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Energy and coordinate dependent effective mass and confined electron states in quantum dots

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Abstract

We present a theoretical study of the electron energy states in narrow gap semiconductor quantum dots (QDs). For a finite height hard-wall 3D confinement potential the problem was solved by using of the effective one electronic band Hamiltonian, the energy and position dependent electron effective mass approximation, and the Ben Daniel–Duke boundary condition. To solve the 3D Schrödinger equation, we employ a numerical scheme by using the finite difference method and the QR algorithm. Our results show that the parabolic band approximation is applicable only for relatively thin cylindrical QDs or for the dots with large radius. We show that the electron wave function localization plays an important role in the dependency of the energy and the electron effective mass. For the excited states, the non-parabolicity effect has been found to be stronger than it at ground state.

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1. Introduction

During the recent decade by using modern nanotechnologies, it has become possible to model quasi-zero-dimensional semiconductor systems — quantum dots (QDs) in laboratories [1–3]. The main advantage of these so called 'artificial atoms' [4] over those 'natural' is a possibility to control electronic confinement potentials ('artificial nuclei') technologically (by conditions of the dot preparation processes) and dynamically (by external fields and changing of the electron number in dots). The electronic state engineering forms novel directions in semiconductor electronics: quantum dot optoelectronics [1,3], single electronic devices [5], and quantum dot computing systems [6].

The theoretical modeling of quantum dot electronic properties can be done by different schemes. Those are pseudopotential and first-principal calculations [7–9], which consume a very large computer power. While large-scale QD calculations using complicated Hamiltonians have become feasible, they are not better than the results calculated with the input parameters and models. At the same time, the large-scale calculations are almost useless to evaluate basic physical tendencies of QD electronic properties. We should conclude that it is necessary to develop computational models and numerical methods that can be used to evaluate electronic properties over a wide range of QD parameters. With this reason one can use multi-band envelope function and the effective mass approximations with single-electron [10–13] and multi-electron descriptions [14–16]. In this class of models one needs an assumption about the electronic confinement potential in the system. Among other used confinement potential models (the parabolic lateral potential or the infinite wall potential), a finite hard wall boundary potential model is the most realistic. However, in this case the eigenvalue problem cannot be solved exactly and requires some reliable numerical methods to compute the energy states and wave functions. Different computational methods have been proposed within multi-electron descriptions. Unfortunately,

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most of practical estimations of the electron energy states were done only within the simple parabolic approximation for the electron effective mass [14–16]. The effective mass is taken as an arbitrary (has to be adjusted) parameter that can be quite different from the electronic band edge effective mass [17,18]. It can produce an error in the electron energy estimations, and this error is much larger than that one with the energy corrections form multi-electron descriptions, lattice-mismatch-induced strains, and piezo-electric fields [15,19]. Therefore, the validity of the constant effective mass approximation needs a special evaluation for small QDs of narrow gap semiconductors.

In this study we present a computational technique that used to obtain the energy states and the corresponding wave functions of an electron confined by an InAs QD embedded in GaAs matrix. To demonstrate the efficiency of the proposed simulation technique we treat the problem within the effective one electronic band Hamiltonian, the energy and position dependent electron effective mass (non-parabolic) approximation, and the Ben Daniel–Duke boundary conditions. The validity of this approximation for semiconductor quantum heterostructures was discussed in literature in details [20,21]. A hard-wall (of finite height) 3D confinement potential is induced by real discontinuity of the conduction band at the edge of the dot. The dot has a disk shape with radius R_0 and thickness z_0. We choose an isotropic effective mass description in this study, but its generalization to the anisotropic model (along R and z directions with a conservation the system symmetry) can be developed similarly. Furthermore, a good control of the dot shape and content in the experimental situation for the self-assembled narrow gap semiconductor QDs was developed recently (see, for instance Ref. [22]). From this reason we believe that the disk shape and step-like confinement potential are reasonable first order approximations in the calculation of QD energy states. In addition, in this work we concentrate on an effect of the effective mass non-parabolicity. From this reason we do not consider stresses or piezo-electric fields (this can be performed in future studies). To solve the 3D Schrödinger equation we employ a numerical scheme by using the finite difference method [23], the balanced and shifted QR algorithm [24–26], and the inverse iteration method [27].

Our main result is that in small cylindrical QDs of narrow gap semiconductors, one can use the parabolic approximation only for relatively thin (small z_0) QDs or for dots with large R_0. The electron wave function penetration into the confinement barrier region in the z direction leads to a weakening of the effects of non-parabolicity. We show that the electron wave function localization plays an important role in the dependency of the energy and the electron effective mass. Furthermore, we also have found the non-parabolicity effect for the excited states are much stronger than it at ground state.

In Section 2, we present a description of the model and method of our calculation. Section 3 is devoted to the discussion of the calculation results. Conclusions are drawn in Section 4.

2. Model and method of calculation

We consider 3D QD structures in the one-band envelope-function formalism in which the effective Hamiltonian is given by [20,21]:

$$\hat{H} = -\frac{\hbar^2}{2}\nabla_{\mathbf{r}}\left(\frac{1}{m(E,\mathbf{r})}\right)\nabla_{\mathbf{r}} + V(\mathbf{r}), \tag{1}$$

where $\nabla_{\mathbf{r}}$ stands for the spatial gradient, $m(E, \mathbf{r})$ is the electron effective mass that depends on both the energy and position. The expression of the $m(E, \mathbf{r})$ is given by:

$$\frac{1}{m(E,\mathbf{r})} = \frac{P^2}{\hbar^2}\left[\frac{2}{E + E_g(\mathbf{r}) - V(\mathbf{r})} + \frac{1}{E + E_g(\mathbf{r}) - V(\mathbf{r}) + \Delta(\mathbf{r})}\right], \tag{2}$$

where $V(\mathbf{r})$ is the confinement potential, $E_g(\mathbf{r})$ and $\Delta(\mathbf{r})$ denote, respectively, the position dependent band gap and the spin-orbit splitting in the valence bands, and P is the momentum matrix element. The approximation above is traditionally used in calculation of quantum well energy states and it well describes the electronic properties of the 2D heterostructures. This is a case when the non-parabolicity effects should be taken into account [20]. We apply here this approximation to calculate energy states of the 3D QDs of a narrow gap semiconductor.

For systems with a sharp discontinuity of the conduction band at the interface between the QD (material 1) and the crystal matrix (material 2), the hard-wall confinement potential can be presented as:

$$V(\mathbf{r}) = \begin{cases} 0, & \mathbf{r} \in \text{material 1} \\ V_0, & \mathbf{r} \in \text{material 2} \end{cases}. \tag{3}$$

Integrating the Schrödinger equation with Hamiltonian (1) along the direction perpendicular to the interface, we obtain the Ben Daniel–Duke boundary conditions for the wave function $\Psi(\mathbf{r})$

$$\Psi_{\text{material 1}}(\mathbf{r}_s) = \Psi_{\text{material 2}}(\mathbf{r}_s)$$
$$\frac{\hbar^2}{2m(E,\mathbf{r}_s)}\nabla_n\Psi(\mathbf{r}_s) = \text{const}, \tag{4}$$

where $\mathbf{r}_s$ denotes the position on the interface. These boundary conditions are dependent on the electron energy and originate from the parameter difference between different materials.We consider a disk-shaped QD with the radius R_0 and the thickness z_0 in the cylindrical coordinates (R, ϕ, z). The origin of the system lies at the center of the disk and the z-axis is chosen along the rotation axis. Since

the system is cylindrically symmetric, the wave function can be represented as:

$$\Psi(\mathbf{r}) = \Phi(R, z)\exp(il\phi), \tag{5}$$

where $l = 0, \pm 1, \pm 2, \ldots$ is the electron orbital quantum number and the problem remains 2D in (R, z) coordinates:

$$-\frac{\hbar^2}{2m_1(E)}\left(\frac{\partial^2}{\partial R^2} + \frac{\partial}{R\partial R} + \frac{\partial^2}{\partial z^2} - \frac{l^2}{R^2}\right)\Phi_1(R, z) = E\Phi_1(R, z), \tag{6}$$

$$R \le R_0,\ |z| \le \frac{z_0}{2},$$

$$-\frac{\hbar^2}{2m_2(E)}\left(\frac{\partial^2}{\partial R^2} + \frac{\partial}{R\partial R} + \frac{\partial^2}{\partial z^2} - \frac{l^2}{R^2}\right)\Phi_2(R, z) + V_0\Phi_2(R, z) = E\Phi_2(R, z),$$

$$R > R_0,\ |z| > \frac{z_0}{2},$$

and the boundary conditions (4) become of the form

$$\Phi_1(R_0, z) = \Phi_2(R_0, z), \qquad |z| \le \frac{z_0}{2}, \tag{7}$$

$$\Phi_1\left(R, \pm\frac{z_0}{2}\right) = \Phi_2\left(R, \pm\frac{z_0}{2}\right), \qquad R \le R_0,$$

$$\frac{1}{m_1(E)}\frac{\partial\Phi_1(R, z)}{\partial R}\bigg|_{R_0} = \frac{1}{m_2(E)}\frac{\partial\Phi_2(R, z)}{\partial R}\bigg|_{R_0}, \qquad |z| \le \frac{z_0}{2},$$

$$\frac{1}{m_1(E)}\frac{\partial\Phi_1(R, z)}{\partial z}\bigg|_{\pm\frac{z_0}{2}} = \frac{1}{m_2(E)}\frac{\partial\Phi_2(R, z)}{\partial z}\bigg|_{\pm\frac{z_0}{2}}, \qquad R \le R_0.$$

The electron energy states here are complicated functions of the dot parameters and the electron angular momentum. We can obtain the solution by means of a numerical approach to solve the Schrödinger Eq. (6) together with the boundary conditions (7).

Due to the energy dependence on the effective electron mass the calculation should consist of some iteration loops to reach a 'self-consistent' energy solution. A feedback nonlinear iteration scheme is proposed here: (i) set initial energy $E = 0$, (ii) compute effective mass m, (iii) solve Schrödinger equation for energy E, (iv) update the newer energy E and back to (ii). The iteration will be terminated when a specified stopping criterion on energy is reached. To obtain a complete numerical solution of the 3D Schrödinger equation in step (iii), a finite difference method [23] with non-uniform mesh technique is firstly applied to discretize the Schrödinger Eq. (6). The discretized Schrödinger equation with the boundary conditions (7) leads to an

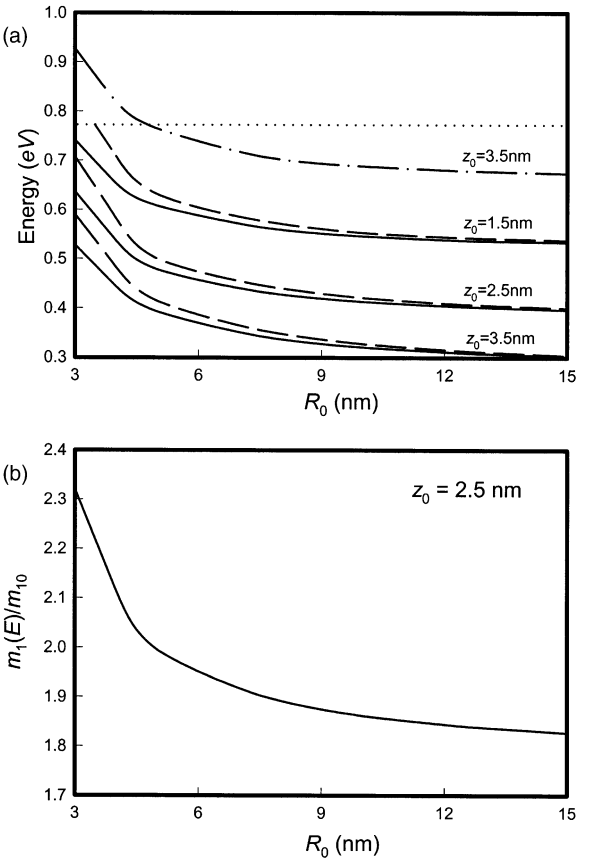

Fig. 1. (a) The ground state {1,0} energies (as a function of the dot size) for the non-parabolic (solid lines) and parabolic (dash lines) approximation, respectively. Horizontal dot line shows the top energy of the well ($E = V_0 = 0.77$ eV). Dash-dot line indicates the non-parabolic approximation results for the infinite well model ($v_0 \to \infty$). (b) Ratio of effective masses in non-parabolic $m_1(E)$ and parabolic m_{10} approximations as a function on the dot radius ($z_{0\,=\,2.5\ nm}$) for the ground state {1,0}.

eigenvalue problem

$$\mathbf{AX} = \lambda\mathbf{X}, \tag{8}$$

where $\mathbf{A}$ is the matrix rising from discretized Schrödinger equation with the boundary conditions, $\mathbf{X}$ and λ are the corresponding eigenvectors (wave functions) and the eigenvalues (energy levels) of the matrix $\mathbf{A}$. In general, the matrix $\mathbf{A}$ is a non-symmetric and sparse matrix, the eigenvalues of such matrix can be very sensitive to small changes in the matrix elements. In order to reduce the sensitivity of eigenvalues, we perform a balancing algorithm [24] to matrix $\mathbf{A}$. Then the next strategy for finding the eigenvalues of the balanced matrix $\mathbf{A}$ is transferring it into a simpler matrix form, Hessenberg form, with a sequence of Householder transformations [24]. The eigenvalues of the Hessenberg matrix are directly computed with the shifted QR method

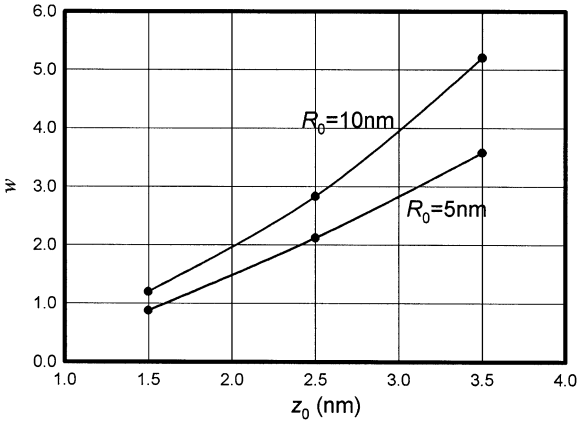

Fig. 2. Plot of the parameter w versus the dot size.

[24,26]. When the eigenvalues are found, we solve the corresponding eigenvectors with the inverse iteration method [27]. The fundamental idea of this method is to solve the following linear system

$$(\mathbf{A} - \zeta I)y = \mathbf{b},$$

where $\mathbf{b}$ is a trial vector and ζ is one of the computed eigenvalues of matrix $\mathbf{A}$. The solution y is right the candidate eigenvector corresponding to ζ.

The problem of finding all eigenvalues of a non-symmetric matrix $\mathbf{A}$ can be quite unstable with respect to perturbations in the coefficients of $\mathbf{A}$. Some traditional methods, such as the power method, the Lanczos algorithm, etc. [28], could be used for solving the eigenvalues of a symmetric matrix. However, the QR method used here is the best-suited general method for the calculations of all eigenvalues of a non-symmetric matrix [25]. From our calculation experience and a mathematical fact that the effective electron mass depends on energy monotonically, the convergence criteria on energies (the maximum norm error is less than (10^{-7} eV) can be fast reached by taking only 8–10 feedback non-linear iterative loops.

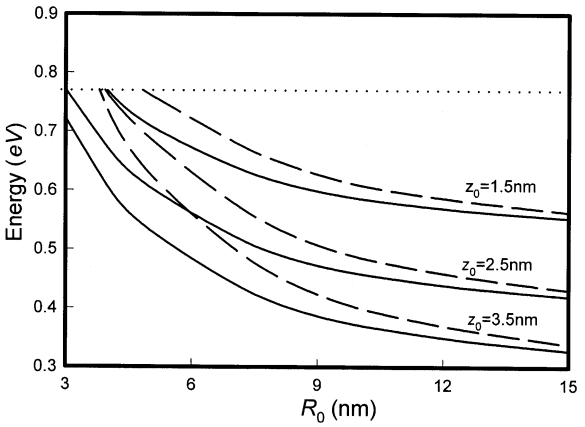

Fig. 3. The first excited state {1,1} energy of the dot.

3. Calculation results

The energy spectrum of the dot consists of a set of discrete levels numerated by the set of numbers $\{n, l\}$, where n is the nth solution of the problem with fixed l. In calculations of the electron energy spectra for narrow gap InAs cylindrical QDs in GaAs matrix we choose the semiconductor band structure parameters for InAs: the energy gap $E_{1g} = 0.42$ eV, the spin-orbit splitting $\Delta_1 = 0.48$ eV, the value of the non-parabolicity parameter $E_{1p} = 3m_0P_1^2/\hbar^2 = 22.2$ eV, and m_0 is the free electron effective mass. For GaAs we choose: $E_{2g} = 1.52$ eV, $\Delta_2 = 0.34$ eV, and $E_{2p} = 24.2$ eV. The band offset is taken as $V_0 = 0.77$ eV.

In Fig. 1(a), we show the calculated ground state {1,0} energy depending on the dot size. The non-parabolic correction leads to a decrease in the state energy. The difference between parabolic (when $m_i(E) = m_{i0},\ i = 1, 2$ — the band edge electronic effective mass) and non-parabolic approximation results gains ~0.05 eV for $R_0 \approx 3$ nm. This difference can exceed known electron–electron interaction corrections [14–16]. As shown in Fig. 1(b), a ratio of effective masses in non-parabolic and parabolic approximations as a function on the dot radius is presented. The actual effective mass $m_1(E)$ in this figure was calculated by substituting the state energy into Eq. (2).

For dots of relatively large radius ($R_0 > 20$ nm) and lower ground state energy we can neglect the non-parabolicity effects. At the same time, dots of a small height ($z_0 \sim 1.5$ nm) demonstrate a suppression of the non-parabolicity effect in spite of the increase of the state energy. This unusual effect is a direct result that the electron effective mass depends energy as well as position and the electron wave functions are tunneling into the barrier along the z direction. To clarify this result we compare 'electronic weights' inside and outside the dot by the following ratio:

$$w = \frac{\int_{\mathbf{r}\in\text{material 1}} d\mathbf{r}^3 |\Phi(R,z)|^2}{\int_{\mathbf{r}\in\text{material 2}} d\mathbf{r}^3 |\Phi(R,z)|^2}.$$

The w parameter depends on the dot size is presented in Fig. 2. For QDs of small heights, the electron wave function spreads out of the dot ($w \sim 1$) and the energy level properties are controlled by band parameters of GaAs matrix. In this situation, an actual electron effective mass is a weak function of the electron energy (as for GaAs). For QDs with relatively large heights, the electron state energy is controlled by InAs band parameters and is affected by the non-parabolicity effect.

Fig. 3 shows the first {1,1} excited energy states of the dot. As it can be seen from the figure, the non-parabolic

approximation leads to large corrections in this case. A difference between parabolic and non-parabolic estimations can gain 0.1 eV.

4. Conclusions

In conclusions, we have studied theoretically electron energy states of cylindrical hard wall QDs with respect to various sizes by using the one-band envelope-function formalism and the non-parabolic approximation. The studied model was solved with the finite difference and QR methods to obtain the all energies and wave functions.

It has been found that the widely used parabolic electron band approximation leads to a large discrepancy in the calculation results for electron energy states in small cylindrical QDs. This approximation can be used only for QDs of large radius or small height. The penetration of the electron wave function into barrier region in the z direction (as a result of the finite hard wall confinement potential) plays an important role for reducing the effective mass dependence on the energy. For the excited states, the non-parabolicity effect is much stronger than it is at the ground state and can exceed electron–electron interaction corrections known from literature. The analysis presented here is useful to clarify principal dependencies of QD energy states on semiconductor material band parameters and QDs sizes.

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