Quantum Monte Carlo studies of quantum dots

The aim of this project is to study the structure of quantum dots using variational Monte Carlo techniques, combining results from Hartree-Fock calculations in order to achieve a as good as possible variational wave function. The project will explore various Monte Carlo optimalization 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 μm. 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.

Different types of quantum dots confinements will be tested in this project, from harmonic oscillator traps to square well traps, allowing thereby for a direct comparison with eperiments on quantum dots. If successful, these results can be published in scientific journals.

The Monte Carlo method is rather flexible when it comes to types of confinements and gives insights about the quantum mechanical systems that are often difficult (or more tedious) to obtain using other many-body methods. It has the possibility to develop deep insights about quantum mechanical systems that are normally achieved only during graduate studies. Topics like correlations energy, density distributions etc are important quantities in studies of interacting quantum mechanical systems. These are topics which seldomly are exposed in regular courses, even at the graduate level.

This project entails thus the development of a variational Monte Carlo (VMC) program to solve Schrödingers equation and obtain various expectation values of interest, such as the energy of the ground state and the expectation energies of the kinetic and potential energies.

The Monte Carlo calculation provides a variationally optimal trial wave function of a many-body system and its pertinent energy. With this wave function it is possible to compute several observables of interest. The Slater determinant for the variational wave function is set up using single-particle wave functions from a Hartree-Fock calculation. It is assumed that a Hartree-Fock calculation has already been performed. The VMC calculations will in turn provide the basis for determining a as good as possible ground state wave function.

The various tasks leading to a professional program for quantum mechanical studies are detailed below.

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

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

The expectation value of the energy E is given by

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

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

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

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

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

This equation expresses the variational Monte Carlo approach.

This project 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.

These calculations will in turn provide the basis for determining a as good as possible ground state wave function. This wave function can in turn, if time allows, be used to define the quantum mechanical density. The density can 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 can in turn be compared with existing density functionals for various quantum dots.

Explict tasks and work plan

Here we detail the various steps that will lead to professional Monte Carlo program for studying interacting electrons. If properly written, the program can easily be extended to three-dimensional electronic systems as well other interacting fermionic or bosonic systems.

We consider a system of electrons confined in a pure two-dimensional isotropic harmonic oscillator potential, with an idealized total Hamiltonian given by

$$\hat{H} = \sum_{i=1}^{N} \left(-\frac{1}{2}\nabla_i^2 + \frac{1}{2}\omega^2 r_i^2 \right) + \sum_{i<j} \frac{1}{r_{ij}}, \tag{5}$$

where natural units ($\hbar = c = e = m_e = 1$) are used and all energies are in so-called atomic units a.u. We will study systems of many electrons N as functions of the oscillator frequency ω using the above Hamiltonian. The Hamiltonian includes a standard harmonic oscillator part

$$\hat{H}_0 = \sum_{i=1}^{N} \left(-\frac{1}{2}\nabla_i^2 + \frac{1}{2}\omega^2 r_i^2 \right), \tag{6}$$

and the repulsive interaction between two electrons given by

$$\hat{H}_1 = \sum_{i<j} \frac{1}{r_{ij}}, \tag{7}$$

with the distance between electrons given by $r_{ij} = |\boldsymbol{r}_1 - \boldsymbol{r}_2|$. We define the modulus of the positions of the electrons (for a given electron i) as $r_i = \sqrt{r_{i_x}^2 + r_{i_y}^2}$.

Part a): In parts a-f we will deal only with a system of two electrons in a quantum dot with a frequency of $\hbar\omega = 1$. The reason for this is that we have exact closed form expressions for the ground state energy from Taut's work for selected values of ω, see M. Taut, Phys. Rev. A **48**, 3561 (1993). The energy is given by 3 a.u. (atomic units) when the interaction between the electrons is included. If only the harmonic oscillator part of the Hamiltonian is included, the so-called unperturbed part,

$$\hat{H}_0 = \sum_{i=1}^{N} \left(-\frac{1}{2}\nabla_i^2 + \frac{1}{2}\omega^2 r_i^2 \right), \tag{8}$$

the energy is 2 a.u. The wave function for one electron in an oscillator potential in two dimensions is

$$\phi_{n_x,n_y}(x,y) = AH_{n_x}(\sqrt{\omega}x)H_{n_y}(\sqrt{\omega}y)\exp\left(-\omega(x^2+y^2)/2. \tag{9}\right.$$

The functions $H_{n_x}(\sqrt{\omega}x)$ are so-called Hermite polynomials while A is a normalization constant. For the lowest-lying state we have $n_x = n_y = 0$ and an energy $\epsilon_{n_x,n_y} = \omega(n_x + n_y + 1) = \omega$. Convince yourself that the lowest-lying energy for the two-electron system is simply 2ω.

The unperturbed wave function for the ground state of the two-electron system is given by

$$\Phi(\boldsymbol{r}_1, \boldsymbol{r}_2) = C \exp\left(-\omega(r_1^2 + r_2^2)/2\right), \tag{10}$$

with C being a normalization constant and $r_i = \sqrt{r_{i_x}^2 + r_{i_y}^2}$. Note that the vector $\boldsymbol{r}_i$ refers to the x and y position for a given particle. What is the total

spin of this wave function? Find arguments for why the ground state should have this specific total spin.

Part b): We want to perform a Variational Monte Carlo calculation of the ground state of two electrons in a quantum dot well with different oscillator energies, assuming total spin $S = 0$ using the Hamiltonian of Eq. (5). Our trial wave function which has the following form

$$\psi_T(\boldsymbol{r}_1, \boldsymbol{r}_2) = C \exp\left(-\alpha\omega(r_1^2 + r_2^2)/2\right) \exp\left(\frac{ar_{12}}{(1 + \beta r_{12})}\right), \qquad (11)$$

where a is equal to one when the two electrons have anti-parallel spins and $1/3$ when the spins are parallel. Finally, α and β are our variational parameters. Note well the dependence on α for the single-particle part of the trial function. It is important to remember this when you use higher-order Hermite polynomials. Find the analytical expressions for the local energy.

Part c): Your task is to perform a Variational Monte Carlo calculation using the Metropolis algorithm to compute the integral

$$\langle E \rangle = \frac{\int d\boldsymbol{r}_1 d\boldsymbol{r}_2 \psi_T^*(\boldsymbol{r}_1, \boldsymbol{r}_2) \hat{H}(\boldsymbol{r}_1, \boldsymbol{r}_2) \psi_T(\boldsymbol{r}_1, \boldsymbol{r}_2)}{\int d\boldsymbol{r}_1 d\boldsymbol{r}_2 \psi_T^*(\boldsymbol{r}_1, \boldsymbol{r}_2) \psi_T(\boldsymbol{r}_1, \boldsymbol{r}_2)}. \qquad (12)$$

Compute the expectation value of the energy using both the analytical expression for the local energy and numerical derivation of the kinetic energy. Compare the time usage between the two approaches. Perform these calculations without importance sampling and also without the Jastrow factor. For the calculations without the Jastrow factor and repulsive Coulomb potential, your energy should equal 2.0 a.u. and your variance should be exactly equal to zero.

Part d): Add now importance sampling and repeat the calculations from the previous exercise but use only the analytical expression for the local energy. Perform also a blocking analysis in order to obtain the optimal standard deviation. Compare your results with the those without importance sampling and comment your results.

Part e): Using either the steepest descent method or the conjugate gradient method, find the optimal variational parameters and perform your Monte Carlo calculations using these. In addition, you should parallelize your program using MPI and/or OpenMP.

Part f): Finally, we wil now analyze and interpret our results for the two-electron systems. Find the energy minimum and discuss your results compared with the analytical solution from Taut's work, see reference [1] below. Compute also the mean distance $r_{12} = |\boldsymbol{r}_1 - \boldsymbol{r}_2|$ (with $r_i = \sqrt{r_{i_x}^2 + r_{i_y}^2}$) between the two electrons for the optimal set of the variational parameters. With the optimal

parameters for the ground state wave function, compute the onebody density. Discuss your results and compare the results with those obtained with a pure harmonic oscillator wave functions. Run a Monte Carlo calculations without the Jastrow factor as well and compute the same quantities. How important are the correlations induced by the Jastrow factor? Compute also the expectation value of the kinetic energy and potential energy using $\omega = 0.01$, $\omega = 0.05$, $\omega = 0.1$, $\omega = 0.5$ and $\omega = 1.0$. Comment your results. Hint, think of the virial theorem.

Discuss also your results with those obtained with Hartree-Fock theory for two electrons.

Part g): The previous parts have prepared you for extending your calculational machinery to other systems. Here we will focus on quantum dots with $N = 6$ and $N = 12$ electrons.

The new item you need to pay attention to is the calculation of the Slater Determinant. This is an additional complication to your VMC calculations. You will also need to use replace the single-particle functions of the Slater determinant with those obtained from your Hartree-Fock calculations. If we stick to harmonic oscillator like wave functions, the trial wave function for say an $N = 6$ electron quantum dot can be written as

$$\psi_T(\boldsymbol{r}_1, \boldsymbol{r}_2, \ldots, \boldsymbol{r}_6) = Det\left(\phi_1(\boldsymbol{r}_1), \phi_2(\boldsymbol{r}_2), \ldots, \phi_6(\boldsymbol{r}_6)\right) \prod_{i<j}^{6} \exp\left(\frac{ar_{ij}}{(1 + \beta r_{ij})}\right),$$
(13)

where Det is a Slater determinant and the single-particle wave functions are the harmonic oscillator wave functions for the $n_x = 0, 1$ and $n_y = 0, 1$ orbitals. Similarly, for the $N = 12$ quantum dot, the trial wave function can take the form

$$\psi_T(\boldsymbol{r}_1, \boldsymbol{r}_2, \ldots, \boldsymbol{r}_{12}) = Det\left(\phi_1(\boldsymbol{r}_1), \phi_2(\boldsymbol{r}_2), \ldots, \phi_{12}(\boldsymbol{r}_{12})\right) \prod_{i<j}^{12} \exp\left(\frac{ar_{ij}}{(1 + \beta r_{ij})}\right),$$
(14)

In this case you need to include the $n_x = 2$ and $n_y = 2$ wave functions as well. Observe that $r_i = \sqrt{r_{i_x}^2 + r_{i_y}^2}$.

Write a function which sets up the Slater determinant. Find the Hermite polynomials which are needed for $n_x = 0, 1, 2$ and obviously n_y as well. Relate these to your Hartree-Fock results. Compute the ground state energies of quantum dots for $N = 6$ and $N = 12$ electrons, following the same set up as in the previous exercises for $\omega = 0.01$, $\omega = 0.05$, $\omega = 0.1$, $\omega = 0.5$, and $\omega = 1.0$. The calculations should include parallelization, blocking, importance sampling and energy minimization using the conjugate gradient approach or similar approaches. To test your Slater determinant code, you should reproduce the unperturbed single-particle energies when the electron-electron repulsion is switched off. Convince yourself that the unperturbed ground state energies for

$N = 6$ is 10ω and for $N = 12$ we obtain 28ω. What is the expected total spin of the ground states?

Part h): With the optimal parameters for the ground state wave function, compute again the onebody density. Discuss your results and compare the results with those obtained with a pure harmonic oscillator wave functions. Run a Monte Carlo calculations without the Jastrow factor as well and compute the same quantities. How important are the correlations induced by the Jastrow factor? Compute also the expectation value of the kinetic energy and potential energy using $\omega = 0.01$, $\omega = 0.05$, $\omega = 0.1$, $\omega = 0.5$, and $\omega = 1.0$. Comment your results.

Part i): The last exercise is a performance analysis of your code(s) for the case of $N = 6$ electrons. Make a performance analysis by timing your serial code with and without vectorization. Perform several runs with the same number of Monte carlo cycles and compute an average timing analysis with and without vectorization. Comment your results. Use at least 10^6 Monte Carlo samples.

Compare thereafter your serial code(s) with the speedup you get by parallelizing your code, running either OpenMP or MPI or both. Do you get a near 100% speedup with the parallel version? Comment again your results and perform timing benchmarks several times in order to extract an average performance time.

With all these steps included, you should now have a professional Variational Monte Carlo program that, if properly written, can be extended to other systems. Furthermore, it provides the basis for making comparisons with experiment and other theories.

Literature.

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