# Electronic Structure of Quantum Dots

C. Hines, S.A. McCarthy, J.B. Wang, and P.C. Abbott

Department of Physics, The University of Western Australia
Perth, WA 6009, Australia, wang@physics.uwa.edu.au

## ABSTRACT

This paper presents a detailed calculation of the electronic structure of quantum dots with various geometries. In particular, non-circular quantum dots are examined and their characteristic properties analysed. In addition, the importance of electron-electron spin exchange and correlation is addressed.

*Keywords*: quantum dot, electronic structure, eigenvalue problem, Hartree-Fock method

## 1 INTRODUCTION

Quantum dots, also known as artificial atoms, are so termed because like real atoms they confine the motion of an electron in space resulting in a discrete energy spectrum. However unlike their naturally occurring brethren, artificial atoms can be manufactured in such a way to exhibit a precise control over this confinement, which has opened up a wide range of possibilities and areas for examination (Kouwenhoven et al 2001). For example, they are not limited to being spherically or circularly symmetric – we can have elliptic dots, rectangular dots, triangular dots, and even dots without any symmetry. Transitions never observed in the spectra of natural atoms can be obtained from the artificial ones. In the future, quantum dots may be used to build more efficient and precisely controlled lasers with otherwise inaccessible wavelengths, and also as vital components of nanoelectronic devices (Reed 1993, Gammon 2000). It is also hoped that quantum dots may one day be able to help realise the dream of quantum computing (DiVincenzo et al. 2000).

This field of study has emerged from recent advances of nano fabrication technology, and thus only very limited theoretical work has been done. Pfannkuche et al. (1993) performed Hartree-Fock self-consistent calculations for a two-electron quantum dot and compared with results obtained from direct numerical diagonalisation of the two-particle Hamiltonian. They obtained good agreement between these two calculations for the triplet state, but markedly different results for the singlet state. This indicates that spin correlation was not properly built in their Hartree-Fock model. Ezaki et al. (1998) applies a brute force approach by numerically diagonalising the *N*-electron Hamiltonian, using Slater determinants composed of 26 single-electron eigenstates. However, such an approach involves the calculation of a very large number of integrals and the inversion of large matrixes, and thus requires considerable numerical resources. Macucci et al. (1997) studied quantum dots with up to 24 electrons using a mean-field local-density-functional approach, in which the exchange and correlation potential was approximated by an empirical polynomial expression. Lee et al. (1998) also studied an N-electron quantum dot using the density functional theory, where the generalized gradient approximation was used for exchange correlation potentials. Exchange interaction comes directly from the anti-symmetrisation of wavefunctions as required by the Pauli exclusion principle. In the density functional theory, this is a major problem since the mathematical object is electron density rather than electron wavefunction, while the exchange interactions can be readily built in the Hartree-Fock method that deals with wavefunctions directly.

In the following sections, we will first look at a single electron in quantum dots of various geometries. The electronic structure of multi electrons will then be examined through the Hartree-Fock formalism.

## 2 SINGLE ELECTRON SYSTEMS

In our model of quantum dot systems, we assume that the confining potential can be separated into a vertical ($z$) component and a lateral $\vec{r} = (x, y)$ component. The confining potential in the vertical direction can be thought of as effectively being a very narrow triangular well, whereas the lateral confining potential $V(\vec{r})$ can be made arbitrarily complex. The energy level of the first excited state in the z direction is generally hundreds of times greater than many of the low energy states in the *x-y* plane. This property allows us to model electron motion in a quantum dot as two-dimensional. The corresponding Schrodinger equation reads

$$\left(-\frac{\hbar^2}{2m^*}\nabla^2 + V(x,y)\right)\psi(x,y) = E\psi(x,y), \qquad (1)$$

where $m^*$ is the effective mass and $\nabla^2$ is the two-dimentional Laplacian.

If we map the wavefunction $\psi(x,y)$ as well as the kinetic and potential energy operator onto a numerical grid, Eq. (1) becomes a matrix equation, which can be solved by

several well-established methods (see, for example, Saad, 1992). In this work, we used the ARPACK library (http://www.caam.rice.edu/software/ARPACK), which was designed to solve large scale eigenvalue problems.

The ARPACK library is so named because of its use of an Implicitly Restarted Arnoldi method. Using this technique, a set of Schur vectors are calculated which give rise to approximate eigenvalues and eigenvectors of the original matrix. The library is written in such a way that when the main routine is called, it returns the user with a vector. The user is requested to multiply the vector by the matrix for which the eigenvalues are required and then re-call the same routine. This process continues until convergence is achieved. When the library determines that a reasonable set of Schur vectors have been calculated, a separate routine is used to calculate the eigenvalues and eigenvectors from the previous iterative refinement.

The ARPACK library is most suitable for use on "sparse" matrices, where the definition of "sparse" is such that the multiplication of a vector by the matrix is an order $O(n)$ operation. That is, if the vector has $n$ elements, and the matrix has $n^2$ elements, most of the elements of the matrix are zero so that the number of multiplications and additions required to multiply the vector by the matrix is only proportional to $n$ (multiplying a dense matrix with $n^2$ non-zero elements is an order $O(n^2)$ process). Thus the time required to complete the calculation is proportional to the length of the eigenvectors and the number of eigenvectors required. Therefore, the ARPACK library is most advantageous when only the first few eigenvectors are required, as is the case, for example, when computing the first few eigenfunctions of a Hamiltonian.

We studied four different quantum dot systems (elliptic, triangle, square and annular ring). Their energy levels are listed in Table 1. The first panel of Figures (1-4) shows the confinement potential of the dot, and the other panels illustrate the wavefunctions of the first five eigen-states of the system.

Table 1: Energy levels (a.u.)

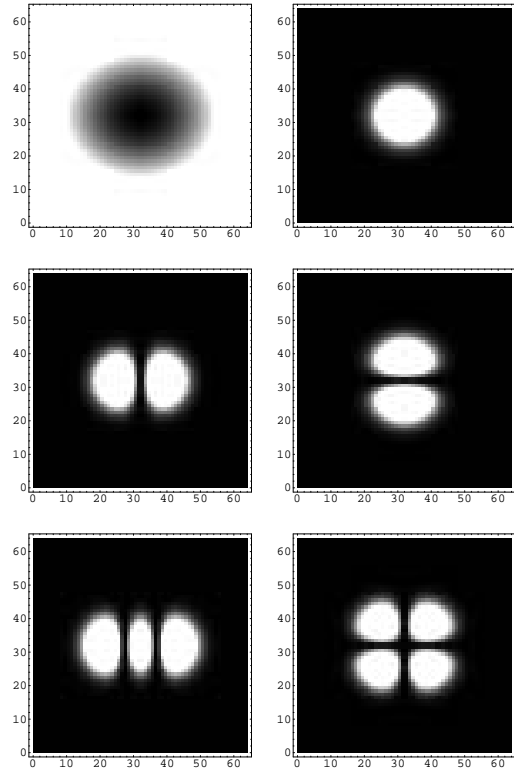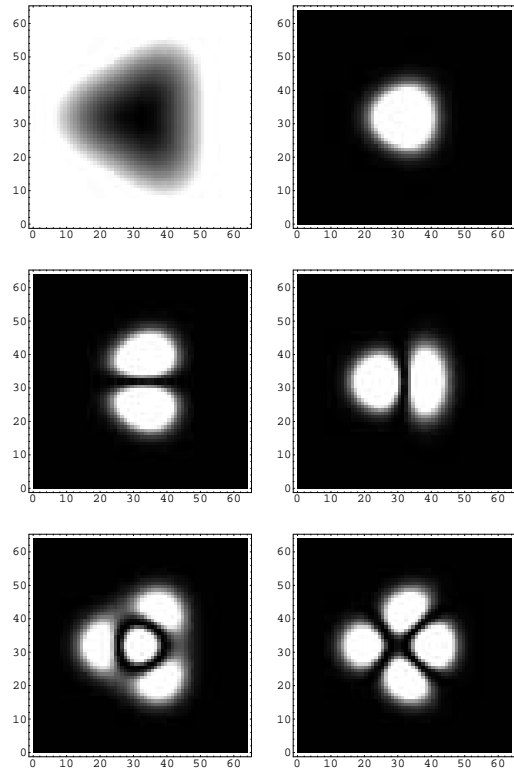|   | Elliptic | Triangle | Square | Ring |
|---|----------|----------|--------|------|
| 1 | 1.1 | 0.99 | 0.99 | 1.00 |
| 2 | 2.1 | 1.96 | 1.97 | 1.12 |
| 3 | 2.3 | 1.96 | 1.97 | 1.12 |
| 4 | 3.1 | 2.88 | 2.76 | 1.45 |
| 5 | 3.3 | 2.97 | 2.93 | 1.47 |

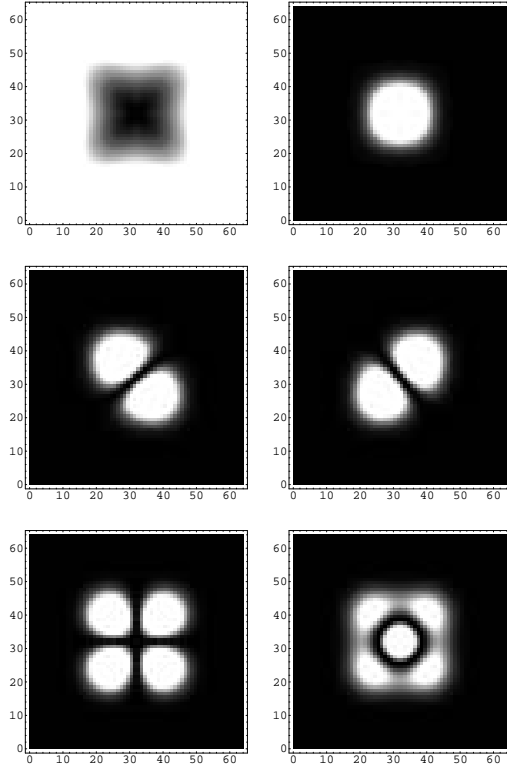

Figure 1. An elliptic QD



Figure 2. A triangular QD

Figure 3. A square QD

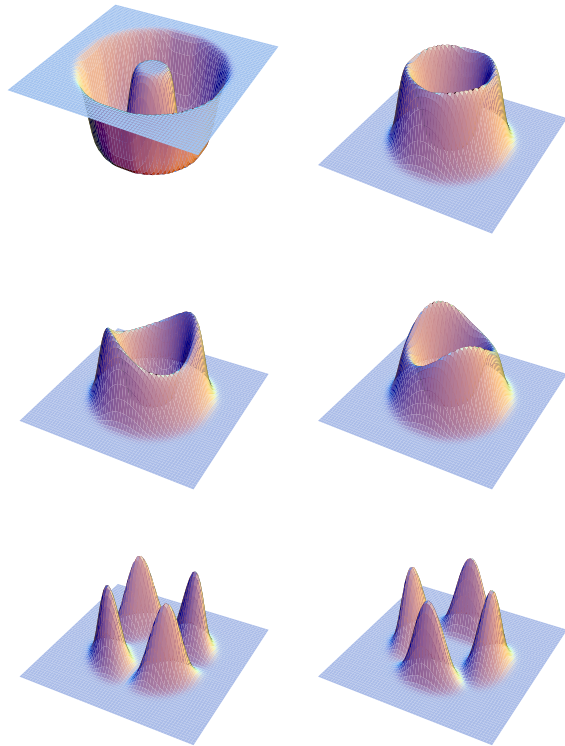

Figure 4. An annular ring QD

# 3 N-ELECTRON SYSTEMS

We now turn to the problem of an N-electron quantum dot system, for which the Schrödinger equation is

$$\left[\sum_{i=1}^{N}\left(-\frac{\hbar^2}{2m^*}\nabla_{r_i}^2 + V(\mathbf{r}_i)\right) + \frac{e^2}{4\pi\varepsilon}\sum_{i>j=1}^{N}\frac{1}{r_{ij}}\right]\psi(q_1,q_1,...q_N) = E\psi(q_1,q_1,...q_N),$$

(2)

where $q_i$ represents collectively both the spatial and the spin coordinate of the $i^{th}$ electron.

To solve the above equation we employ the Hartree-Fock method. The Hartree-Fock approach is a particular case of the variational method, in which the trial wavefunction is assumed to be a Slater determinant,

$$\Psi(q_1, q_2, ...q_N) = \frac{1}{\sqrt{N!}}\begin{vmatrix} \psi_a(q_1) & \psi_b(q_1) & \cdots & \psi_n(q_1) \\ \psi_a(q_2) & \psi_b(q_2) & \cdots & \psi_n(q_2) \\ & & \prod & \\ \psi_a(q_N) & \psi_b(q_N) & \cdots & \psi_n(q_N) \end{vmatrix},$$

(3)

where $\psi_l(q_i)$ are the individual electron spin-orbitals.

The basic assumption in the Hartree-Fock approach is that each electron moves in the time-averaged charge distribution due to all the other electrons. From Eqs. (2) and (3), the Hartree-Fock equation describing such an electron can be readily derived as

$$\left[-\frac{1}{2}\nabla_{r_i}^2 + V(q_i) + V_l^d(q_i) - V_l^{ex}(q_i)\right]\psi_l(q_i) = E_l\,\psi_l(q_i),$$

(4)

with

$$V_l^d(q_i) = \sum_{m\alpha l} V_m^d(q_i) = \sum_{m\alpha l} \left\langle \frac{\psi_m(q_j)}{r_{ij}} \right\rangle,$$

$$V_l^{ex}(q_i)\psi_l(q_i) = \sum_{m\alpha l} \delta_{m_s^m, m_s^l}\left\langle \frac{\psi_m(q_j)\psi_l(q_i)}{r_{ij}} \right\rangle,$$

where $\chi_{1/2,m_s^\mu}$ is the spin part of the wavefunction, the direct potential $V_l^d(q_i)$ represents the average Coulomb interaction due to the $N-1$ electrons, and the exchange potential $V_l^{ex}(q_i)$ represents an exchange term due to Pauli's exclusion principle. A self-consistent procedure is applied, in which an initial guess is made for the wavefunctions of the $N-1$ electrons and Eq. (4) is solved for the $N^{th}$ electron. This wavefunction is then used as an improved guess for that electron in the calculation of the wavefunction of another electron. Self-consistency is obtained by repeating this process iteratively until no further change of the wavefunctions.

We have carried out such a calculation for a circularly symmetric quantum dot with up to $n=18$ electrons. Calculations for other geometries are underway and will be reported at the conference. Our results for the circularly symmetric quantum dot are presented in Figures (5-7). The addition energy is defined as

$$\Delta\mu(N) = E(N+1) - 2E(N) + E(N-1), \qquad (5)$$

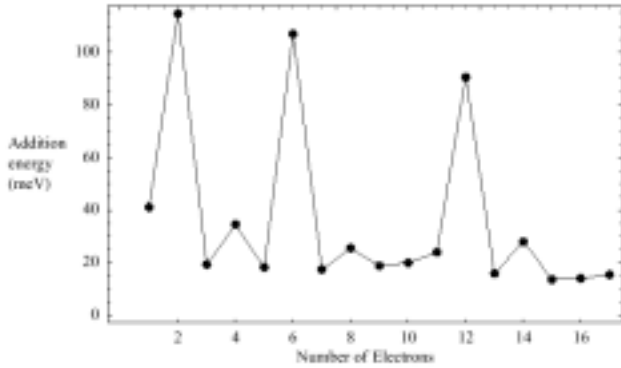where $E(N)$ is the ground state energy for a $N$-electron system.



Figure 5. Addition energies for a circularly symmetric QD with confinement potential $V(x,y) = k^2(x^2 + y^2)/2$.
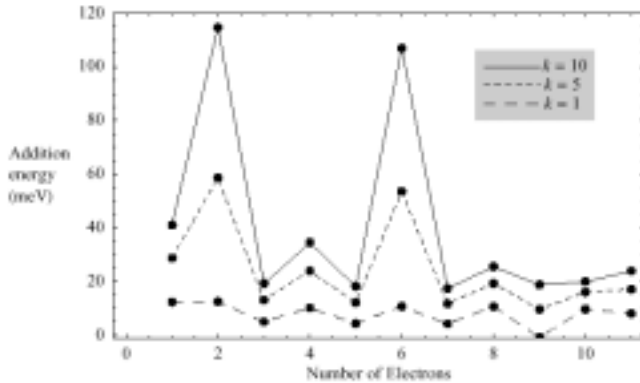
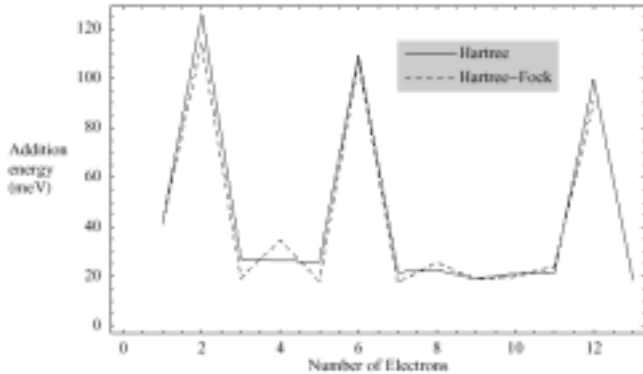

Figure 6. Addition energies for different $k$ values.



Figure 7. Comparison of a Hartree (without exchange) and a Hartree-Fock (with exchange) calculation for $k = 10$.

Tarucha et al (1996) were able to experimentally measure the addition energy of a quantum dot structure. As a negative voltage is applied to the side gate, the diameter of the dot becomes smaller and excess electrons are forced out one at a time until there are none left in the conduction band. A current will flow only if the number of electrons in the dot changes. This will only happen when certain energies are supplied, and so current peaks are observed at the corresponding voltages. The voltage difference between current peaks is a measure of the addition energy, which is shown in Figure 8. Note that the experimental peaks tend to fall off more rapidly than the theoretical results probably due to the fact that the calculations are performed for fixed $k$, whereas the experimental technique of slowly altering the voltage to remove the electrons would scan through a range of $k$ values.
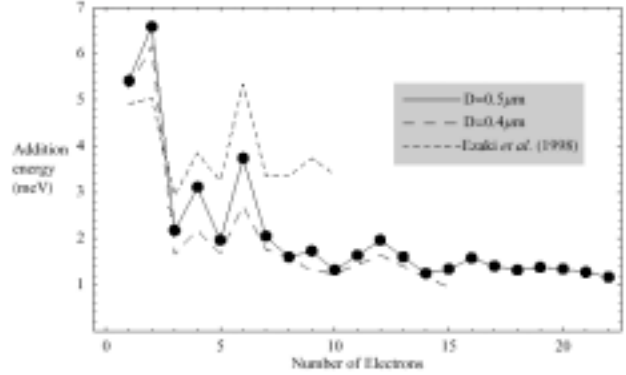


Figure 8. Addition energies measured by Tarucha *et al* (1996) and calculated by Ezaki *et al* (1998).

## 4   CONCLUSION

In this work we have examined the electronic structure of quantum dots with various geometries. As a first step, we investigated the single electron solutions to the Schrödinger equation. A general method was developed which allows us to treat a wide range of different confining potentials. We also studied a circularly symmetric dot with up to 18 electrons. Our results are in good agreement with the theoretical work by Lee *et al* (1998) and also compares well with the experimental results of Tarucha *et al* (1996). The spin exchange interaction is found to be responsible for the secondary peaks in the addition energy spectrum.

## REFERENCES

DiVincenzo DP. Bacon D. Kempe J. Burkard G. Whaley KB. 2000, Nature 408, 339-342

Ezaki T. Sugimoto Y. Mori N. Hamaguchi C. 1998, Semicond. Sci. Technol. 13, A1–A3.

Gammon D. 2000, Nature 405, 899-900

Kouwenhoven LP. Austing DG. Tarucha S. 2001, Rep. Prog. Phys. 64, 701-736

Lee IH. Rao V. Martin RM. Leburton JP. 1998, Phys. Rev. B 57, 9035-9042

Lehoucq RB. and Sorensen DC. 1996, SIAM J. Matrix Analysis and applications 17, 789-821

Macucci M. Hess K. Iafrate GJ. 1997, Phys. Rev. B 55, R4879-R4882

Pfannkuche D. Gudmundsson V. Maksym P. 1993, Phys. Rev. B 47, 2244-2250

Reed M.A. 1993, Scientific American, January, 98-102

Saad Y. 1992, *Numerical methods for large eigenvalue problems*, Halsted Press

Tarucha S. et al. 1996 Phys. Rev. Lett. 77, 3613-3616