

Thesis title: Monte Carlo and density functional theory studies of quantum dots

The aim of this thesis is to study the structure of quantum dots using variational and Green's function Monte Carlo techniques, combining results from Hartree-Fock calculations in order to achieve a as good as possible variational wave function. The thesis will explore various Monte Carlo optimization strategies and use these to define the best possible density functional. Semiconductor quantum dots are structures where charge carriers are confined in all three spatial dimensions, the dot size being of the order of the Fermi wavelength in the host material, typically between 10 nm and 1 μ m. The confinement is usually achieved by electrical gating of a two-dimensional electron gas (2DEG), possibly combined with etching techniques. Precise control of the number of electrons in the conduction band of a quantum dot (starting from zero) has been achieved in GaAs heterostructures. The electronic spectrum of typical quantum dots can vary strongly when an external magnetic field is applied, since the magnetic length corresponding to typical laboratory fields is comparable to typical dot sizes. In coupled quantum dots Coulomb blockade effects, tunneling between neighboring dots, and magnetization have been observed as well as the formation of a delocalized single-particle state.

Of particular interest for this thesis is the application of quantum dots to solar cells, as they can increase the efficiency such cells. If quantum dots are grown within a second and wider band of semiconductor material, then the quantum dot introduces a discrete level within the band gap of the host semiconductor. Different types of quantum dots confinements will be tested in this thesis, from harmonic oscillator traps to square well traps. The Monte Carlo method is rather flexible when it comes to types of confinements.

This thesis entails the development of variational Monte Carlo (VMC) and Green's function Monte Carlo (GFMC) programs to solve Schrödinger's equation and obtain various expectation values of interest, such as the energy of the ground state and excited states. The GFMC approach allows, in principle, for a numerically exact solution of Schrödinger's equation. However, it needs a reasonable starting point. It is there where a variational Monte Carlo calculation of the same system provides a variationally optimal trial wave function of a many-body system and its pertinent energy. The Slater determinant for the variational wave function is set up using single-particle wave functions from a Hartree-Fock calculation. The methods are briefly described in the following section. These calculations will in turn provide the basis for determining a as good as possible ground state wave function. This wave function will in turn be used to define the quantum mechanical density. The density will be used to construct a density functional for quantum dots using the adiabatic-connection method as described by Teale *et al* in J. Chem. Phys. **130**, 104111 (2009). The results will be compared with existing density functional for quantum dots.

The reliability of the Monte Carlo method as function of the externally applied magnetic field will be compared with ab initio coupled-cluster and large-scale diagonalization techniques. The latter two calculations will be performed by other Master of science students.

The results are expected to be published in leading journals.

A progress plan for this thesis project is given at the end.

Variational Monte Carlo

The variational quantum Monte Carlo (VMC) has been widely applied to studies of quantal systems. The recipe consists in choosing a trial wave function $\psi_T(\mathbf{R})$ which we assume to be as realistic as possible. The variable \mathbf{R} stands for the spatial coordinates, in total $2N$ if we have N particles present. The trial wave function serves then as a mean to define the quantal probability distribution

$$P(\mathbf{R}) = \frac{|\psi_T(\mathbf{R})|^2}{\int |\psi_T(\mathbf{R})|^2 d\mathbf{R}}. \quad (1)$$

The expectation value of the energy E is given by

$$\langle E \rangle = \frac{\int d\mathbf{R} \Psi^*(\mathbf{R}) H(\mathbf{R}) \Psi(\mathbf{R})}{\int d\mathbf{R} \Psi^*(\mathbf{R}) \Psi(\mathbf{R})}, \quad (2)$$

where Ψ is the exact eigenfunction. Using our trial wave function we define a new operator, the so-called local energy,

$$E_L(\mathbf{R}) = \frac{1}{\psi_T(\mathbf{R})} H \psi_T(\mathbf{R}), \quad (3)$$

which, together with our trial PDF allows us to rewrite the expression for the energy as

$$\langle H \rangle = \int P(\mathbf{R}) E_L(\mathbf{R}) d\mathbf{R}. \quad (4)$$

This equation expresses the variational Monte Carlo approach.

The first part of this thesis deals thus with a VMC calculation of spherical quantum dots as functions of the strength of the applied magnetic field.

The trial wave function is a combination of a Slater determinant and a correlation part. The Slater determinant will be constructed using single-particle wave functions based on Hartree-Fock theory.

Green's function Monte Carlo (GFMC)

The GFMC method is based on rewriting the Schrödinger equation in imaginary time, by defining $\tau = it$. The imaginary time Schrödinger equation is then

$$\frac{\partial \psi}{\partial \tau} = -\hat{\mathbf{H}}\psi, \quad (5)$$

where we have omitted the dependence on τ and the spatial variables in ψ .

A Green's function Monte Carlo, although it allows for a formally exact solution of Schrödinger's equations, needs a clever starting point for the energy. This trial energy is initially chosen to be the VMC energy of the trial wave function, and is updated as the simulation progresses. Use of an optimised trial function minimises the difference between the local and trial energies, and hence minimises fluctuations in the calculations. A wave function optimised using VMC is ideal for this purpose, and in practice VMC provides the best method for obtaining wave functions that accurately approximate ground state wave functions locally.

The final aim of this thesis is thus to develop a GFMC program for studying quantum dots, based on a VMC calculation first.

These calculations will in turn provide the basis for determining a as good as possible ground state wave function. This wave function will in turn be used to define the quantum mechanical density. The density will be used to construct a density functional for quantum dots using the adiabatic-connection method as described by Teale *et al* in J. Chem. Phys. **130**, 104111 (2009). The results will be compared with existing density functionals for various quantum dots.

Progress plan and milestones

The aims and progress plan of this thesis are as follows

- Fall 2010: Develop first a Hartree-Fock code for electrons trapped in a single harmonic oscillator in two dimensions. This part entails developing a code for computing the Coulomb interaction in two dimensions in the laboratory system.
- The Hartree-Fock interaction is then used as input to the variational Monte Carlo code. The results will be compared with large scale diagonalization and coupled-cluster techniques for 2, 6, 12 and 20 electrons in a single harmonic oscillator well.

- Fall 2010: Write a code which solves the variational Monte-Carlo (VMC) problem for quantum dots. Both closed-core and open shell quantum dots will be studied. Construct a Green's function Monte Carlo code based on the Variational Monte Carlo code. The GFMC code receives as input the optimal variational energy and wave function from the VMC calculation and solves in principle the Schrödinger equation exactly. The Slater determinant used in the VMC calculation includes also single-particle wave functions from Hartree-Fock calculations.
- Spring 2011: The obtained ground states will in turn be used to define a as exact as possible density functional for quantum dots using the adiabatic-connection method. The density functional can in turn be used to model systems with a large number of electrons in quantum dots. Comparisons with a density functional derived from coupled-cluster methods will also be made in order to test the validity of the Monte Carlo approach. Possible applications are to mechanical studies of solar cells.

The thesis is expected to be handed in June 1 2011.

References

- [1] A. M. Teale, S. Coriani, and T. Helgaker, J. Chem. Phys. **130**, 104111 (2009).
- [2] Jenny Olsen, *The Physics of Solar Cells* (Imperial College, Press, London, 2009).