Electron energy state dependence on the shape and size of semiconductor quantum dots

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In this article we present a unified model for studying the effect of the sizes and shapes of small semiconductor quantum dots on the electron and hole energy states. We solved the three-dimensional effective one band Schrödinger equation for semiconductor quantum dots with disk, lenticular, and conical shapes. For small InAs/GaAs quantum dots we found a substantial difference in the ground state and first excited state electron energies for dots with the same volume but different shapes. Electron energy dependence on volume is found to be quite different from the commonly quoted $V^{-2/3}$. The exponent can vary over a wide range and depends on the dot shapes. [DOI: 10.1063/1.1412578]

I. INTRODUCTION

Recent advances in the fabrication of semiconductor quantum dots have generated huge quantities of experimental and theoretical data.[1,2] The three-dimensional confinement of charge carriers in various structures provides fascinating optical and magnetic characteristics for many important device applications. The intensive investigation, to a large part, is driven by the prospect of fabricating a new generation of electronic and photonic devices (quantum dot lasers for instance).[2]

The spectral broadening in semiconductor quantum dots caused by the nonuniformity in their size and shape is of primary concern for practical laser applications.[2–5] Many studies were carried out for fabrication of quantum dots with a small size variation.[6–11] The best result so far is achieved in the so-called self-assembled InAs quantum dots grown on GaAs substrate by the Stranski–Krastanow mode. It has been shown experimentally that InAs dots can be designed with sharp electronic multilevel shells[12–15] in which the exited state emission is narrower than the ground state emission.[3,4] Various experimental results suggest, not yet without a controversy, that InAs/GaAs quantum dots can have disk, lens, or cone shapes with a circular top view cross section and a large area-to-height aspect ratio.[16–23] Unfortunately, no consistent description of the dot shape can be drawn from the literature due to the different conditions used in the dot formation.

The shape of quantum dots is debated intensively in theoretical works since an accurate calculation (and explanation) of the electronic structure depends on the dot shape itself. A wide range of shapes and sizes have been used in the theoretical models to simulate InAs dot properties. Most of them use numerical methods. The commonly used shapes include disk,[24,25] lens,[26,27] and cone shapes.[28,29] Spherical,[30–32] pyramidal,[33–38] and cubic[39,40] shapes were also used. The energy level calculation has been done using the effective-mass approximation with[24,25,28,29,32,33,37,40] and without[26,27,38,39] the coordinate dependence for the effective mass. The multiband **k·p** method with finite[33,34,36] and infinite[30,31] confinement potentials, and the pseudopotential method were used in the calculation.[35]

The diversity in the theoretical model and approach makes it difficult to compare the theoretical results of different authors and to verify the models on the basis of experimental results. A comprehensive analysis of the influence of the dot size and shape on the electron energy states by using a unified model of the semiconductor band structure has not been done yet. While large-scale calculations using complicated Hamiltonians have become feasible, the results are not better than those using the input parameters and dot shape models. For instance, the multielectron interaction and other factors in small quantum dots generally affect the electron energy in the order of a few meV. But, at the same time, the variations of the dot size and shape can produce an energy change up to an order of 0.1 eV in the strong confinement region.

In this study we calculate and compare the electron energy spectra for three-dimensional small InAs/GaAs quantum dots of four different shapes (see Fig. 1): disk (DI),

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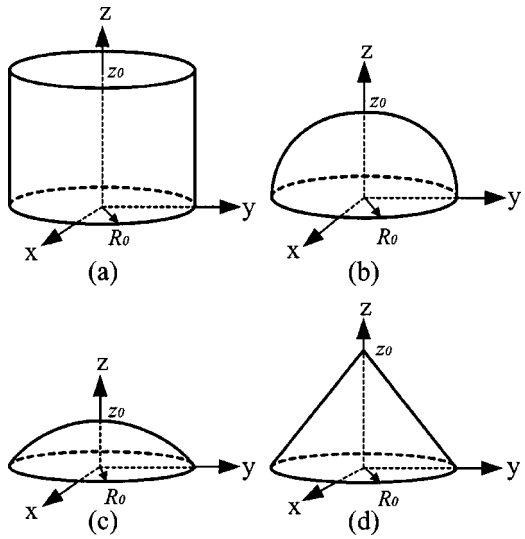

FIG. 1. Schematic diagrams for quantum dots of four different shapes: (a) DI, (b) EL, (c) CL, and (d) CO.

ellipsoidal lens (EL), cut sphere lens (CL), and conical shape (CO). All of them are cylindrically symmetric (with the circular top view cross section). We use the effective one electronic band Hamiltonian, the energy and position dependent effective mass approximation, and the Ben Daniel–Duke boundary conditions. To solve the three-dimensional Schrödinger equation we employ a robust numerical scheme by using the finite difference method,[41,42] a shifted and balanced QR algorithm,[43,44] and the inverse iteration technique.[43,45]

We show that the dependence of the electron energy level on the dot volume V can be quite different from the commonly quoted $V^{-2/3}$ rule. It can be formulated as $V^{-\gamma}$, where the effective exponent γ depends on the dot's shape. The effective exponent is different with respect to the ground state and the exited states.

This article is organized as follows. Section II introduces the theoretical models and the calculation methods. Section III describes the calculated results illustrating the dependence of the electron energy level on the dot volume for different dot's shapes. Section IV draws conclusions.

II. THEORETICAL MODEL AND CALCULATION METHOD

We consider semiconductor quantum dots in the one-band envelope-function formalism for electrons and holes in which the effective Hamiltonian is given by[46]

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

where $\nabla_{\mathbf{r}}$ stands for the spatial gradient. The electron effective mass $m(E,\mathbf{r})$, depending on both energy E and position, is taken as

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

where $V(\mathbf{r})=E_c(\mathbf{r})$ is the confinement potential, $E_c(\mathbf{r})$, $E_g(\mathbf{r})$, and $\Delta(\mathbf{r})$ denote, respectively, the position dependent electron band edge, band gap, and the spin-orbit splitting in the valence band, and P is the momentum matrix element. The hole effective mass $m_h(\mathbf{r})$ is assumed to be only position dependent.

We investigate quantum dots with shapes of DI, EL, CL, and CO with the base (top view) radius R_0 and height z_0 in the cylindrical coordinates $(R,\ \phi,\ z)$. Since the system is cylindrically symmetric, the wave function can be written as

$$\Psi(\mathbf{r})=\Phi(R,z)\exp(il\phi), \tag{3}$$

where $l=0,\pm1,\pm2,...$ is the electron orbital quantum number. The problem remains two dimensional in $(R,\ z)$ coordinates:

$$-\frac{\hbar^2}{2m_i(E)}\left(\frac{\partial^2}{\partial R^2}+\frac{\partial}{R\partial R}+\frac{\partial^2}{\partial z^2}-\frac{l^2}{R^2}\right)\Phi_i(R,z)+V_i(R,z)\Phi_i(R,z)=E\Phi_i(R,z), \tag{4}$$

where $V_1(R,z)=0$ $(i=1)$ inside and $V_2(R,z)=V_0$ $(i=2)$ outside the dot. The boundary conditions are

$$\Phi_1(R,z)=\Phi_2(R,z),\ \ z=f_S(R),$$
$$\frac{1}{m_1(E)}\left\{\frac{\partial\Phi_1(R,z)}{\partial R}+\frac{df_S}{dR}\frac{\partial\Phi_1(R,z)}{\partial z}\right\}\Bigg|_{z=f_S(R)}=\frac{1}{m_2(E)}\left\{\frac{\partial\Phi_2(R,z)}{\partial R}+\frac{df_S}{dR}\frac{\partial\Phi_2(R,z)}{\partial z}\right\}\Bigg|_{z=f_S(R)}, \tag{5}$$

where $z=f_S(R)$ $(S=\mathrm{DI,EL,CL,CO})$ is the contour of the structure's cross section on the $\{R,\ z\}$ plane. The structure shape is generated by the rotation of this contour around the z axis.

Based on the fact that the electron effective mass is a spatial and energy dependent function, the Schrödinger equation is a nonlinear equation in energy. A computational method for such a nonlinear problem has been proposed and successfully implemented for the spin-splitting quantum dot problem by us[47] recently. Due to the energy dependence of the electron effective mass, our calculation consists of iteration loops to reach a ''self-consistent'' energy solution. In each iteration we use a central difference method with a nonuniform mesh technique[41] to discretize the two-dimensional Schrödinger equation. The discretized Schrödinger equation together with its boundary conditions Eq. (5) leads to the eigenvalue problem

$$A\mathbf{X}=\lambda\mathbf{X},$$

where A is the matrix rising from the discretized Schrödinger equation and boundary conditions, and $\mathbf{X}$ and λ are the corresponding eigenvectors (wave functions) and the eigenvalues (energy levels), respectively. Because the matrix A is an energy dependent, five diagonal and nonsymmetric matrix,[47] we perform a balancing algorithm to reduce the sensitivity of eigenvalues of the matrix A to small changes in the matrix elements.[43] Then the matrix A is transformed into a simpler upper Hessenberg form. The eigenvalues of the upper Hes-

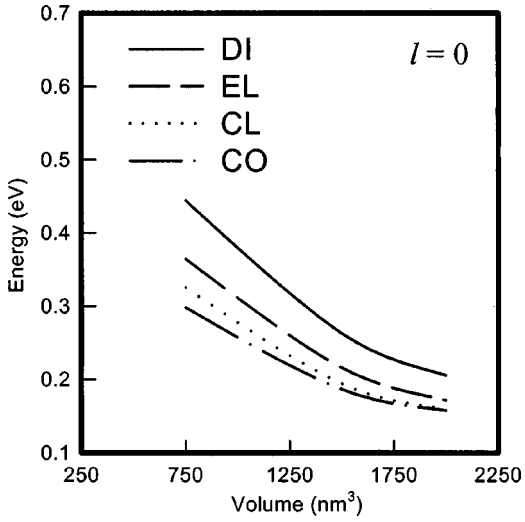

FIG. 2. The electron ground state ($l=0$) energy levels for InAs/GaAs quantum dots versus the dot volume. The structure's base radius is fixed at 10 nm for all shapes. The solid, dash, dot, and dash-dot lines correspond to DI, EL, CL, and CO shapes, respectively.

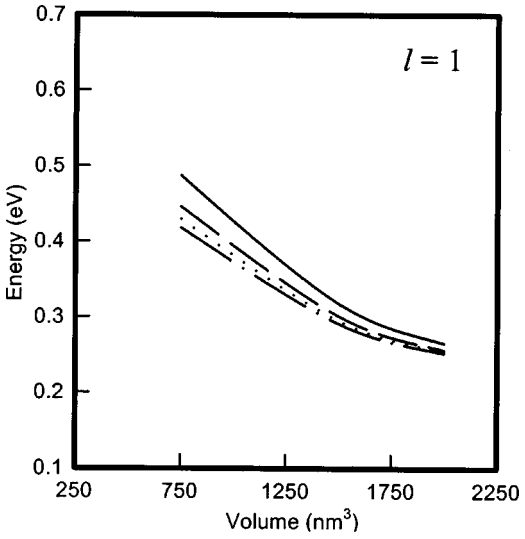

FIG. 4. The energy position of the first excited electron states ($l=1$) for the dots with the same parameters of Fig. 2.

senberg matrix are directly computed with the QR method.[43] When the eigenvalues are found, we solve the corresponding eigenvectors with the inverse iteration method.[45] In our calculation experience, the proposed computational method converges monotonically, and a strict convergence criteria on energies (the maximum norm error is less than 10^{-12} eV) can be reached by only 12–15 feedback nonlinear iterative loops.

III. CALCULATION RESULTS

In Fig. 2 we present the calculated electron energy levels for InAs/GaAs quantum dots as functions of the dot volume. The results are plotted relative to the InAs conduction band edge. For InAs, the energy gap E_{1g} is 0.42 eV, spin-orbit Δ_1 is 0.42 eV, the value of the nonparabolicity parameter E_{1p} is $3m_0P_1^2/\hbar^2=22.2$ 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 $V_{e0}=0.77$ eV.[46] The base radius of the dots is fixed at $R_0=10$ nm for all shapes. Notice that the range of the dot volume and the radius of the base were take from available experimental data.[7] Our model predicts rather different electron energy dependences on the volume for dots of different shapes. When the dot volume increases the energy states of different shapes converge. The most sensitive to the dot volume variation is the quantum disks and the least is that of the conical shape dots. This is no surprise since the electron wave function is the best confined for the disk geometry when the volume and the radius are fixed. The electron ground state wave functions with a fixed 750 nm^3 dot volume for all shapes are plotted in Fig. 3. The wave function shape confirms weaker confinement for conical shaped dots. The first excited state ($l=1$), however, has demonstrated a weaker sensitivity to the dot shape and volume[3,4] (see Fig. 4). This is because that the electron wave functions of the

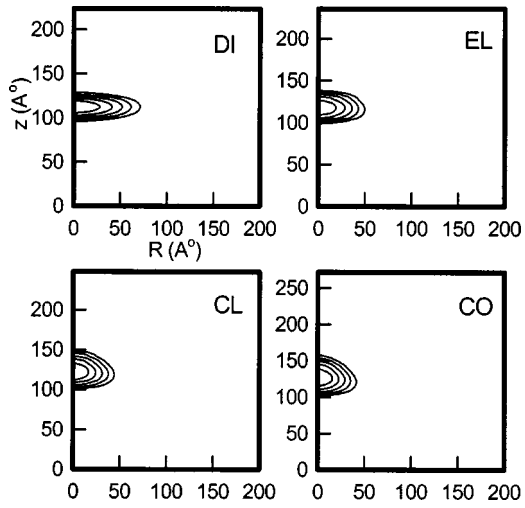

FIG. 3. Contour plot of the electron ground state wave functions where the dot volume is fixed at 750 nm^3 for all shapes. All parameters are the same as in Fig. 2.

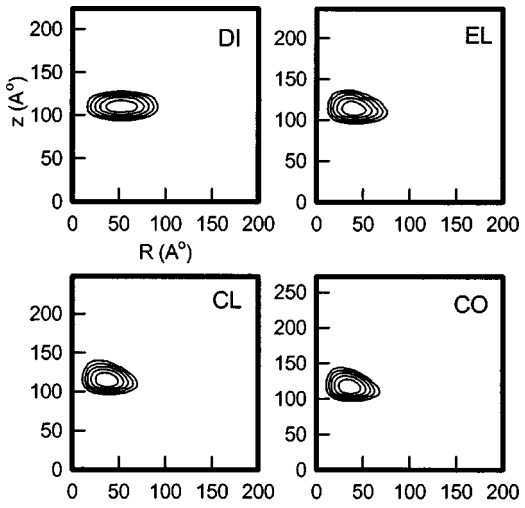

FIG. 5. Contour plot of the first excited electron state wave functions where the dot volume is fixed at 750 nm^3 for all shapes. All parameters are the same as in Fig. 2.

TABLE I. Parameter γ for electron energy levels.

Base radius	5 nm		10 nm		15 nm	
State	$l=0$	$\|l\|=1$	$l=0$	$\|l\|=1$	$l=0$	$\|l\|=1$
DI	0.31	0.12	0.74	0.61	0.76	0.63
EL	0.30	0.12	0.72	0.60	0.74	0.61
CL	0.30	0.12	0.69	0.58	0.70	0.59
CO	0.30	0.12	0.65	0.56	0.66	0.56

excited states (see Fig. 5) are less confined and, therefore, are less sensitive to the dot shape and size.

To investigate the dependence of electron energy level on the dot volume V more generally we fitted the dependence $E \sim V^{-\gamma}$ to our calculation results. The parameter γ obtained by this fitting is presented in Table I for different shapes and base radii of the dots. It is clear from Table I that the parameter is rather different for all dot shapes and can vary widely from the commonly quoted 2/3 value. Based on the calculation we did, the first excited state of a cylindrical dot with $R_0=5$ nm has the lowest γ parameter 0.12. Based on the wave function confinement discussed above, the largest γ parameter was also obtained from the disk shape quantum dot. The ground state energy of the disk with $R_0=15$ nm has a γ value of 0.76. In contrast to the result of Ref. 32 the geometry variation allows us to obtain a wide range of γ values different from the conventional 2/3.

Using the same calculation method we obtained hole energy states for dots of the same shapes. The hole effective mass was taken as $m_{1h}=0.4m_0$ and $m_{2h}=0.5m_0$,[46] respectively. The hole band offset is taken as $V_{h0}=0.33$ eV. The hole energy states were found to be least dependent on the dot shapes. With base radius $R_0=10$ nm, the fitted γ parameters for those dependencies are presented in Table II. The difference in γ obtained for $e_0 \rightarrow h_0$ and $e_1 \rightarrow h_0$ suggests that the transition lines should converge in optical experiments when the size of the dot decreases. A similar tendency has been observed experimentally in InAs quantum dots.[4,12]

IV. CONCLUSIONS

We have presented a calculational approach to compare the electron energy states for small quantum dots of four different shapes. This simple method with very limited computational demands is useful to analyze the dependence of the quantum dot spectra (ground and excited states) on the dot size and shape variations. We found a large (about 0.1 eV) difference in the electron ground state energy of InAs/GaAs dots with the same volume but different shapes. It is found that the γ parameter in the $V^{-\gamma}$ rule derived from curve fitting deviates from the conventional 2/3 value. Furthermore, it is dependent on dot shape. The excited states as a rule have smaller γ parameter than the ground states.

TABLE II. Parameter γ for energy of transitions.

Transition	$e_0 \rightarrow h_0$	$e_1 \rightarrow h_0$
DI	0.38	0.36
EL	0.35	0.32
CL	0.31	0.29
CO	0.27	0.26

In our calculations we also derived hole energy states for the dots and estimated hole–electron transition energies. Our results suggest that the inhomogeneous broadening of excited level transitions is sufficiently less than that for the ground state transitions. Different volume dependence of the energy states for different dot shapes can be useful in tuning the intersublevel energy spacing when we prepare the quantum dots with different sizes and shapes.

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