Electron pair in a Gaussian confining potential

J. Adamowski, M. Sobkowicz, B. Szafran, and S. Bednarek
Faculty of Physics and Nuclear Techniques, University of Mining and Metallurgy (AGH), Kraków, Poland
(Received 10 December 1999; revised manuscript received 17 February 2000)

Two electrons confined in quantum dots (QD's) are studied under an assumption of a Gaussian confining potential and its parabolic approximation. We have calculated the energy levels of singlet and triplet states as functions of the range and depth of the confining potential in the two-dimensional (circular) and three-dimensional (spherical) QD's, and determined the critical values of these parameters, for which the electrons form bound states. The present results allow us to explain why the commonly used parabolic approximation fails in a description of transport-spectroscopy data for QD's. The applicability of the Gaussian potential to the QD's has been discussed.

Quantum dots (QD's) fabricated by modern semiconductor nanotechnology[1] possess various shapes. Semiconductor nanocrystals with nearly spherical boundaries,[2] embedded in insulating materials, are examples of spherical QD's. The vertical gated nanodevice[3] consists of the cylindrical QD, which is treated by some authors[4,5] as a two-dimensional (2D) circular nanostructure. Excess electrons confined in nanometer-sized QD's can form bound atomiclike states. The confining potential, which results from a conduction-band discontinuity at the QD boundary and external voltages applied to the nanostructure, is usually approximated by a three-dimensional (3D) potential well (for spherical QD's)[6,7] and a two-dimensional parabolic potential (for cylindrical QD's).[4,5,8–11] Dipole optical transitions for the electrons confined in the parabolic potential are independent of the number of electrons. This property, known as a generalized Kohn's theorem,[12] was found to be approximately fulfilled in $GaAs/Al_xGa_{1-x}As$ (Ref. 13) and $In_xGa_{1-x}As/GaAs/AlAs$ (Ref. 14) QD's. However, recent experimental data[15] show pronounced deviations from the predictions of this theorem. The parabolic potential possesses infinite depth and range. Therefore, it is inappropriate for a description of the experimentally measured charging of the QD by the finite number of excess electrons.[16] These experimental results suggest that the real confining potential is nonparabolic and possesses a well-like shape. The charging of QD's was qualitatively described[6,7] with the use of a 3D spherical rectangular potential well of finite depth. The application of the 3D cylindrical potential well allowed us[17] to obtain a quantitative agreement with the capacitance-spectroscopy data for self-assembled QD's.[16] It was also shown[17] that a consistent description of these data is not possible under the assumption of parabolic confinement.

In the present paper, we propose a Gaussian attractive confining potential to study the properties of excess electrons in the QD's. This potential possesses the finite depth and range and—in the vicinity of the dot center—can be approximated by the parabolic potential, i.e., it can account for the charging of the QD by the finite number of excess electrons and the approximate validity of the generalized Kohn's theorem. Anharmonic corrections to the parabolic approximation are responsible for the observed deviations[15] from this theorem. Contrary to the rectangular potential well, the Gaussian potential well is smooth at the QD boundaries, which permits us to model a compositional modulation within the QD's.[18]

The Gaussian potential was used in nuclear physics for a description of scattering of complex nuclei.[19] The one-electron eigenvalue problem with the attractive Gaussian potential does not admit analytical solutions. Different approximate methods[20–23] were implemented to solve this problem. In the present paper, we have applied a variational method to the eigenvalue problems for both the one- and two-electron systems in the 2D (circular) and 3D (spherical) attractive Gaussian potentials.

We consider the electrons in the QD with the confining potential

$$V(r) = -V_0 \exp(-r^2/2R^2), \tag{1}$$

where the depth of the potential well $V_0 > 0$, R is the range of the confinement potential, which corresponds to a radius of the QD, $r = |\mathbf{r}|$, where $\mathbf{r} = (x,y)$ for the 2D system, and $\mathbf{r} = (x,y,z)$ for the 3D system. For $r/R \ll 1$, Gaussian potential (1) can be approximated by the harmonic-oscillator (parabolic) potential

$$\tilde{V}(r) = -V_0 + \gamma^2 r^2, \tag{2}$$

where $\gamma^2 = V_0/2R^2$. Throughout the present paper, we use the donor Rydberg $R_D = m_e^\star \mathrm{Ry}/\varepsilon^2$ as a unit of energy, and the donor Bohr radius $a_D = \varepsilon a_B / m_e^\star$ as a unit of length, where Ry is the atomic Rydberg, a_B is the atomic Bohr radius, $m_e^\star = m_e/m_{e0}$ is the ratio of the electron conduction-band mass m_e to the electron rest mass m_{e0}, and ε is the effective dielectric constant of the QD. The one-electron energy spectrum for parabolic potential (2) is given by

$$E_{nm}^{2D} = -V_0 + 2\gamma(2n + |m| + 1) \tag{3}$$

for the 2D case, and

$$E_{nl}^{3D} = -V_0 + \gamma(4n + 2l + 3) \tag{4}$$

for the 3D case, where $n, l = 0,1,\ldots$, and $m = 0, \pm 1, \ldots$.

For the single electron confined in potential (1) we have solved the Schrödinger equation by the variational method with the trial wave function expanded in the basis $r^p \exp(-\alpha_i r^2)$, where α_i are the variational parameters. The

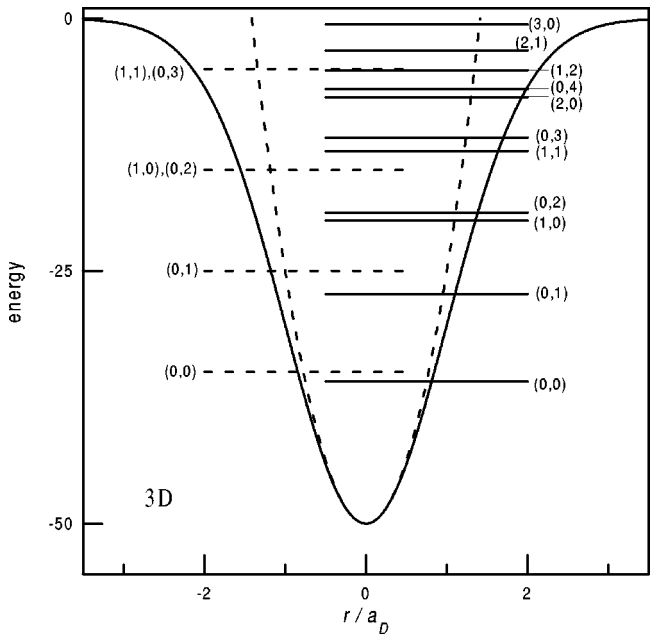

FIG. 1. One-electron energy levels for the Gaussian (solid lines) and harmonic-oscillator (dashed lines) 3D potential with $V_0 = 50R_D$ and $R = a_D$. The energy levels are labelled by quantum numbers (n,l). The shapes of both the potential energies are also shown as functions of distance r from the QD center. Energy is expressed in donor Rydbergs R_D and length in donor Bohr radii a_D.

parameter p is taken on as follows: $p = |m|$ for the 2D system and $p = 0$ for the 3D system. We have performed the calculations with the 20-element basis and checked that our results agree very well with those given in Refs. 20–23. The one-electron energy levels calculated for the 3D system are displayed in Fig. 1 and compared with those for the parabolic potential. We note that only for the ground state the parabolic potential can be regarded as a fairly good approximation of the nonparabolic Gaussian potential. The excited-state energy levels for the parabolic potential exhibit large deviations from those for the Gaussian potential, which are both qualitative (degeneracy of states) and quantitative (considerable shifts of levels).

It is well known that the eigenvalue problem for two electrons in the parabolic potential can be separated into center-of-mass and relative coordinates, which leads to the simple Schrödinger equation for the relative motion.[8–10,24–26] For certain values of γ, this equation can even be solved analytically.[24] For arbitrary γ, accurate numerical methods are available,[8–10,26] which allow us to obtain exact solutions to the relative-motion eigenvalue problem. For the Gaussian confinement potential, the two-electron eigenvalue problem is not separable. In order to solve the corresponding Schrödinger equation, we have proposed the following variational trial wave function:

$$\Psi(r_1, r_2, r_{12}) = (1 \pm P_{12}) \sum_{i=1}^{N_B} c_i \exp(-\alpha_i r_1^2 - \beta_i r_2^2 - \gamma_i r_{12}^2), \tag{5}$$

where r_1 and r_2 are the electron-dot center separations, r_{12} is the electron-electron distance, sign $+(-)$ corresponds to the singlet (triplet) states, P_{12} is the permutation operator interchanging the indices 1 and 2, c_i are the linear variational parameters, α_i, β_i, and γ_i are the nonlinear variational parameters, which have been chosen according to geometrical progressions. Two-electron trial wave function (5) explicitly includes the electron-electron correlation via the r_{12}-dependent terms. We have performed test calculations with the increasing number N_B of basis elements in trial wave function (5) and checked that taking on $N_B = 55$ is sufficient to achieve the required accuracy. For example, the differences between the results obtained with $N_B = 55$ and $N_B = 190$ do not exceed $10^{-3} R_D$. Trial wave function (5) has been also tested for two electrons in the harmonic-oscillator potential and a very good agreement has been found with the exact results[24,26] for both the singlet and triplet states.

Figures 2(a) and 2(b) show the results obtained for the 2D and 3D few-electron systems confined in the Gaussian and parabolic potentials. The singlet and triplet two-electron states with the total angular momentum $L = 0$ (S states) are denoted by ^1S and ^3S, respectively. Figures 2(a) and 2(b) as well display the lowest one-electron energy levels for s-like states, i.e., the states with $m = 0$ for 2D and $l = 0$ for 3D systems. The potential-well depth $V_0 = 50R_D$ corresponds to the GaAs QD's in the $Al_{0.3}Ga_{0.7}As$ matrix, for which $R_D = 6$ meV and $a_D = 10$ nm. The plots in Figs. 2 correspond to the QD's with the strong and intermediate confinement, i.e., $R \leqslant 2a_D$. The condition of binding of N-electron quantum state ν with energy $E_\nu(N)$ has the form[6,7]

$$E_\nu(N) < E_{th}(N). \tag{6}$$

The continuum-threshold energy $E_{th}(N)$ for the N-electron system is determined as follows:[6,7] $E_{th}(N) = E_0(N-1)$ and $E_{th}(1) = 0$, where $E_0(N-1)$ is the ground-state energy of the $(N-1)$-electron system. For the harmonic-oscillator potential given by Eq. (2), we have introduced the artificial continuum threshold (cf. Fig. 1), corresponding to $\tilde{V}(r) = 0$, in order to compare the results for both the potentials. Figures 2(a) and 2(b) show that the qualitative properties of energy levels for the 2D and 3D systems are similar. However, the quantitative differences are also visible: all the energy levels for the 2D system are located slightly below the corresponding levels for the 3D system. This effect results from the enhanced effective confinement of electrons in the 2D nanostructure. We note that in both the 2D and 3D systems the energy levels for the Gaussian confining potential are considerably shifted downwards with respect to the corresponding levels for the parabolic confinement. The calculations performed in the weak-confinement regime show that the ground-state energy levels for both the potentials start to coincide with each other for $R \gg a_D$ [cf. the (0,0) and ^1S levels at $R \simeq 2a_D$]. However, the excited-state energy levels are still different even for large QD's.

Based on the present results, we can speculate about expected properties of many-electron systems in QD's with nonparabolic confinement. The many-electron quantum states are mainly built from the excited one-electron states, for which the energy levels for the Gaussian and parabolic potential essentially differ between themselves (cf. Fig. 1). This will cause large deviations of the results obtained in the

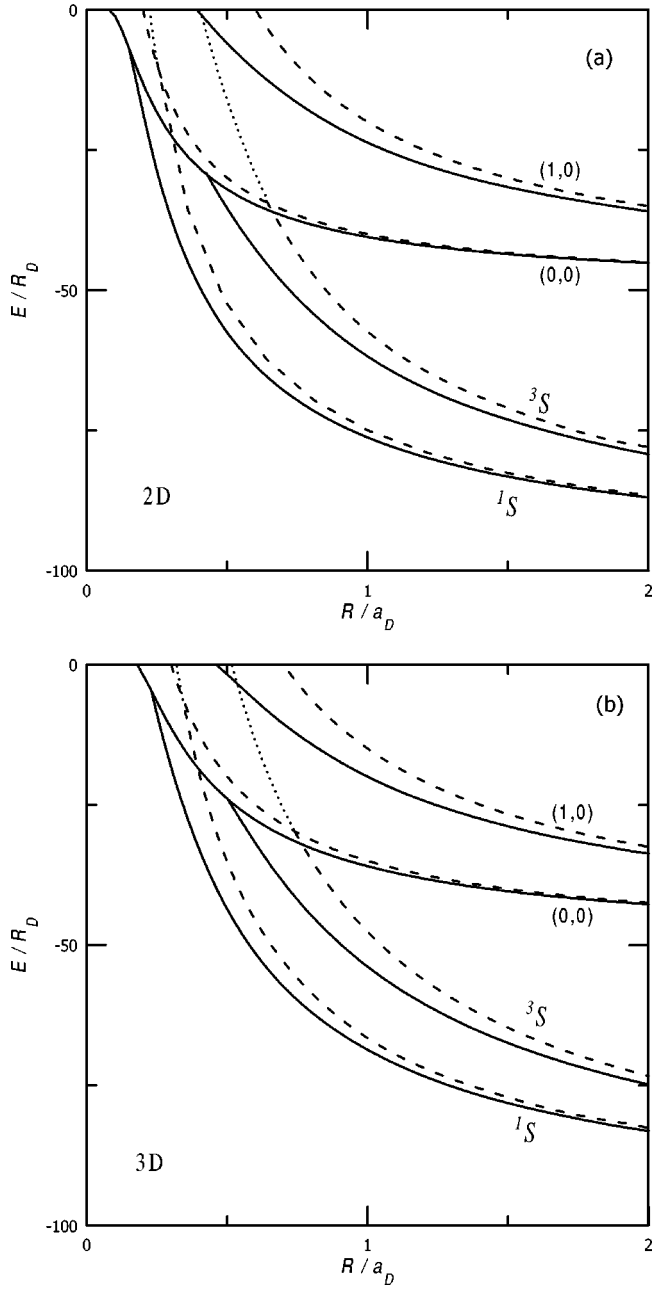

FIG. 2. Energy levels of singlet (^1S) and triplet (^3S) states of two electrons confined in Gaussian (solid curves) and harmonic-oscillator (dashed curves) potential as functions of range R for the 2D (a) and 3D (b) systems. One-electron energy levels (0,0) and (1,0) are also shown for both potentials. Dotted curves correspond to the two-electron states, which are bound in the parabolic potential and possess higher energies than the one-electron ground state. The units are the same as in Fig. 1.

frame of the harmonic-oscillator model from those for the nonparabolic potential. The largest differences will appear for the weakly bound many-electron states (with energies near the continuum threshold), which determine the upper limit for the filling of the QD by the excess electrons. This effect is already visible for the two-electron triplet excited state [Figs. 2(a) and 2(b)], for which the difference between the energy levels for the Gaussian and parabolic potentials is considerably larger than that for the singlet ground state and remains noticeable up to $R \simeq 10a_D$. The common usage of

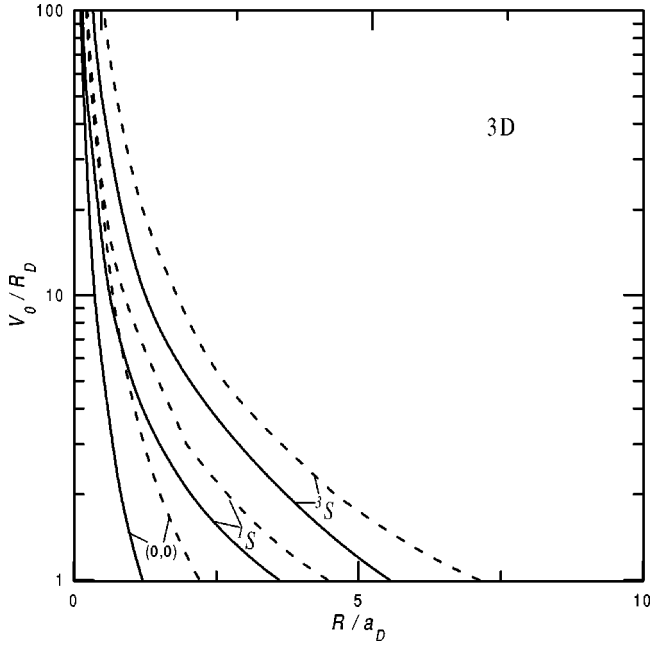

FIG. 3. ''Phase diagrams'' on the $R-V_0$ plane for electrons in 3D QD's. Solid (dashed) curves display the critical values of the QD parameters, above which the corresponding quantum state is bound for the Gaussian (parabolic) confining potential. Solid curves correspond to the conditions at which the electrons can tunnel through the QD. The units are the same as in Fig. 1.

the parabolic confining potential[4,5,8–11] is based on the assumption that—in the vicinity of the dot center—the real confining potential can be approximated by a quadratic function of electron-dot center distance. The present results show that this approximation can only be justified for the QD's with the confined-electron energy levels, which lie sufficiently deeply below the continuum-threshold energy. Nevertheless, in transport experiments with QD's,[3,16] the major contributions originate from the electron states with the energies near the continuum threshold, for which the parabolic approximation fails.

Condition (6) allows us to determine critical values of QD parameters (R and V_0), above which the excess electrons form atomiclike bound states. We have performed the calculations for different values of R and V_0 and established ''phase transitions'' for the few-electron systems. The results for the 3D QD's are reported in Fig. 3. These ''phase transitions'' can be interpreted as follows: if the values of R and V_0 exceed those depicted by solid curves, then the quantum capacity[6] of the QD is sufficiently large in order for the corresponding N-electron quantum state to be bound. The critical values of R and V_0 for the electron binding increase with the increasing number of electrons. Therefore, the curves in Fig. 3 also provide lower bounds on the critical values of the QD parameters for the binding of N-electron states with $N>2$. The results shown in Fig. 3 can be useful in determining whether or not the QD, characterized by the certain parameters, can be charged by electrons. The transport experiments[3] are performed with entirely filled QD's,[27] which correspond to the solid curves in Fig. 3, for which the differences between the Gaussian and harmonic-oscillator potential are large. We note that for the commonly used parabolic potential of the form $\gamma^2 r^2$, all the N-electron states

are bound and the dashed curves, which result from the artificially introduced continuum-energy threshold, do not appear.

The present results are qualitatively similar to those obtained in our previous paper[6] for few-electron systems in the finite rectangular potential well of spherical symmetry. This means that the depth and range of the potential well are the most important parameters characterizing the model confining potential. We note, however, that the Gaussian potential does not exhibit the discontinuity at the QD boundary, which is the undesirable property of the rectangular potential, since it leads to the discontinuity of the second derivative of the wave function. Due to its continuity and finite depth and range, the Gaussian potential fairly well approximates the real confinement potential in QD's. In particular, the Gaussian confinement potential can be applied to the gate-controlled QD's,[3] for which the real confinement potential calculated[27] from the Poisson equation exhibits the strong nonparabolicity, and to the self-organized QD's with the compositional modulation.[18] The softness of the Gaussian potential enables us to model the slowly varying confining potential, which results from a nonabrupt interface between the QD and matrix regions.[18] Therefore, the attractive Gaussian potential is sufficiently flexible and possesses all the required properties to be applicable as a realistic confining potential in the QD's.

In summary, we have applied the Gaussian confining potential to a description of few-electron states in semiconductor QD's. We have determined the energy levels of the singlet and triplet states as functions of the potential depth and range for the 2D and 3D systems, and determined the critical values of the QD parameters for the binding of the few-electron states. The same calculations performed with the parabolic approximation of the Gaussian potential lead to the results, which are qualitatively and quantitatively different. The results of the present paper allow us to recommend the Gaussian potential as a good approximation of the real confining potential, which should be especially useful for a description of many-electron systems in self-organized QD's with a varying composition.

[1] *Nanotechnology*, edited by G. Timp (Springer-Verlag, New York, 1999).

[2] A.P. Alivisatos, J. Phys. Chem. **100**, 13 226 (1996).

[3] S. Tarucha, D.G. Austing, T. Honda, R.J. van der Hage, and L.P. Kouwenhoven, Phys. Rev. Lett. **77**, 3613 (1996).

[4] M. Eto, Jpn. J. Appl. Phys. **36**, 3924 (1997).

[5] O. Steffens, M. Suhrke, and U. Rössler, Physica B **256-258**, 147 (1998).

[6] B. Szafran, J. Adamowski, and S. Bednarek, Physica E (Amsterdam) **4**, 1 (1999).

[7] S. Bednarek, B. Szafran, and J. Adamowski, Phys. Rev. B **59**, 13 036 (1999).

[8] U. Merkt, J. Huser, and M. Wagner, Phys. Rev. B **43**, 7320 (1991).

[9] D. Pfannkuche, V. Gudmundsson, and P.A. Maksym, Phys. Rev. B **47**, 2244 (1993).

[10] J.-L. Zhu, Z.-Q. Li, J.-Z. Yu, K. Ohno, and Y. Kawazoe, Phys. Rev. B **55**, 15 819 (1997).

[11] R.M.G. Garcia-Castelan, W.S. Choe, and Y.C. Lee, Phys. Rev. B **57**, 9792 (1998).

[12] P.A. Maksym and T. Chakraborty, Phys. Rev. Lett. **65**, 108 (1990).

[13] R.C. Ashoori, H.L. Stormer, J.S. Weiner, L.N. Pfeiffer, S.J. Pearton, K.W. Baldwin, and K.W. West, Phys. Rev. Lett. **68**, 3088 (1992).

[14] H. Drexler, D. Leonard, W. Hansen, J.P. Kotthaus, and P.M. Petroff, Phys. Rev. Lett. **73**, 2252 (1994).

[15] D. Heitman, K. Bollweg, V. Gudmundsson, T. Kurth, and S.P. Riege, Physica E (Amsterdam) **1**, 204 (1997).

[16] B.T. Miller, W. Hansen, S. Manus, R.J. Luyken, A. Lorke, J.P. Kotthaus, S. Huant, G. Medeiros-Ribeiro, and P.M. Petroff, Phys. Rev. B **56**, 6764 (1997).

[17] B. Szafran, J. Adamowski, and S. Bednarek, Phys. Rev. B **61**, 1971 (2000).

[18] P.D. Siverns, S. Malik, G. McPherson, D. Childs, C. Roberts, R. Murray, B.A. Joyce, and H. Davock, Phys. Rev. B **58**, R10 127 (1998).

[19] B. Buck, H. Friedrich, and C. Wheatley, Nucl. Phys. A **275**, 246 (1977).

[20] N. Bessis, G. Bessis, and B. Joulakian, J. Phys. A **15**, 3679 (1982).

[21] C.S. Lai, J. Phys. A **16**, L181 (1983).

[22] R.E. Crandall, J. Phys. A **16**, L395 (1983).

[23] M. Cohen, J. Phys. A **17**, L101 (1984).

[24] M. Taut, Phys. Rev. A **48**, 3561 (1993).

[25] M. Taut, A. Ernst, and H. Eschrig, J. Phys. B **31**, 2689 (1998).

[26] B. Szafran, J. Adamowski, and S. Bednarek, Physica E (Amsterdam) **5**, 185 (2000).

[27] S. Bednarek, B. Szafran, and J. Adamowski, Phys. Rev. B **61**, 4461 (2000).