Electronic structure of artificial atoms

Jingbo Wang and Chris Hines
School of Physics, The University of Western Australia, Australia
E-mail: wang@physics.uwa.edu.au

Abstract. Quantum dots are artificially fabricated “atoms and molecules”, in which charge carriers are confined in all three dimensions similar to the electrons in real atoms and molecules. Consequently, they exhibit similar properties normally associated with real atoms and molecules, such as quantized energy levels and shell structures. The detailed electronic structure of quantum dot systems depends on many of their physical properties such as material, size and geometry. Therefore, they can be readily engineered and manipulated to test fundamental concepts in quantum mechanics, to build more efficient and precisely controlled lasers and electronic devices, and to process vast amount of quantum information. This opens up a wide range of possibilities and areas for exploring new physics and new applications. In this paper, we present \textit{ab initio} calculations on quantum dot systems by solving the corresponding Schrödinger's equations.

1. Introduction

Quantum mechanics is one of the most successful theories ever developed. It has provided extraordinarily accurate predictions of many physical systems, with a wide range of practical applications such as lasers and electronic devices. However the scalability of quantum calculations and the sheer resources necessary to make an accurate practical calculation limits the ability to study nano-structured devices of reasonable size using first principles of quantum mechanics.

This paper is concerned with approximations and computational schemes developed in order to perform quantum mechanical calculations with limited resources. In particular, we will describe an iterative refinement configuration interaction method (IRCI) and a spin adapted configuration interaction method (SACI). Both methods take into account full electron and spin correlation effects, giving a level of accuracy not available in alternative approaches such as the density functional theory or the self-consistent Hartree-Fock method. They are also advantageous over the standard configuration interaction method, in that a much smaller basis is required by choosing the most appropriate set of Slater determinants through an iterative process and by using spin eigenfunctions already possessing the Fermionic symmetry.

2. Theory and Computational Schemes
The Hartree-Fock method and the density functional theory are both self-consistent mean-field models, although they are fundamentally different in the way of treating exchange and correlation effects [1,2]. In the Hartree-Fock method, exchange is considered exactly by the proper anti-symmetrization of wavefunctions using a Slater determinant, while correlation in the motion of the electrons is neglected since only a time-averaged effective potential is used in this formulation. In the density functional theory, both exchange and correlation effects are included exactly in principle, but only approximately in practice in most situations. Perhaps worse still, there is no clear route for the density functional theory to provide convergent results.

A more accurate formalism is to use a linear combination of many Slater determinants, each of which describes a different configuration, namely the configuration interaction (CI) method [3]. This method provides a convergent route to obtain numerically exact solutions of multi-electron systems. The main difficulty of the CI method lies in the rapid explosion of the number of Slater determinants to be included in the expansion as the number of spin-orbitals increases. For example, to represent a six-electron quantum dot with 10 available spin-orbitals we need 44,100 Slater determinants in the expansion, while with 20 spin-orbitals the number of Slater determinants to be included is 1,502,337,600. As a result, a truncated CI approach is almost inevitable in practice. In the following we describe two ways to significantly reduce the basis size while maintain high numerical accuracy.

2.1. Iterative refinement configuration interaction method (IRCI)

The number of Slater determinants increases drastically as the number of spin-orbitals increases:

\[ N_D = \binom{n_B}{n_T} \binom{n_B}{n_I} \]

However, upon examining the configuration interaction coefficients, some Slater determinants do not contribute significantly to the final solution. If these determinants are excluded, the accuracy of the solution can be improved by substituting more significant determinants.

The IRCI scheme starts with a large list of determinants, from which a subset is selected and a CI calculation is performed. Determinants with CI coefficients smaller than a certain threshold are replaced with new determinants that may contribute more to the solution, as shown in figure 1. This process iterates until the entire list is exhausted.

Figure 1. Left panel: original CI coefficients; Right panel: sorted CI coefficients.
2.2. Spin adapted configuration interaction method (SACI)

The spin-adapted CI method expands the multi-electron wavefunction as a linear combination of antisymmetrised products of basis spatial wavefunctions and spin eigenfunctions. This method has a distinct advantage over standard CI, in that a much smaller basis is needed since the spin-adapted orbitals already possess the required Fermionic symmetry [4,5,6].

**Elementary Spin functions:**

\[ \theta_k(m_S) = \sigma(1)\sigma(2) \cdots \sigma(N) \]

where \( \sigma(i) = \begin{cases} \alpha(i) & \text{represents spin up or down of } i^{th} \text{ electron.} \\ \beta(i) & \end{cases} \)

Note \( \theta_k(m_S) \) are eigenfunctions of \( \hat{S}_z \) but not \( \hat{S}^2 \).

**Spin basis functions:**

\[ X(S, m_S) = \sum_k C_k \theta_k(m_S) \]

where \( C_k \) are determined by \( \hat{S}^2 X(S, m_S) = S(S+1) X(S, m_S) \).

**Spatial basis function:**

\[ \Phi = \phi_1 \phi_2 \cdots \phi_N \]

where \( \phi_i \) are single electron spatial wavefunctions.

**Spin-adapted orbital:**

Finally, we combine both spin and spatial functions to form an orthonormal and properly antisymmetrised basis:

\[ \mathcal{A} \Phi(1, 2 \cdots N) X(S, m_S) \]

where \( \mathcal{A} = \sum_P (-1)^P P \) sums over all possible permutations. The symmetry properties of the spin eigenfunctions dictate with which spatial functions it can be combined.

3. Results

The electron densities for up to 11 electrons in a triangular dot and a square dot are shown in figures 2 and 3, respectively. For each system, density distributions for spin up and spin down electrons are also shown. Furthermore, we studied coupled quantum dots, such as the Quantum Cellular Automata (QCA) systems, which can be used to have been proposed to process both classical and quantum information. The first lowest electronic states for 12 and 16 coupled quantum dots are shown in figures 4 and 5, respectively.

![Figure 2. Total, spin up, and spin down density functions for the ground state of a triangular quantum dot (up to 12 electrons).](image-url)
4. Conclusions

Numerical calculations provide detailed and often very accurate information about nanostructured systems. With further development of theoretical models and advanced computational algorithms, many novel applications as well as potential problems which previously could only be speculated on can now be addressed in great detail.

References

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