Energy Spectra of an Electron in a Pyramid-shaped Quantum Dot in Effective Mass Approximation with Even Mirror Boundary Conditions

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Abstract: - We study the problem of an electron confined in a semiconductor quantum dot shaped as a square-base pyramid, assuming specular reflection of a particle from quantum dot boundaries. The application of boundary condition equals the wave function in an arbitrary point inside the dot with its image formed in the walls, allowing to find wave the functions and energy spectra of the particle. The comparison of the resulting energies with published experimental data shows reasonable agreement between the theory and the experiment.

Key-Words: - Quantum dot, Pyramid, Schrödinger equation, Mirror boundary conditions

1 Introduction

Quantum dots (QDs) take a central place in modern material science. Among these, pyramidal-shaped formations have a special importance as they appear in semiconductor devices of different kinds, such as lasers [1], photonic crystals [2], solar cells [3], etc. Naturally, formations with such a well-defined geometry should significantly influence the energy spectrum of the electron. Even roughly-pyramidal shapes formed on CdSe annealed surfaces change considerably the optical spectra of the material [4], making it timely and important to study the problem of an electron concealed in pyramid-shape quantum dots.

The solution of the Schrödinger equation for this geometry is complicated, because even in the two-dimensional case Laplace's equation becomes non-analytical in the vicinity of certain corner angles [5], so that the system can be studied only numerically. Currently, there was a considerable progress in finite volume modeling of square-based pyramid with the Jacobi-Davidson method [6]. A similar simulation technique was applied in [7] to a truncated pyramid made of InP, covered with InAs quantum dots. However, it is extremely important and interesting to find analytical solutions of the problem, even if this would lack several states due to restrictions of non-analyticity.

To tackle similar problems we proposed mirror-reflection boundary conditions, which were useful for the solution of the Schrödinger equation for triangular and hexagonal dots [8]. The idea of the method consists in a specular reflection of the particle wave-function from the sides of the quantum dot, forming a standing wave pattern. It should be noted that the concept of particle reflection from the boundary of the nanostructure is evidently favorable for the effective mass approximation, because it increases the effective path of a particle in the semiconductor material.

According to Refs. [8, 9], the treatment of QD boundaries as mirrors allows to equalize the Ψ-functions for a point inside the dot and its image by the absolute value, because the physical meaning of the wave function is associated with Ψ*Ψ. Thus, we can introduce *even* and *odd* mirror boundary conditions depending if the wave function sign is allowed to change or not. The case of odd boundary condition is equivalent to impenetrable walls, making the Ψ-function vanish at the boundary. Evidently, this situation corresponds to the case of strong quantum confinement. The even mirror boundary conditions describe the milder case of weak confinement, when a particle can penetrate into the barrier (with non-vanishing Ψ at the boundary) and return into the confined volume afterwards. In this paper, we will focus on even mirror boundary conditions.

2 Theoretical Model

Let us consider a pyramid with a square base $a \times a$ and a height $a/2$, Fig. 1. The figure also shows the central cross-section of the pyramid with an arbitrary point within it and reflections thereof in the sides of the pyramid. To solve the Schrödinger equation, we will consider a set of successive reflections of a plane wave propagating perpendicularly to the planes delimiting the quantum dot.

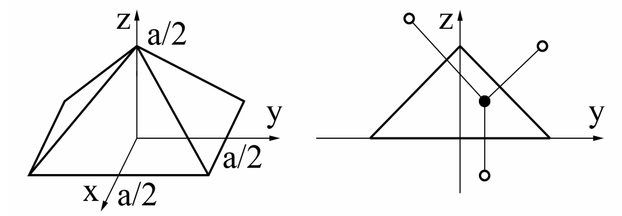

Fig. 1. Pyramidal quantum dot with square base; the right panel shows its central cross-section (including z-axis) with original (filled) and reflected (hollow) points marked.

The conditions applied to the wave function vary with reflection plane. For the base plane z=0 they are

$$\Psi(x,y,z)=\Psi(x,y,-z)\,. \tag{1}$$

For the side planes $\pm x + z = a/2$ they are

$$\begin{aligned}\Psi(x,y,z)&=\Psi(-z+a/2,y,-x+a/2)\\ \Psi(x,y,z)&=\Psi(z-a/2,y,x+a/2)\end{aligned}\;; \tag{2}$$

for the planes $y + z = a/2$ and $-y + z = 0$ they are

$$\begin{aligned}\Psi(x,y,z)&=\Psi(x,-z+a/2,-y+a/2)\\ \Psi(x,y,z)&=\Psi(x,z-a/2,y+a/2)\end{aligned}\,. \tag{3}$$

Let us consider a Ψ-function describing a wave propagating along the z-axis with wave vector $\mathbf{k} = -k\,\mathbf{e}_z$. This wave will get reflected from the base of the pyramid in the direction of $+\mathbf{e}_z$. The two waves further reflect from the planes $\pm x + z = a/2$ and $\pm y + z = a/2$ along the directions $\pm\mathbf{e}_x$ and $\pm\mathbf{e}_y$, respectively.

The resulting Ψ-function can be found as a linear combination of the aforementioned waves:

$$\begin{aligned}\Psi &= Ae^{ikx}+Be^{-ikx}+Ce^{iky}\\ &+De^{-iky}+Ee^{ikz}+Fe^{-ikz}\end{aligned} \tag{4}$$

From Eq. (1) it follows that $F = E$; similarly, by applying boundary conditions (2) one will obtain $E = B\exp(-ika/2) = A\exp(-ika/2)$ and $F = A\exp(ika/2) = B\exp(ika/2)$. The conditions (3) imply that $E = D\exp(-ika/2) = C\exp(-ika/2)$ and $F = C\exp(ika/2) = D\exp(ika/2)$. The coefficients can then be expressed as $A = B = C = D = N\exp(\pm ika/2)/2$ and $E = F = N/2$ for any arbitrary amplitude N. The quantization condition $\exp(ika/2) = \exp(-ika/2)$ is equivalent to $\exp(ika) = 1$, yielding the wave number $k = 2\pi n/a$. The solution is therefore given by

$$\Psi = N\left\{\cos\left[k(x-\frac{a}{2})\right]+\cos\left[k(y-\frac{a}{2})\right]+\cos(kz)\right\} \quad (5)$$

with the corresponding energy

$$E = \frac{\hbar^2 k^2}{2m} = \frac{h^2 n^2}{2ma^2}. \quad (6)$$

For the sake of presentation simplicity, we denote the effective mass of the particle as m in (6) and the further expressions for the energy.

Alternatively, one considers a wave propagating normal to the plane $x + z = a/2$ with wave vector $\mathbf{k} = (\mathbf{e}_x + \mathbf{e}_z)/\sqrt{2}$. The reflected wave undergoes another reflection from the pyramid's base $z = 0$, resulting in $\mathbf{k} = \pm(-\mathbf{e}_x + \mathbf{e}_z)/\sqrt{2}$, normal to the plane $-x + z = a/2$. These four waves reflect from the planes $\pm y + z = a/2$, yielding the wave vectors $\pm(\mathbf{e}_y + \mathbf{e}_z)/\sqrt{2}$, $\pm(-\mathbf{e}_y + \mathbf{e}_z)/\sqrt{2}$, $\pm(\mathbf{e}_x + \mathbf{e}_y)/\sqrt{2}$ and $\pm(-\mathbf{e}_x + \mathbf{e}_y)/\sqrt{2}$.

The final Ψ-function describing the system can be found as linear combination of those participating waves:

$$\begin{aligned}\Psi &= A_1 e^{\frac{ik}{\sqrt{2}}(y+z)} + A_{-1} e^{\frac{ik}{\sqrt{2}}(y-z)} + A_{-2} e^{\frac{ik}{\sqrt{2}}(-y+z)} \\ &+ A_{-2} e^{\frac{ik}{\sqrt{2}}(-y-z)} + B_1 e^{\frac{ik}{\sqrt{2}}(x+z)} + B_{-1} e^{\frac{ik}{\sqrt{2}}(x-z)} \\ &+ B_2 e^{\frac{ik}{\sqrt{2}}(-x+z)} + B_{-2} e^{\frac{ik}{\sqrt{2}}(-x-z)} + C_1 e^{\frac{ik}{\sqrt{2}}(x+y)} \\ &+ C_2 e^{\frac{ik}{\sqrt{2}}(x-y)} + C_3 e^{\frac{ik}{\sqrt{2}}(-x+y)} + C_4 e^{\frac{ik}{\sqrt{2}}(-x-y)}\end{aligned} \quad (7)$$

where k is the absolute value of the wave vector.

Proceeding as before, we use Eq. (1) to obtain $A_{-1} = A_1$, $A_{-2} = A_2$, $B_{-1} = B_1$, $B_{-2} = B_2$. From boundary conditions (2) it follows that:

$B_{-2} = B_1\exp(ika/\sqrt{2})$, $C_1 = A_{-1}\exp(-ika/\sqrt{8})$,
$C_2 = A_{-2}\exp(-ika/\sqrt{8})$, $C_3 = A_1\exp(ika/\sqrt{8})$,
$C_4 = A_2\exp(-ika/\sqrt{8})$,

and

$B_2 = B_{-1}\exp(-ika/\sqrt{2})$, $C_1 = A_1\exp(ika/\sqrt{8})$,
$C_2 = A_2\exp(ika/\sqrt{8})$, $C_3 = A_{-1}\exp(-ika/\sqrt{8})$,
$C_4 = A_{-2}\exp(-ika/\sqrt{8})$.

Finally, from Equations (3) one will obtain

$A_{-2} = A_1\exp(ika/\sqrt{2})$, $C_1 = B_{-1}\exp(ika/\sqrt{8})$,
$C_2 = B_1\exp(ika/\sqrt{8})$, $C_3 = B_{-2}\exp(-ika/\sqrt{8})$,
$C_4 = B_2\exp(-ika/\sqrt{8})$,

and

$A_{-1} = A_2\exp(ika/\sqrt{2})$, $C_1 = B_1\exp(ika/\sqrt{8})$,
$C_2 = B_{-1}\exp(-ika/\sqrt{8})$, $C_3 = B_2\exp(ika/\sqrt{8})$,
$C_4 = B_{-2}\exp(-ika/\sqrt{8})$.

The coefficients in Eq. (7) express as $A_{-1} = A_1 = A_{-2} = A_2 = B_{-1} = B_1 = B_{-2} = B_2 = N\exp(\pm ika/\sqrt{8})/2$ and $C_1 = C_2 = C_3 = C_4 = N/4$, for any arbitrary value of N. The quantization condition is $\exp(ika/\sqrt{8}) = \exp(-ika/\sqrt{8})$, equivalent to $\exp(ika/\sqrt{2}) = 1$, leading to $k/\sqrt{2} = 2\pi n/a$.

Therefore, the solution for the wave propagating normal to a side of the pyramid is:

$$\begin{aligned}\Psi = N\Big\{&\cos\left[k\left(y-\frac{a}{2}\right)\right]\cos(kz) \\ &+\cos\left[k\left(x-\frac{a}{2}\right)\right]\cos(kz) \\ &+\cos\left[k\left(x-\frac{a}{2}\right)\right]\cos\left[k\left(y-\frac{a}{2}\right)\right]\Big\}\end{aligned} \quad (8)$$

with the energy of the particle

$$E = \frac{\hbar^2 k^2}{2m} = \frac{h^2 n^2}{ma^2}. \quad (9)$$

It should be noted that in the two-dimensional analog of the studied QD (quantum well shaped as a bilateral rectangular triangle) the energy is described by the same expression (9) and the wave function represents a particular solution mentioned above; Ref. [12] shows good correlation of calculated energy spectra with the experimental data. The largest length of de Broglie wave (for the smallest wave vector, $n = 1$) is $\lambda_{max} = a/\sqrt{2}$, which corresponds to a standing wave normal to the sides and reflected at 45° from the bottom of the triangle.

3 Discussion

It is convenient to compare the current results with those published in Ref. [9], which uses the similar methodology to solve the Schrödinger equation for quantum dots shaped as rectangular prism and a sphere. In the case of a prism with sides a, b and c, the energy spectrum is the same as that obtained in the classic approach of impenetrable walls (although the form of the wave function is different):

$$E = \frac{h^2}{8m}\left(\frac{n_x^2}{a^2}+\frac{n_y^2}{b^2}+\frac{n_z^2}{c^2}\right) \quad (9)$$

For a spherical QD with diameter a, the energy spectrum obtained with mirror boundary conditions is

$$E = \frac{h^2}{8ma^2}(2n+1)^2, \quad n = 0,1,2,... \quad (10)$$

In the classical solution for a sphere with impenetrable surface the coefficient $h^2/8ma^2$ is multiplied by squares of even integers, whereas in (10) these are odd integers.

Besides, in Ref. [12] we obtained an expression similar to (9) for the energy of a particle confined in a pyramid with the base representing an equilateral triangle with side a and height $a\sqrt{6}$ (the pyramid formed by the planes $x = 0$, $y = 0$, $z = 0$, and $x + y + z = a/\sqrt{2}$ in Cartesian coordinates):

$$E = \frac{3}{4}\frac{h^2}{ma^2}n^2 \tag{12}$$

In all cases studied the energy spectrum of a particle is proportional to the square of quantum numbers and inversely proportional to the square of the QD size; the wave functions are considerably different for each particular geometry. It appears that the position of the lowest level and the separation between the levels is larger for the case of a pyramidal QD in comparison with other geometries. To verify this, we consider a rectangular prism with a square base ($a = c$) and the height $b = a/2$. From Equation (9) it follows that

$$E = \frac{h^2}{8ma^2}(n_x^2 + 4n_y^2 + n_z^2)\,, \tag{13}$$

which corresponds to a higher energy scale, similar to the effect observed for the particle energy of a pyramidal dot. Comparing the energy expressions for different QDs, we see that the smaller volume of the dot leads to higher particle energy.

Since the formula for the energy spectra obtained with mirror boundary conditions is not different from that obtained with traditional solution methodology, one can use the classification scheme developed for quantum confinements types [13, 14]. The case of strong confinement is defined by the criterion $a/2 \ll a_B$, the Bohr radius for an exciton:

$$a_B = \frac{\hbar^2\varepsilon}{\mu e^2}\,,\ \mu = \frac{m_e m_h}{m_e + m_h} \tag{14}$$

Here $m_{e,h}$ are the effective masses for electron/hole and ε is a dielectric constant of the material. Following Refs. [13, 14], one can introduce the effective mass of the carriers m directly into the expressions for the energy spectra. We also considered the apparent "increase" of QD size due to multiple reflections from its walls. The separation between quantum levels is about $\hbar^2/ma^2$, which makes the Coulomb interaction energy $e^2/\varepsilon a$ negligible. Therefore, the energy spectrum of a particle will be defined only by the quantum confinement effect.

Among the experimental investigations of pyramidal QDs, many publications are dedicated to InAs dots with strong confinement (as it follows from their energy spectra [11, 15-21]). In particular, the influence of QD aspect ratio (relation between pyramid's height and base length) upon the energy levels was reported in [15] for InAs dots grown on GaAs substrate by MBE technique. The obtained structures were characterized with base $a = 12 \pm 1$nm in width. The height taken from cross-section TEM images is about 5 nm for the sample that we designate QD1 and 10 nm for the samples QD3. These data were used for numerical estimations with Formula (9) for QD1-type samples. For the sample QD3 one should expect smaller level separation due to the larger height of the quantum dot. The material parameters used in our calculations [22] are as follows: $m_e/m_0 = 0.023$, $m_h/m_0 = 0.42$, $\varepsilon = 12$, $E_g = 0.36$ eV. The calculated Bohr radius for an exciton is 29 nm (exceeding QD dimensions), validating the strong confinement criterion. In the effective mass approximation, one should use the expression for the shift of optical absorption relative to band gap

$$\Delta E = \hbar\omega_{01} - E_g = h^2/\mu a^2, \tag{15}$$

rendering $\Delta E = 0.952$ eV for the sample QD1. The corresponding experimental results reported in Ref. [15] are 0.865 eV for QD1 and 0.71 eV for QD3. Taking into account possible errors in estimation of the pyramids' height from the TEM data, one obtains a reasonable agreement between the experiment and theoretical predictions. It is worth noting that the confinement effect is pronounced (the difference between the exciton energy and the band gap is two – three times larger than the band gap itself), but yields smaller values than those reported in the literature.

The series of experimental data refers to inverted pyramids made of GaAs [1, 23-24] showing interconnected quantum structures (QDs, wires and wells), including a pyramid. The exact dimensions of these formations are not specified, but the pattern of inverted pyramids has a pitch of 300 to 500 nm. The pyramids have triangular base, so that the expression (12) can be used. The emission line corresponding to the ground state of an exciton at 10K has the energy of 1.553eV, confirming week confinement effect. Using the parameters of GaAs (E_g(10K) = 1.52eV and effective mass of the exciton 0.056 m_0) with formula (12), one can calculate the length of pyramid's base as $a = 52$ nm, which correlates well with the scale of the whole structure.

4 Conclusion

The mirror-type boundary conditions proved to be a simple and reliable approach in the solution of the Schrödinger equation for electrons confined in quantum dots of pyramidal shape. Our theoretical predictions were compared to experimental data, showing a reasonable agreement of the data.

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