Electronic Levels of Cubic Quantum Dots

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We introduce an efficient variational method to solve the three-dimensional Schrödinger equation for any arbitrary potential $V(x, y, z)$. The method uses a basis set of localized functions which are build up as products of one-dimensional cubic β-splines. We calculate the energy levels of GaAs/AlGaAs cubic quantum dots and make a comparison with the results from two well-known simplification schemes based on a decomposition of the full potential problem into three separate one-dimensional problems. We show that the scheme making a sequential decomposition gives eigenvalues in better agreement with the ones obtained variationally, but an exact solution is necessary when looking for highly precise values.

In recent years, the progress in growing techniques and the present degree of control of the fundamental mechanisms involved in the epitaxy of strained layers has allowed the built up of not only arrays of quantum dots but also isolated quantum dots (QDs). These three-dimensional (3D) structures are considered as "artificial atoms" whose dimension can be tailored. These "atoms" are the subject of intense research in solid state physics and device engineering [1]. They are also useful for studying fundamental problems in the physics of mesoscopic systems, like Coulomb [2] or spin blockades [3]. To develop computational schemes which can produce the electronic states in these confined systems is always an important issue since they will help to understand not only the basic mechanisms that control their energy values but also their transport or optical properties. It is clear that the external shape of the QD will determine such confinement energy. However, more important than the geometrical shape of the QD is the potential profile $V(x, y, z)$ in which such confinement is produced. To the date, the envelope function approximation within the effective mass theory has proven to be the more achievable scheme [4–6] with wide applicability, despite the inherent simplifications involved in the method. Following this scheme, we introduce a variational method that solves the 3D Schrödinger equations associated with any potential profile $V(x, y, z)$ defining a corresponding QD structure. In order to test the accuracy of our method, we have employed two different simplified approaches that can be used to reduced the full 3D problem into three one-dimensional (1D) problems when the potential has a given geometrical shape: i) the first consists of the simplest decomposition of the potential $V(x, y, z) \approx V(x) + V(y) + V(z)$ and ii) the other is a sequential decomposition of the given potential. Comparing the results obtained from our method, *i.e.*, comparing the exact solution of the Schrödinger equation to both approximations, we conclude that such a sequential decomposition is the best approach when trying to find eigenstates in very short computational times, even though a considerable deviation is present for quite small QDs.

The problem to be solved is the 3D Schrödinger equation (in a. u.):

$$-\frac{1}{2m^*}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}\right)\Psi(x, y, z) + V(x, y, z)\Psi(x, y, z) = E\Psi(x, y, z), \tag{1}$$

where m^* is the electron effective mass that, for simplicity, we assume to be homogeneous and isotropic and $V(x,y,z)$ is an arbitrary potential. The variational method expands the wavefunction Ψ as a linear combination of a finite number N of localized functions $\phi_i(x,y,z)$, each one centered at the position i of a grid of initially selected points (x_i, y_i, z_i) defining the 3D space:

$$\Psi(x,y,z) = \sum_{i=1}^{N} c_i \phi_i(x,y,z). \qquad (2)$$

Each localized function is formed as a product of one-dimensional cubic β-spline, i.e., piecewise C^2-smooth cubic polynomials [7]

$$\phi_i(x,y,z) = \beta_i(x)\beta_i(y)\beta_i(z). \qquad (3)$$

Cubic polynomials were chosen because they are the lowest-order polynomials having nonzero second-order derivatives. They have been successfully employed as a basis set in lower-order Schrödinger equations; a rich variety of 1D and 2D problems have been treated by using the corresponding variational algorithms [8–12]. Any function ϕ_i follows the usual procedure for forming a finite-element basis set. We start by introducing 3D mesh points defining I, J, and K knots in the

x, y, and z-axis, respectively, which can be uniformly distributed initially. This is not a restrictive condition, and we can always define an additional number of knots at selected positions to increase the accuracy of the method, which is important in those problems showing local abrupt changes in the potential, such as interfaces and surfaces. Along any selected direction, the corresponding 1D spline $\beta_i(x)$ is defined at a knot (x_i, y_i, z_i) and will extend from the knot (x_{i-2}, y_i, z_i) on the left up to the knot (x_{i+2}, y_i, z_i) on the right.

The procedure will generate a total basis set consisting of $N = (I-4) \times (J-4) \times (K-4)$ independent ϕ_i's that are non-orthogonal. We shall remark that the wavefunction Ψ defined in Eq. (2) is set automatically equal to zero outside the region where the mesh points are defined. By using the standard procedure, the expansion of Eq. (2) will convert the differential equation problem expressed in Eq. (1) into the following matrix problem:

$$(H_{ij} - ES_{ij})\, c_j = 0, \qquad (4)$$

where H_{ij} and S_{ij} describe all the elements of the Hamiltonian and the overlap matrices, respectively. H_{ij} are defined by the following integrals:

$$\begin{aligned} H_{ij} &= -\frac{1}{2m^*} \int_i \beta_i(x)\beta_i''(x)dx \int_i \beta_i(y)\beta_i(y)dy \int_i \beta_i(z)\beta_i(z)dz \\ &\quad -\frac{1}{2m^*} \int_i \beta_i(x)\beta_i(x)dx \int \beta_i(y)\beta_i''(y)dy \int \beta_i(z)\beta_i(z)dz \\ &\quad -\frac{1}{2m^*} \int \beta_i(x)\beta_i(x)dx \int \beta_i(y)\beta_i(y)dy \int \beta_i(z)\beta_i''(z)dz \\ &\quad +\frac{1}{2m^*} \int \int \int \beta_i(x)\beta_i(y)\beta_i(z)V(x,y,z)\beta_j(x)\beta_j(y)\beta_j(z)dxdydz, \end{aligned} \qquad (5)$$

where β'' stands for the second derivative of the 1D spline with respect to the argument inside the parenthesis. S_{ij} are given by

$$S_{ij} = \int \int \int \beta_i(x)\beta_i(y)\beta_i(z)\beta_j(x)\beta_j(y)\beta_j(z)dxdydz. \qquad (6)$$

Since every function β_i and its derivatives are polynomials, almost all the integrals above can be done exactly. The last integral in Eq. (5), which involves the potential profile, can also be calculated exactly for some analytical potentials.

The variational parameters in our method are the coefficients c_i given in Eq. (2). Exact eigenvalues of the corresponding potential problem are always lower bounds for our numerical results. This procedure does verify

the Rayleigh-Ritz variational principle: an error ε in the wavefunction implies an error ε^2 in the eigenvalues.

The matrix problem expressed in Eq. (4) is a symmetric generalized eigenvalue problem whose solutions must be obtained numerically. Nevertheless, thanks to the use of a basis set of localized functions, the matrices H_{ij} and S_{ij} are sparse; therefore, the computation time can be substantially reduced [13].

We shall describe a single QD with a prism shape as follows:

$$V(x,y,z) = V_b \Theta(x^2 - \frac{L_x^2}{4})\Theta(y^2 - \frac{L_y^2}{4})\Theta(z^2 - \frac{L_z^2}{4}). \qquad (7)$$

V_b is the barrier height and $\Theta(x)$ is the step function $[\Theta(x) = 1$ if $x \geq 0$, $\Theta(x) = 0$ if $x \geq 0]$.

We considered the case of GaAs cubic dots surrounded by an $Al_xGa_{1-x}As$ background. We used a fixed elec-
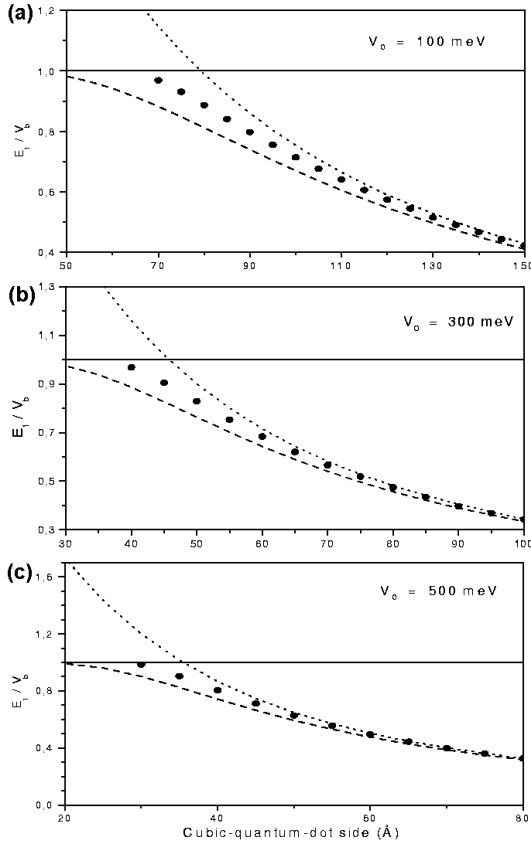
Fig. 1. Evolution of the ground state E_1 (normalized to the barrier height, V_b), vs the side of a cubic quantum dot. The symbols (full circles) represent the energies calculated variationally using the exact potential, Eq. (10). The dotted lines define the values obtained using the standard linear decomposition, Eq. (11). The dashed lines define the values resulting from a sequential decomposition, Eq. (12). The parameters employed in the calculations were $m^* = 0.067$ and (a) $V_b = 100$ meV, (b) $V_b = 300$ meV, and (c) $V_b = 500$ meV.

tron effective mass of $m^* = 0.067$, and several potential heights that simulated different aluminum concentrations. For each QD, the fundamental level E_1 was made exact up to the fourth digit by analyzing its behavior as a function of the number of basis set employed in its description.

Since the method introduced here allows us to obtain the fundamental level of the QD, we can use this value to test the accuracy of other simplifying methods based on the reduction of Eq. (1) to a 1D problem. The standard procedure splits the 3D potential in the following manner:

$$V(x,y,z) \approx V_b\Theta(x^2 - \frac{L_x^2}{4}) + V_b\Theta(y^2 - \frac{L_y^2}{4})$$
$$+ V_b\Theta(z^2 - \frac{L_z^2}{4}). \qquad (8)$$

This splitting make the corresponding Schrödinger equa-

tion separable, so the eigenenergies can be written as

$$E(n_x, n_y, n_z) = \varepsilon(n_x) + \varepsilon(n_y) + \varepsilon(n_z), \qquad (9)$$

where $\varepsilon(n)$ are the eigenvalues of a 1D quantum well with barrier V_b, whose values can be easily found [14]. The values obtained by using this approach are represented by the dotted lines in Fig. 1. In comparison with the variational calculation, this procedure is exact in the limit of infinite barriers, but it fails to predict the correct energy values as the barrier height becomes finite or as the length of the QW along any direction decreases, in which case it predicts unbounded states while the exact potential still has bounded-state solutions.

To correct the drawbacks associated with such a naive procedure, we can use, instead, a sequential decomposition based on the following picture: the origin of a level in a QD of dimension $L_x \times L_y \times L_z$ is a confined level ϵ_{ij} of a quantum-well wire of section $L_x \times L_y$, which is further confined by another QW of thickness L_z and with an effective barrier height $V_b - \epsilon_{ij}$. Moreover, the level ϵ_{ij} can be considered as originating from a bound level ϵ_i of a QW of thickness L_x whose parabolic dispersion along y is quantized by a second QW of length L_y and effective barrier $V_b - \epsilon_i$. Therefore, to calculate the fundamental level E_1, we have to proceed as follows: First, we obtain the fundamental level ϵ_1 for a QW of thickness L_x and barrier V_b. Second, the same is done to get the ground level ϵ_{11} of a QW of dimension L_y and barrier $V_b - \epsilon_1$. Third, we consider a QW of thickness L_z and barrier height $V_b - \epsilon_{11}$. If ϵ_{111} is the fundamental level of the third QW problem, the approximate solution for the full 3D problem is $E_1 = \epsilon_1 + \epsilon_{11} + \epsilon_{111}$. This solution corresponds to the following decomposition of the potential in Eq. (7):

$$V(x,y,z) \approx V_b\Theta(x^2 - \frac{L_x^2}{4}) + (V_b - \epsilon_1)\Theta(y^2 - \frac{L_y^2}{4})$$
$$+ (V_b - \epsilon_{11})\Theta(z^2 - \frac{L_z^2}{4}) \qquad (10)$$

In comparison with the standard decomposition, Eq. (8), the y and z parts of the potential are now related to each other and to the x-part through the eigenvalues ϵ_{11} and ϵ_1, respectively. The procedure applied to calculate the fundamental level can be easily extrapolated to obtain any confined level inside the QD. The values E_1 obtained with this approach are defined by the dashed lines in Fig. 1.

The comparison between the calculated ground levels allows us to conclude that this sequential decomposition has several advantages: 1) Compared with the exact treatment done by the variational method, it always produces lower values, but they can be obtained with a hand calculator; 2) compared with the widely used linear decomposition in Eq. (8) it makes good predictions about the bound character of the level inside the QD. Certainly, if the barrier height and/or the QD dimension

produce ground levels deep in the QW, any potential decomposition can be considered good. The conclusions above are very encouraging and support the use of simplified schemes for QDs with a pyramidal shape, but, unfortunately, the existence of QDs with such a shape is still controversial. However, special cases, as for example when one grows drops of InGaAs on the top of a GaAs surface [15], demand a full calculation, and the variational method presented here is in order.

In conclusion, we have introduced a variational method able to solve the 3D Schrödinger equation for QDs of any potential shape. The fundamental energies of several AlGaAs/GaAs cubic QDs have been calculated, and we have tested two different simplified schemes that can reduce the 3D QD potential problem into three 1D problems. Results for the case of cubic QDs have been compared with the values obtained variationally. We have shown that a sequential decomposition of the potential is competitive as a first approach to obtaining the true levels, but exact solutions as presented here are indispensable under certain conditions.

ACKNOWLEDGMENTS

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