frac{2\delta^{\text{in}}}{\xi^{\text{in}}} - \frac{n(n+1)}{(\xi^{\text{in}})^2} \right] g^{\text{in}} = 0. \quad (23)$$

The solution of this equation, which satisfies the finiteness condition of the wave function, can be represented by the Coulomb function

$$g^{\text{in}}(\xi^{\text{in}}) = D^{\text{in}} \frac{2^n e^{-\pi\delta^{\text{in}}/2} |\Gamma(n+1+i\delta^{\text{in}})|}{\Gamma(2n+2)} e^{-i\xi^{\text{in}}} (\xi^{\text{in}})^{n+1} M(n+1-i\delta^{\text{in}}, 2n+2, 2i\xi^{\text{in}}), \quad (24)$$

where $\Gamma(z)$ is the Euler gamma-function [17].

When $r > a$, equations (11)–(12) can be written

$$-\frac{1}{2b^{\text{out}}} (\Delta_n R^{\text{out}}) - \frac{1}{\varepsilon^{\text{out}} r} R^{\text{out}} - \tilde{E}^{\text{out}} R^{\text{out}} = 0.$$

For the second type of states $b^{\text{out}} = 1/(\gamma_1^{\text{out}}(1 - \mu^{\text{out}}))$, $\Delta_n = \Delta_f$, $R^{\text{out}} = (R_f^f)^{\text{out}}$, and for the third type of states $b^{\text{out}} = 1/(\gamma_1^{\text{out}}(1 + 2\mu^{\text{out}}))$, $b^{\text{out}} = 1/(\gamma_1^{\text{out}}(1 + 2\mu^{\text{out}}))$, $R^{\text{out}} = (R_0^1)^{\text{out}}$. In both cases $\tilde{E}^{\text{out}} = E - U_0$. After introducing dimensionless variables and other parameters $\xi^{\text{out}} = \alpha^{\text{out}} r$, $(\alpha^{\text{out}})^2 = -8b^{\text{out}}\tilde{E}^{\text{out}}$, $\lambda^{\text{out}} = 2b^{\text{out}}/(\varepsilon^{\text{out}}\alpha^{\text{out}})$, $R^{\text{out}}(\xi^{\text{out}}) = g^{\text{out}}(\xi^{\text{out}})/\xi^{\text{out}}$, we obtain the Whittaker equation. The solution of this equation, which satisfies the finiteness condition of the wave function, in the limit $r \rightarrow \infty$, takes the form [17]:

$$g^{\text{out}}(\xi^{\text{out}}) = D^{\text{out}} \frac{\exp(-\xi^{\text{out}}/2) (\xi^{\text{out}})^{-n}}{\Gamma(-n - \lambda^{\text{out}})} \int_0^{\infty} dt \exp(-\xi^{\text{out}} t) t^{-n - \lambda^{\text{out}} - 1} (1+t)^{-n + \lambda^{\text{out}} - 1}. \quad (25)$$

From the boundary conditions and the normalization condition one can define the eigenvalues and eigenfunctions of an acceptor impurity. The potential energy $V_p(r)$ will be taken into consideration in first-order perturbation theory.

Equation (10) for an acceptor impurity has no exact solution, even if we neglect the $V_p(r)$ term. Therefore, we will find its solution expanding the exact radial wave function over the functions of the problem without an impurity (17)–(18):

$$\left(\begin{array}{c} (R_f^{f-1})_a \\ (R_f^{f+1})_a \end{array} \right) = \sum_i c_i \left(\begin{array}{c} (R_f^{f-1})_i \\ (R_f^{f+1})_i \end{array} \right). \quad (26)$$

Inserting (26) into (10) and multiplying it by the conjugate row-vector $\left(\begin{array}{cc} (R_f^{f-1})_{i'}^* & (R_f^{f+1})_{i'}^* \end{array} \right)$ gives the homogeneous system of equations with coefficients c_i :

$$\sum_i ((E_i - E_a) \delta_{i'i} + V_{i'i}) c_i = 0, \quad (27)$$

where E_i is the specific h hole energy level without an impurity, which is determined by solutions (17)–(18), and

$$V_{i'i} = \int dr r^2 (V_c(r) + V_p(r)) \left[(R_f^{f-1})_{i'}^* (R_f^{f-1})_i + (R_f^{f+1})_{i'}^* (R_f^{f+1})_i \right].$$

Equating the determinant of the homogeneous system (27) to zero, we get the equation to determine the hole energy E_a with the acceptor presence. Using system (27) and the normalization condition $\sum_i |c_i|^2 = 1$, one can define the eigenfunctions of an acceptor impurity.

2.5. Exciton ground-state energy

The ground-state energy of the electron-hole pair without an impurity can be calculated by perturbation theory. The expression of the electron-hole coupling is given by [18]. The exciton wave function was constructed as a product of the electron and hole wave functions

$$\Psi_{\text{ex}} = \psi_{1,0,0}(\vec{r}_1) \psi_{1,1,M}^{0,2}(\vec{r}_2), \quad (28)$$

where \vec{r}_1 , \vec{r}_2 are the electron and hole coordinates, respectively. The solutions of the electron Schrödinger equation in the spherical QD, neglecting the effective mass anisotropy in the QD, were defined similar to [18, 19]. The polarization charges at the separation boundary of the media were taken into the consideration therein.

Therefore, adding the electron and hole energies and taking into account the electron-hole interaction, the exciton energy was obtained.

3. The analysis of results

All results have been obtained for the spherical Si/SiO₂ QD heterostructure. We take the basic parameters of crystals, which form the heterostructure, to be identical to those in [12]: $\gamma_1^{(\text{in})} = 4.22$, $\gamma_2^{(\text{in})} = 0.53$, $\gamma_3^{(\text{in})} = 1.38$, $\varepsilon^{(\text{in})} = 11.4$, $\gamma_1^{(\text{out})} = 1/5$, $\gamma^{(\text{out})} = 0$, $\varepsilon^{(\text{out})} = 3.9$, $U_0 = 4.3$ eV. The introduced isotropic effective electron mass in Si is considered equal to the mean value of $m_{\perp}^* = 0.19$ and $m_{\parallel}^* = 0.916$ effective electron masses.

Figure 1 shows the dependences of the ground and lower hole energy levels on the QD size for three types of states (7)–(9). Also, the effect of polarization charges at the interface was taken into account. It is seen from figure 1 that with the decrease the QD radius, the hole energy increase is caused by the enhancement of the effect of the spatial confinement and polarization charges at the interface.

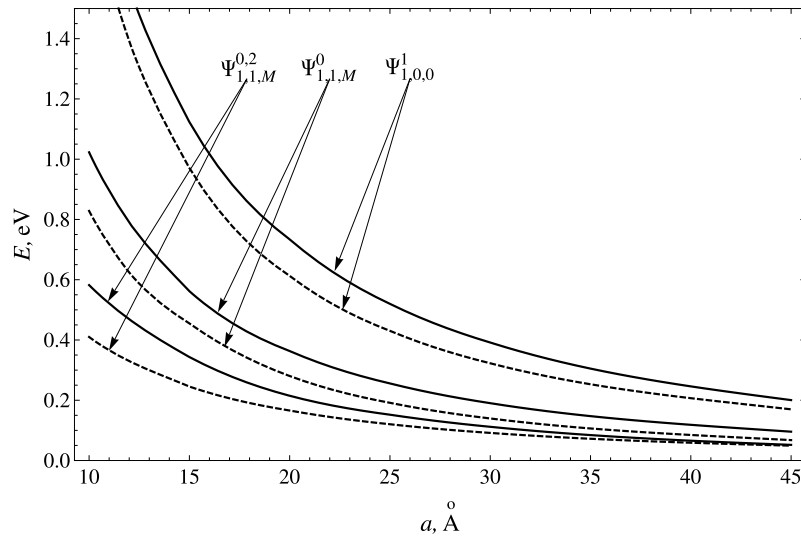


Figure 1. Dependence of the hole energy on the QD radius without (dashed curves) and with (solid curves) taking into account polarization charges.

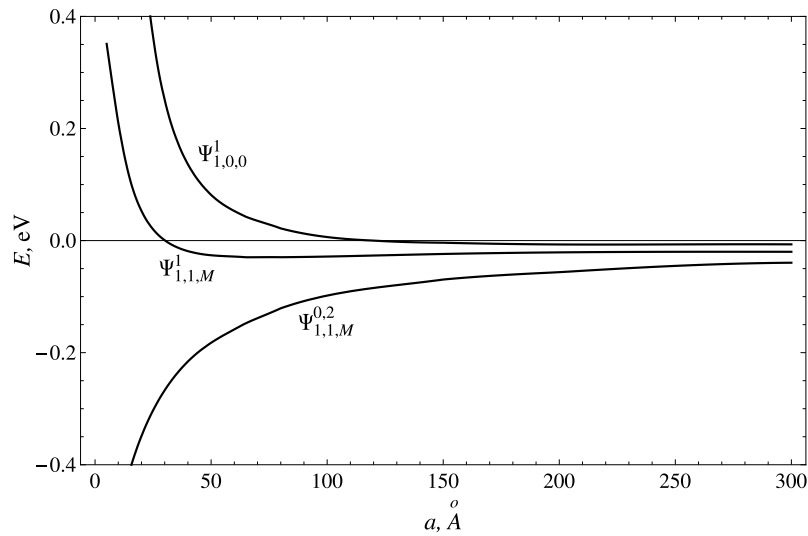


Figure 2. The acceptor hole energy spectrum.

The acceptor impurity energy spectrum, with taking into account the total potential energy (2), is represented in figure 2. Therein and further in the paper, the polarization term $V_p(r)$ is taken into consideration. The energy levels smoothly approach the values, which correspond to the acceptor impurity hole levels in the bulk silicon crystal in the limit of a large QD radius. The reduction of the QD radius causes two competing effects: the rise of the spatial confinement, which increases the hole energy, and the rise of the quantum well depth $U^*(a)$ (4), which in this case decreases the hole energy. As is shown in [6], the effect of the effective potential well on the particle energy is clear for the ground and first lower states. Therefore, the decrease of the QD radius for $\psi_{1,1,M}^{0,2}$ (7) leads to the reduction of the particle energy. Moreover, the reason of such energy behavior is the large hole effective mass in the matrix, which is taken $m_v = 5$, like in [12]. As for the states $\psi_{1,1,M}^1$, $\psi_{1,0,0}^1$ (8)–(9), the hole energy decreases slightly first (which is unnoticeable in figure 2) and then rises.

In order to study the dependence of the acceptor energy levels on the matrix dielectric permittivity and to clearly show the effect of the effective potential well on the hole spectrum, the calculation of the acceptor ground-state energy is performed for different ϵ^{out} . The result of the calculation is shown in figure 3.

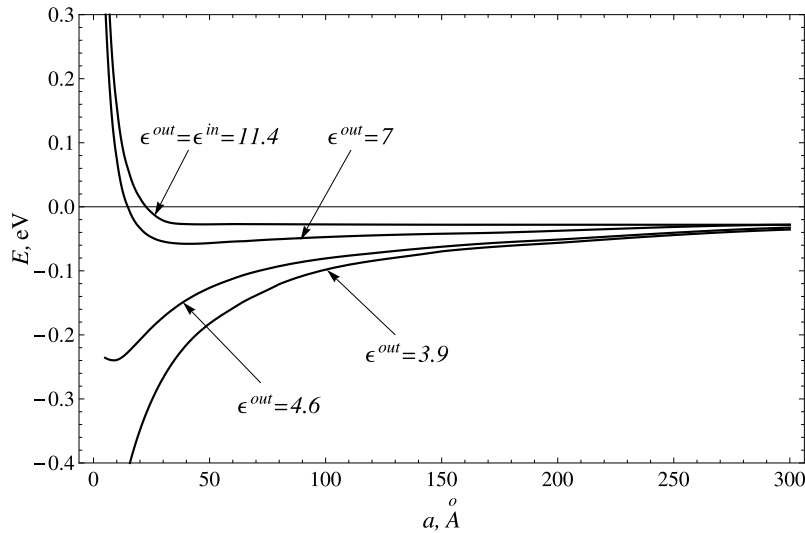


Figure 3. The acceptor ground-state energy for different values of the matrix dielectric permittivity.

If $\epsilon^{\text{out}} = 3.9$, like in [12], then the effective potential well in the QD is negative and in its absolute value is larger than the kinetic energy of the particle, and thus the total energy decreases. In case the dielectric permittivity of the matrix $\epsilon^{\text{out}} = 4.6$ [20], then contributes less to the total energy, but when $a \leq 5$ Å, the total energy begins to increase. If the matrix dielectric permittivity $\epsilon^{\text{out}} = 7$, then the particle energy is larger than that in the previous case. When $a \approx 25$ Å, the minimum is observed in figure 3. When the dielectric permittivity of the matrix equals the QD dielectric permittivity $\epsilon^{\text{out}} = \epsilon^{\text{in}} = 11.4$, then $U^*(a) = 0$ and all polarization corrections equal zero. In that case we get a monotonous increase of the particle energy, if the QD radius decreases. However, in all the cases for large QD radii the acceptor ground-state energy leads to the corresponding energy in the bulk silicon crystal: $E = -0.031$ eV.

After having analysed the hole spectrum, one can consider the electron-hole bound state in the QD. Figure 4 shows the dependences of the ground-state exciton energy as a function of the QD radius. The experimental data obtained in [21–25], are presented too. As is seen from figure 4, our calculation results are both in good qualitative and quantitative agreement with the experimental data. Taking account of the multiband effective-mass approximation, interparticle interaction and interaction of particles with polarization charges allows us to determine the dependence of the exciton energy (denoted in figure 4 with “1”).

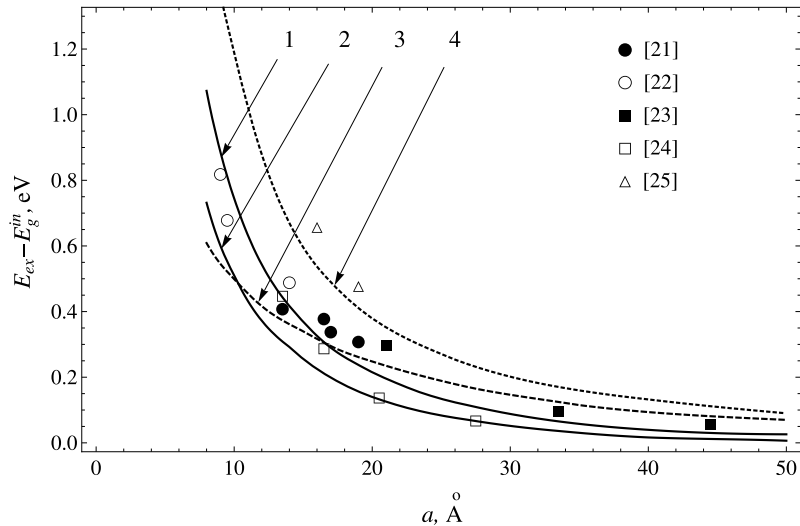


Figure 4. The exciton ground-state energy within (solid curves 1, 2) and without (dashed curve 3) the multiband effective-mass approximation. The ground-state electron-hole energy without the electron-hole interaction (dotted curve 4).

If the dielectric permittivity of the matrix equals that of the QD, then the exciton energy will be described by curve 2. It is found that the increase of the QD radius leads to the decrease of the effect of the matrix properties (in particular, the dielectric properties). Therefore, curves 1 and 2 begin to approach each other if $a > 40$ Å. If the complex band spectrum is neglected and the heavy hole effective mass is employed in calculations, similar to [26], then the exciton ground-state energy will be described by a dashed curve 3. In addition, figure 4 shows the dependence of the electron-hole ground-state energy without taking into account the electron-hole interaction (dotted curve 4). Therefore, the exciton energy depends on the dimensional quantization of the electron and hole energies in the QD and dielectric properties of the matrix. Besides, the calculations show that taking account of the complex band structure numerically changes the results as compared with the single valence-band model.

4. Summary

In the present work the hole and acceptor energy spectra in the QD have been studied. All the energies in this paper are calculated, when the polarization charges are taken into account. The calculation has been done in the multiband effective-mass approximation in case of the weak spin-orbit interaction. Within the frames of this model and in the finite spherical potential-well approximation, the exact hole solutions of all the radial equations have been obtained for the heterostructure. As for an acceptor impurity, the exact solutions have been derived for two types of states, which are described by a single differential equation. The matrix radial equation of the acceptor impurity has been solved approximately, using the exact hole solutions without an impurity. On the basis of exact electron and hole solutions, the exciton wave function has been written, and the exciton ground-state energy has been defined. It is found that the difference between the QD and matrix dielectric permittivity leads to an increase of the exciton energy. This is in good agreement with the data of other theoretical and experimental works. It is shown that the complex band structure being taking into account changes the exciton energy as compared with the single valence-band model. The comparison of the obtained results with experimental data has shown good numerical agreement.

Appendix

For example, we briefly show the scheme of obtaining the boundary condition (14). In order to find the normal component of the velocity operator

$$\mathbf{V}_n = \frac{\vec{r}}{r} \frac{\partial \mathbf{H}}{\partial \vec{\mathbf{p}}} = \left(n_+ \frac{\partial \mathbf{H}}{\partial \mathbf{p}_+} + n_- \frac{\partial \mathbf{H}}{\partial \mathbf{p}_-} + n_z \frac{\partial \mathbf{H}}{\partial \mathbf{p}_z} \right),$$

where $n_+ = (x + iy)/r = -\sqrt{2}\sqrt{4\pi/3}Y_{1,1}$, $n_- = (x - iy)/r = \sqrt{2}\sqrt{4\pi/3}Y_{1,-1}$, $n_z = z/r = \sqrt{4\pi/3}Y_{1,0}$, we write (1) in the matrix form, using the following operators $\mathbf{p}_x = (\mathbf{p}_+ + \mathbf{p}_-)/2$, $\mathbf{p}_y = (\mathbf{p}_+ - \mathbf{p}_-)/(2i)$, $\mathbf{p}^2 = \mathbf{p}_z^2 + \mathbf{p}_+\mathbf{p}_-$, where $[\mathbf{p}_+, \mathbf{p}_-] = 0$ property is taken into account, and the explicit form of \mathbf{J}_i is used

$$\mathbf{J}_x = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \quad \mathbf{J}_y = \frac{i}{\sqrt{2}} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & -1 \\ 0 & 1 & 0 \end{pmatrix}, \quad \mathbf{J}_z = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}.$$

It is found

$$\mathbf{V}_n = \begin{pmatrix} (\mathbf{V}_n)_{11} & (\mathbf{V}_n)_{12} & (\mathbf{V}_n)_{13} \\ (\mathbf{V}_n)_{21} & (\mathbf{V}_n)_{22} & (\mathbf{V}_n)_{23} \\ (\mathbf{V}_n)_{31} & (\mathbf{V}_n)_{32} & (\mathbf{V}_n)_{33} \end{pmatrix}, \quad (A1)$$

where

$$\begin{aligned} (\mathbf{V}_n)_{11} &= (\mathbf{V}_n)_{33} = -\left(A - \frac{B}{2}\right)(n_+\mathbf{p}_- + n_-\mathbf{p}_+) - 2(A - B)n_z\mathbf{p}_z, \\ (\mathbf{V}_n)_{12} &= -[(\mathbf{V}_n)_{21}]^* = -(\mathbf{V}_n)_{23} = [(\mathbf{V}_n)_{32}]^* = \frac{3B}{\sqrt{2}}(n_z\mathbf{p}_- + n_m\mathbf{p}_z), \\ (\mathbf{V}_n)_{13} &= -[(\mathbf{V}_n)_{31}]^* = 3Bn_m\mathbf{p}_-, \\ (\mathbf{V}_n)_{22} &= -(A - B)(n_+\mathbf{p}_- + n_-\mathbf{p}_+) - 2(A + 2B)n_z\mathbf{p}_z. \end{aligned}$$

The normal component of the velocity operator is proportional to the normal component of the probability density flux. Therefore, we write condition for the I-type of states in the form

$$\left(\mathbf{V}_n^{\text{in}}\psi_{f,M}^{f-1,f+1}\right)\Big|_{r=a} = \left(\mathbf{V}_n^{\text{out}}\psi_{f,M}^{f-1,f+1}\right)\Big|_{r=a}. \quad (A2)$$

Left and right parts of the previous equation are similar, that is why we continue to work only with the left part and the index in is omitted:

$$\mathbf{V}_n\psi_{f,M}^{f-1,f+1} = \mathbf{V}_n \left(R_f^{f-1}\Phi_{f,M}^{f-1} + R_f^{f+1}\Phi_{f,M}^{f+1} \right). \quad (A3)$$

The explicit form of spinors is

$$\Phi_{f,M}^{f-1} = \frac{1}{\sqrt{2f(2f-1)}} \begin{pmatrix} \sqrt{(f+M)(f+M-1)}Y_{f-1,M-1} \\ \sqrt{2(f-M)(f+M)}Y_{f-1,M} \\ \sqrt{(f-M)(f-M-1)}Y_{f-1,M+1} \end{pmatrix}, \quad (A4)$$

$$\Phi_{f,M}^{f+1} = \frac{1}{\sqrt{2(f+1)(2f+3)}} \begin{pmatrix} \sqrt{(f-M+1)(f-M+2)}Y_{f+1,M-1} \\ \sqrt{2(f-M+1)(f+M+1)}Y_{f+1,M} \\ \sqrt{(f+M+1)(f+M+2)}Y_{f+1,M+1} \end{pmatrix}. \quad (A5)$$

Operator \mathbf{V}_n contains \mathbf{p}_\pm , \mathbf{p}_z . Those operators will operate on the function $R(r)Y_{l,m}(\theta, \varphi)$. Therefore, we derive the following relation for the spherical harmonics:

$$\mathbf{p}_z(RY_{lm}) = -i\sqrt{\frac{(l+m+1)(l-m+1)}{(2l+3)(2l+1)}}Y_{l+1,m}(\mathbf{B}_l^+R) - i\sqrt{\frac{(l+m)(l-m)}{(2l+1)(2l-1)}}Y_{l-1,m}(\mathbf{B}_l^-R), \quad (A6)$$

$$\mathbf{p}_+(RY_{lm}) = \sqrt{\frac{(l+m+2)(l+m+1)}{(2l+3)(2l+1)}} Y_{l+1,m+1}(\mathbf{B}_l^+ R) - \sqrt{\frac{(l-m)(l-m-1)}{(2l+1)(2l-1)}} Y_{l-1,m+1}(\mathbf{B}_l^- R), \quad (A7)$$

$$\mathbf{p}_-(RY_{lm}) = -\sqrt{\frac{(l-m+2)(l-m+1)}{(2l+3)(2l+1)}} Y_{l+1,m-1}(\mathbf{B}_l^+ R) + \sqrt{\frac{(l+m)(l+m-1)}{(2l+1)(2l-1)}} Y_{l-1,m-1}(\mathbf{B}_l^- R). \quad (A8)$$

Insertion (A4)–(A5) into (A3), the three-component vector-column was obtained. This vector-column was multiplied by the corresponding conjugated spinors $(\Phi_{f,M}^{f-1})^+$ and $(\Phi_{f,M}^{f+1})^+$ by turns. Two expressions were obtained. Then, those expressions were integrated by the angular variables. After simple, but very huge simplifications we obtain the condition (14). In the same manner the equations (15)–(16) were derived too.

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Діркові, домішкові та екситонні стани у сферичній квантовій точці

В.І. Бойчук, І.В. Білінський, Р.Я. Лешко

Кафедра теоретичної фізики, Дрогобицький державний педагогічний університет ім. І. Франка,
вул. Стрийська 3, Дрогобич, 82100

Для сферичної наногетероструктури Si/SiO₂, нехтуючи гофрованістю ізоенергетичних поверхонь в k -просторі, використано дірковий $\vec{k}\vec{p}$ гамільтоніан 3×3 для огинаючих хвильових функцій (гамільтоніан ефективної маси) при обчислення дискретних станів дірки та водневоподібної акцепторної домішки як функції розмірів квантової точки. Дослідження проведено для скінченного потенціалу на межі гетероструктури. Визначено залежність енергетичного спектру дірки від поляризаційних зарядів, що виникають на гетеромежах, та від діелектричної проникності матриці. На основі точних розв'язків для електрона та дірки побудовано хвильову функцію і визначено енергію основного стану екситона. Проведено порівняння з експериментальними даними.

Ключові слова: *домішка, багатозонна модель, поляризаційні заряди*