Many-Body Approaches to Quantum Dots

Patrick Merlot

Department of Computational Physics

November 4, 2009

Table of contents

Quantum Dots
Model, Methods and Implementation
Results and Discussions

What is a quantum dot?
Physics of QDs
Applications of QDs

Table of contents

Quantum Dots
Model, Methods and Implementation
Results and Discussions

What is a quantum dot?
Physics of QDs
Applications of QDs

What is a quantum dot?

Definition

- Semiconductor whose charge-carriers are confined in space.
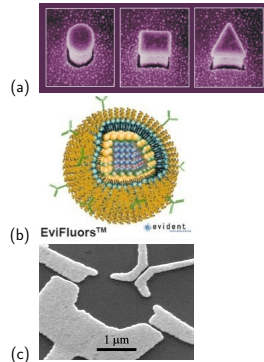- $\neq$ types/shapes/fabrication $\Rightarrow \neq$ QDs

Figure 1: Possible types/shapes of QDs.
(a) Various shapes of QDs pillars$\sim 0.5 \mu m$,
(b) Colloidal QD (InGaP+ZnS+lipid)$\sim 10 nm$,
(c) QD defined by 5 metallic gates on GaAs where 2-DEG is trapped.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

What is a quantum dot?
Physics of QDs
Applications of QDs

Physics of QDs

Quantum dot properties

- Semiconductor band gap increased by size quantization
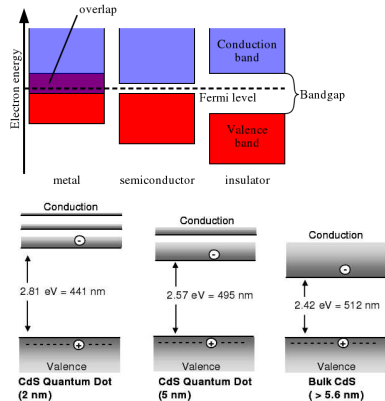
Figure 2: Electronic band structure of semiconductor and quantum dots (Courtesy of J.Winter[4]).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

What is a quantum dot?
Physics of QDs
Applications of QDs

Physics of QDs

Quantum dot properties

- Semiconductor band gap increased by size quantization
- Tunable optical/electrical properties
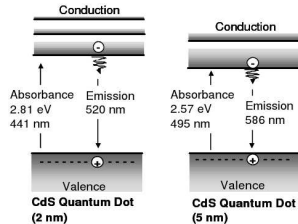- Perfect system for computational studies

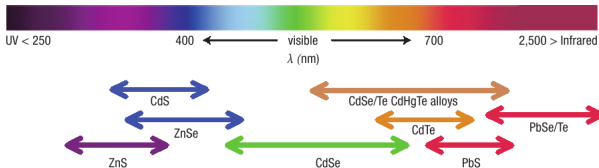Figure 3: Fluorescent emission (Courtesy of J.Winter[4]).

Figure 4: Emission spectra of various quantum dots.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

What is a quantum dot?
Physics of QDs
Applications of QDs

Applications of QDs

Figure 5: QDs imaging in live animals compared to classical organic dyes. (Courtesy of X. Gao)

Possible applications

- Biological nano-sensors
- Qubits for QCA
- LEDs
- Solar cells

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Table of contents

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Model of a quantum dot

Simple model of a quantum dot

- **Atomic scale** problem $\Rightarrow$ **Quantum mechanics** for an accurate description of the system.
  $\Rightarrow$ at rest, solve the time-independent Schrödinger equation $\hat{H}|\Psi\rangle = E|\Psi\rangle$.

- Not modelling all nuclei/electrons $\Rightarrow$ just model the few quasiparticles confined by the semiconductor.
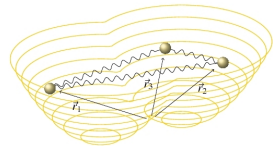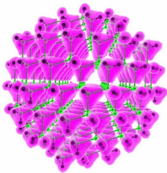

Figure 6: Illustration of a quantum dot model (Courtesy of S.Kvaal[5]).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Model of a N-particle system

The Schrödinger equation

- The Schrödinger equation of a N-particle system

$$\hat{H}(\mathbf{r_1}, \mathbf{r_2}, \ldots, \mathbf{r_N})|\Psi_\kappa(\mathbf{r_1}, \mathbf{r_2}, \ldots, \mathbf{r_N})\rangle = E_\kappa|\Psi_\kappa(\mathbf{r_1}, \mathbf{r_2}, \ldots, \mathbf{r_N})\rangle \quad (1)$$

where $\mathbf{r_i}$ reprensents the (spatial/spin) coordinates of quasiparticle i,
κ stands for all quantum numbers needed to classify a given
N-particle state,
$|\Psi_\kappa\rangle$ and E_κ are the eigenstates and eigenenergies of the system.

How to define our Hamiltonian?

- $\hat{H} = \sum_{i=1}^{N_e} \dfrac{\mathbf{p_i}^2}{2m^*} + \ldots$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Definitions of the interactions/potentials

Forces/Fields acting on the quasiparticles:

- Forces confining the particles $\Rightarrow$ Confining potential

- Interactions between the particles $\Rightarrow$ Interaction potential

The Hamiltonian of our two-electron quantum dot model

- Confining potential $\Rightarrow$ the harmonic oscillator (parabolic) potential

$$\hat{H} = \sum_{i=1}^{N_e=2} \frac{\mathbf{p_i}^2}{2m^*} + \sum_{i=1}^{N_e=2} \frac{1}{2} m^* \omega_0^2 \|\mathbf{r_i}\|^2 + \frac{e^2}{4\pi\epsilon_0\epsilon_r} \frac{1}{\|\mathbf{r_1} - \mathbf{r_2}\|}, \qquad (2)$$

- Interaction potential $\Rightarrow$ the two-body Coulomb interaction

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Applying an external magnetic field

1. $p_i \longrightarrow p_i + eA$, where A is the vector potential defined by $B = \nabla \times A$.

- in coordinate space $p_i \rightarrow -i\hbar\nabla_i$,

- using a Coulomb gauge $\nabla \cdot A = 0$ (by choosing $A(r_i) = \dfrac{B \times r_i}{2}$),

$$(p_i + eA)^2 \rightarrow \left(-\frac{\hbar^2}{2m^*}\nabla_i^2 - i\hbar\frac{e}{m^*}A \cdot \nabla_i + \frac{e^2}{2m^*}A^2 \right). \qquad (3)$$

In terms of B, the linear and quadratic terms in A have the form

$\dfrac{-i\hbar e}{m^*}A \cdot \nabla_i = \dfrac{e}{2m^*}B \cdot L$, and $\dfrac{e^2}{2m^*}A^2 = \dfrac{e^2}{8m^*}B^2 r_i^2$. where $L = -i\hbar(r_i \times \nabla_i)$

is the orbital angular momentum operator of the electron i.

2. B also acts on spin with the additional energy term: $\hat{H}_s = g_s^* \dfrac{\omega_c}{2}\hat{S}_z$, where $\hat{S}$

is the spin operator of the electron and g_s^* is its effective spin gyromagnetic
ratio and $\omega_c = eB/m^*$ is known as the cyclotron frequency.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Final Hamiltonian

The final Hamiltonian reads:

$$
\hat{H} = \sum_{i=1}^{N_e} \left(\frac{-\hbar^2}{2m^*}\nabla_i^2 + \overbrace{\frac{1}{2}m^*\omega_0^2\|\mathbf{r_i}\|^2}^{\substack{\text{Harmonic ocscillator} \\ \text{potential}}} \right) + \overbrace{\frac{e^2}{4\pi\epsilon_0\epsilon_r}\sum_{i<j}\frac{1}{|\mathbf{r_i}-\mathbf{r_j}|}}^{\substack{\text{Coulomb} \\ \text{interactions}}}
$$

$$
+ \underbrace{\sum_{i=1}^{N_e}\left(\frac{1}{2}m^*\left(\frac{\omega_c}{2}\right)^2\|\mathbf{r_i}\|^2 + \frac{1}{2}\omega_c\hat{L}_z^{(i)} + \frac{1}{2}g_s^*\omega_c\hat{S}_z^{(i)}\right)}_{\substack{\text{single particle interactions} \\ \text{with the magnetic field}}}, \tag{4}
$$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Scaling the problem: dimensionless form of $\hat{H}$

New constant, the oscillator frequency $\omega = \sqrt{\omega_0 + \left(\frac{\omega_c}{2}\right)^2}$,

New units:

- the energy unit $\hbar\omega$,
- the length unit, the oscillator length defined by $l = \sqrt{\hbar/(m^*\omega)}$.

The dimensionless Hamiltonian is now

$$\hat{H} = \sum_{i=1}^{N_e} \left[-\frac{1}{2}\nabla_i^2 + \frac{1}{2}r_i^2 \right] + \lambda \sum_{i<j} \frac{1}{r_{ij}} + \sum_{i=1}^{N_e} \left(\frac{1}{2}\frac{\omega_c}{\hbar\omega}m_l^{(i)} + \frac{1}{2}g_s^*\frac{\omega_c}{\hbar\omega}m_s^{(i)} \right). \quad (5)$$

where the new dimensionless parameter $\lambda = l/a_0^*$ describes the strength of the electron-electron interaction (a_0^* being the effective Bohr radius).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The Hamiltonian solved in this project

$$\lambda(B) = \frac{1}{a_0^*} \left(\frac{4\hbar^2}{4\omega_0^2 m^{*2} + e^2 B^2} \right)^{\frac{1}{4}}$$

Role of B?

- Squeezing the particles should increase the strength of the electron-electron interaction.

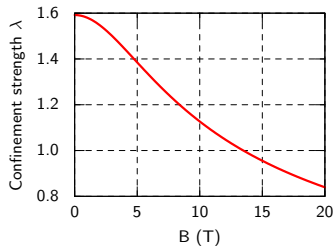- λ only decreases as the magnetic field increases in this model.

Figure 7: Dimensionless confinement strength λ as a function of the magnetic field strength in GaAs.

In the rest of the project, we will solve the following Hamiltonian:

$$\hat{H} = \sum_{i=1}^{N_e} \left[-\frac{1}{2} \nabla_i^2 + \frac{1}{2} r_i^2 \right] + \lambda \sum_{i<j} \frac{1}{r_{ij}}. \tag{6}$$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The method of variational calculus

Definition

Method to solve the Schrödinger eq. more efficiently than using numerical integration.

- Based on the method of Lagrange multipliers, where the functional to minimize (the energy functional) is an integral over the unknown wave function $|\Phi\rangle$

$$E[\Phi] = \frac{\langle\Phi|H|\Phi\rangle}{\langle\Phi|\Phi\rangle} = \frac{\int \Phi^* H\Phi d\tau}{\int \Phi^*\Phi d\tau}, \tag{7}$$

  while subject to a normalization constraint $\langle\Phi|\Phi\rangle = 1$.

- The method introduces new variables for each of the constraints (the Lagrange multipliers ϵ) and defines the Lagrangian (Λ) with respect to $|\Phi\rangle$ as

$$\Lambda(\Phi, \epsilon) = E[\Phi] - \epsilon\left(\langle\Phi|\Phi\rangle - 1\right), \tag{8}$$

- Find stationnary solutions by solving the set of equations by writing $\dfrac{\partial\Lambda}{\partial\Phi} = 0$.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The Hartree-Fock method

Definition

The HF method is a particular case of variational method in accordance with

- the independent particle approximation,
- the Pauli exclusion principle.

The approximated wave function

To fullfil these criteria, the wave-function must be antisymmetric with respect to an interchange of any two particles:

$$\Phi(\mathbf{r_1}, \mathbf{r_2}, \ldots, \mathbf{r_i}, \ldots, \mathbf{r_j}, \ldots, \mathbf{r_N}) = -\Phi(\mathbf{r_1}, \mathbf{r_2}, \ldots, \mathbf{r_j}, \ldots, \mathbf{r_i}, \ldots, \mathbf{r_N}). \qquad (9)$$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The Hartree-Fock wave function

The Slater determinant is an antisymmetric product of the single particle orbitals:

$$\Phi(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r_N}, \alpha, \beta, \ldots, \sigma) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \psi_\alpha(\mathbf{r}_1) & \psi_\alpha(\mathbf{r}_2) & \ldots & \psi_\alpha(\mathbf{r}_N) \\ \psi_\beta(\mathbf{r}_1) & \psi_\beta(\mathbf{r}_2) & \ldots & \psi_\beta(\mathbf{r}_N) \\ \vdots & \vdots & \ddots & \vdots \\ \psi_\sigma(\mathbf{r}_1) & \psi_\sigma(\mathbf{r}_2) & \ldots & \psi_\sigma(\mathbf{r}_N) \end{vmatrix}, \quad (10)$$

It can be rewritten as

$$\Phi_T(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r_N}, \alpha, \beta, \ldots, \sigma) = \frac{1}{\sqrt{N!}} \sum_p (-)^p P \Psi_\alpha(\mathbf{r_1}) \Psi_\beta(\mathbf{r_2}) \ldots \Psi_\sigma(\mathbf{r_N}) \quad (11)$$

$$= \sqrt{N!} \mathcal{A} \left(\Psi_\alpha(\mathbf{r_1}) \Psi_\beta(\mathbf{r_2}) \ldots \Psi_\sigma(\mathbf{r_N}) \right), \quad (12)$$

by introducing the antisymmetrization operator $\mathcal{A} = \dfrac{1}{N!} \sum_P (-)^p \hat{P}$.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Matrix elements calculations

Definition

We write the Hamiltonian for N electrons as $\hat{H} = \hat{H}_0 + \hat{H}_1 = \sum_{i=1}^{N} \hat{h}_i + \sum_{i<j}^{N} v(\mathbf{r_i}, \mathbf{r_j})$,

where $r_{ij} = \|\vec{r_i} - \vec{r_j}\|$, $\hat{h}_i$ and $v(\mathbf{r_i}, \mathbf{r_j})$ are respectively the one-body and the two-body Hamiltonian.

Using properties of $\mathcal{A}$ and commutation rule with $\hat{H}_0$ and $\hat{H}_1$, one can write:

$$\int \Phi_T^* \hat{H}_0 \Phi_T d\tau = \sum_{\mu=1}^{N} \int \Psi_\mu^*(\mathbf{r}) \hat{h} \Psi_\mu(\mathbf{r}) d\mathbf{r} = \sum_{\mu=1}^{N} \langle \mu | h | \mu \rangle. \tag{13}$$

$$\int \Phi_T^* \hat{H}_1 \Phi_T d\tau = \frac{1}{2} \sum_{\mu=1}^{N} \sum_{\nu=1}^{N} \langle \mu\nu | V | \mu\nu \rangle_{AS}. \tag{14}$$

where we define the antisymmetrized matrix element as

$\langle \mu\nu | V | \mu\nu \rangle_{AS} = \langle \mu\nu | V | \mu\nu \rangle - \langle \mu\nu | V | \nu\mu \rangle$, with the following shorthands
$\langle \mu\nu | V | \mu\nu \rangle = \int \Psi_\mu^*(\mathbf{r_i}) \Psi_\nu^*(\mathbf{r_j}) V(r_{ij}) \Psi_\mu(\mathbf{r_i}) \Psi_\nu(\mathbf{r_j}) d\mathbf{r_i} d\mathbf{r_j}$.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The Hartree-Fock energy in the harmonic oscillator basis

The energy functional

The energy functional is our starting point for the Hartree-Fock calculations.

$$E[\Phi_T] = \langle \Phi_T | \hat{H}_0 | \Phi_T \rangle + \langle \Phi_T | \hat{H}_1 | \Phi_T \rangle \tag{15}$$

$$= \sum_{\mu=1}^{N} \langle \mu | h | \mu \rangle + \frac{1}{2} \sum_{\mu=1}^{N} \sum_{\nu=1}^{N} \langle \mu\nu | V | \mu\nu \rangle_{AS}. \tag{16}$$

We expand each single-particle eigenvector Ψ_i in terms of a convenient complete set of single-particle states $|\alpha\rangle$ (the harmonic oscillator eigenstates in our case),

$$\Psi_i = |i\rangle = \sum_{\alpha} c_i^{\alpha} |\alpha\rangle. \tag{17}$$

The energy functional now reads

$$E[\Phi] = \sum_{i=1}^{N} \sum_{\alpha\gamma} C_i^{\alpha*} C_i^{\gamma} \langle \alpha | h | \gamma \rangle + \frac{1}{2} \sum_{i,j=1}^{N} \sum_{\alpha\beta\gamma\delta} C_i^{\alpha*} C_j^{\beta*} C_i^{\gamma} C_j^{\delta} \langle \alpha\beta | V | \gamma\delta \rangle_{AS}. \tag{18}$$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The Hartree-Fock equations (1)

Remember the method of the Lagrange multipliers

1. Define a functional $E[\Phi_T]$,

2. Identify the constraints: $\langle\Psi_i|\Psi_j\rangle = \delta_{ij}$ which implies $\langle\Phi_T|\Phi_T\rangle = 1$,
   with $\langle\Psi_i|\Psi_j\rangle = \sum_{\alpha\beta} C_i^{\alpha*} C_j^{\beta} \underbrace{\langle\alpha|\beta\rangle}_{\delta_{\alpha\beta}} = \sum_{\alpha} C_i^{\alpha*} C_j^{\alpha}$

3. Define the Lagrangian Λ

$$\Lambda(C_1^{\alpha}, C_2^{\alpha}, \ldots, C_N^{\alpha}, \epsilon_1, \epsilon_2, \ldots, \epsilon_N) = E[\Phi_T] - \sum_{i=1}^{N} \epsilon_i \left(\sum_{\alpha} C_i^{\alpha*} C_i^{\alpha} - \delta_{ij} \right). \quad (19)$$

where ϵ_i are the Lagrange multipliers for each of the normalization constraints.

4. Get the system of equations to solve by setting Λ

$$\frac{d\Lambda}{d\Phi_T} \equiv \frac{d}{dC_i^{\alpha*}} \left[\Lambda(C_1^{\alpha}, C_2^{\alpha}, \ldots, C_N^{\alpha}, \epsilon_1, \epsilon_2, \ldots, \epsilon_N) \right] = 0, \quad \forall\ i \in \mathbb{N}^*. \quad (20)$$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The Hartree-Fock equations (2)

Treating C_i^α and $C_i^{\alpha*}$ as independent, we arrive at the Hartree-Fock equations (one equation for each basis state $|\alpha\rangle$)

$$\sum_\gamma \langle\alpha|h|\gamma\rangle\ C_i^\gamma + \sum_\gamma \overbrace{\left[\sum_{j=1}^{N} \sum_{\beta\delta} C_j^{\beta*} \underbrace{\langle\alpha\beta|V|\gamma\delta\rangle_{AS}}_{\substack{\text{Two-body interaction}\\ \text{matrix element}\, V_{\alpha\beta\gamma\delta}}} C_j^\delta\right]}^{\substack{\text{Effective potential}\\ \langle\alpha|U|\gamma\rangle}} C_i^\gamma = \epsilon_i\ C_i^\alpha, \qquad (21)$$

which we can rewrite as $\displaystyle\sum_\gamma \mathcal{O}_{\alpha\gamma}\ C_i^\gamma = \epsilon_i\ C_i^\alpha, \quad \forall\ \alpha \in \mathcal{H}.$

$\Rightarrow$ System of non-linear equations in the $C_i^{\alpha*}$, since $\mathcal{O}_{\alpha\gamma}$ depends itself on the unknowns.

$\Rightarrow$ To be solved by an iterative procedure.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The Hartree-Fock (self-consistent) iterative procedure

1. Compute the effective Coulomb interaction potential $\langle\alpha|U^{(0)}|\gamma\rangle$ with an initial guess of the $C_i^{\alpha(0)}$.

2. Build the resulting Fock matrix $\mathcal{O}$.

3. Solve the linearised system given by the equations (Fock matrix diagonalization)

$$\sum_\gamma \left[\langle\alpha|h|\gamma\rangle + \langle\alpha|U|\gamma\rangle\right] C_i^\gamma = \epsilon_i \ C_i^\alpha.$$

   at iteration (k), store the output eigenenergies $\epsilon_i^{(k)}$ and the coefficients of the new eigenvectors $C_i^{\alpha(k)}$.

4. Substitute back the new coefficients to compute a new Coulomb interaction potential.

5. ...

6. Continue the process until self-consistency is reached.
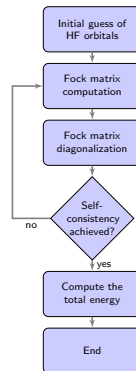
Figure 8: Flowchart of Hartree-Fock algorithm.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Many-body perturbation theory

Take the Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}'$, and treat $\hat{H}'$ as a perturbation, such as the Coulomb interaction.

Suppose Φ_n eigenfunctions of $\hat{H}_0$ corresponding to the eigenvalues E_n: $\hat{H}_0\Phi_n = E_n\Phi_n$.
Consider the effect of the perturbation on a particular state Φ_0.
We denote by Ψ_0 the state into which Φ_0 changes under the action of the perturbation, so that Ψ_0 is an eigenfunction of $\hat{H}$, corresponding to the eigenvalue E.

$$\hat{H}_0\Phi_0 = E_0\Phi_0. \tag{22}$$

$$\hat{H}\Psi_0 = E\Psi_0. \tag{23}$$

Therefore Φ_0 and Ψ_0 denote the ground states of the unperturbed and perturbed systems respectively.
Since $\hat{H}_0$ is Hermitian, one can show that:

$$E - E_0 = \frac{\langle\Phi_0|\hat{H}'|\Psi_0\rangle}{\langle\Phi_0|\Psi_0\rangle}. \tag{24}$$

which is an exact expression and independent of any particular perturbation method.
Since Ψ_0 is unknown, using a *projection operator* **R** for the state Φ_0 defined by the equation

$$\mathbf{R}\Psi = \Psi - \Phi_0\langle\Phi_0|\Psi\rangle, \tag{25}$$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The perturbed energy

The perturbed energy can be derived from the iterated Ψ_0 which gives

$$E - E_0 = \sum_{n=0}^{\infty} \langle \Phi_0 | \hat{H}' \left(\frac{\mathbf{R}}{E_0 - \hat{H}_0} (E_0 - E + \hat{H}') \right)^n | \Phi_0 \rangle. \tag{26}$$

We shall write

$$\Delta E = E - E_0 = \Delta E^{(1)} + \Delta E^{(2)} + \Delta E^{(3)} + \dots$$

where the mth-order energy correction $\Delta E^{(m)}$ contains the mth-order power of the perturbation $\hat{H}'$.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The many-body perturbation corrections

- The 1st-order correction is

$$\Delta E^{(1)} = \langle \Phi_0 | \hat{H}' | \Phi_0 \rangle. \tag{27}$$

- The 2nd-order correction is

$$\Delta E^{(2)} = \langle \Phi_0 | \hat{H}' \frac{\mathbf{R}}{E_0 - \hat{H}_0} (E_0 - E + \hat{H}') | \Phi_0 \rangle. \tag{28}$$

- The 3rd-order energy correction reads

$$\Delta E^{(3)} = \sum_{n=0}^{\infty} \sum_{n=0}^{\infty} \frac{\langle \Phi_0 | \hat{H}' | \Phi_m \rangle \langle \Phi_m | \hat{H}' | \Phi_n \rangle \langle \Phi_n | \hat{H}' | \Phi_0 \rangle}{(E_0 - E_m)(E_0 - E_n)} \tag{29}$$
$$- \langle \Phi_0 | \hat{H}' | \Phi_0 \rangle \sum_{n=0}^{\infty} \frac{\langle \Phi_0 | \hat{H}' | \Phi_n \rangle \langle \Phi_n | \hat{H}' | \Phi_0 \rangle}{(E_0 - E_n)^2}.$$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The MBPT corrections expanded in a basis set

It is possible to rewrite the many-body energy corrections in particle and hole state formalism by using the expression of $\hat{H}'$ as expressed in terms of anihilation (c_k) and creation ($c_k^\dagger$) operators

$$\hat{H}' = \frac{1}{2} \sum_{ijkl} \langle ij|v|kl \rangle c_i^\dagger c_j^\dagger c_l c_k,$$

The previous many-body perturbation corrections now read

$$\Delta E^{(1)} = \langle \Phi_0 | \hat{H}' | \Phi_0 \rangle. = \frac{1}{2} \sum_{h_1 h_2} \langle h_1 h_2 | v | h_1 h_2 \rangle_{as}, \qquad (30)$$

$$\Delta E^{(2)} = \sum_{n=0}^{\infty} \frac{\langle \Phi_0 | \hat{H}' | \Phi_n \rangle \langle \Phi_n | \hat{H}' | \Phi_0 \rangle}{E_0 - E_n} = \frac{1}{4} \sum_{h_1 h_2 p_1 p_2} \frac{|\langle h_1 h_2 | v | p_1 p_2 \rangle|_{as}^2}{\epsilon_{h_1} + \epsilon_{h_2} - \epsilon_{p_1} - \epsilon_{p_2}},$$

where h_i and p_i are respectively hole states and particles states, and ϵ_i are the single particle energies of the basis set.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The MBPT corrections expanded in a basis set

The 3rd-order many-body perturbation correction reads

$$\Delta E^{(3)} = \sum_{n=0}^{\infty} \sum_{n=0}^{\infty} \frac{\langle \Phi_0 | \hat{H}' | \Phi_m \rangle \langle \Phi_m | \hat{H}' | \Phi_n \rangle \langle \Phi_n | \hat{H}' | \Phi_0 \rangle}{(E_0 - E_m)(E_0 - E_n)}$$

$$- \langle \Phi_0 | \hat{H}' | \Phi_0 \rangle \sum_{n=0}^{\infty} \frac{\langle \Phi_0 | \hat{H}' | \Phi_n \rangle \langle \Phi_n | \hat{H}' | \Phi_0 \rangle}{(E_0 - E_n)^2}$$

$$= \Delta E_{4p-2h}^{(3)} + \Delta E_{2p-4h}^{(3)} + \Delta E_{3p-3h}^{(3)}, \qquad (31)$$

where

- $\Delta E_{4p-2h}^{(3)}$ is the contribution to the third-order energy correction due to the 4-particle/2-hole excitations,

- $\Delta E_{2p-4h}^{(3)}$ is the contribution to the third-order energy correction due to the 2-particle/4-hole excitations,

- $\Delta E_{3p-3h}^{(3)}$ is the contribution to the third-order energy correction due to the 3-particle/3-hole excitations.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

The MBPT corrections expanded in a basis set

The contributions to the third-order energy correction can be written as

$$\Delta E_{4p-2h}^{(3)} = \frac{1}{8} \sum_{h_1 h_2 p_1 p_2} \left(\frac{\langle h_1 h_2 | v | p_1 p_2 \rangle_{as}}{\epsilon_{h_1} + \epsilon_{h_2} - \epsilon_{p_1} - \epsilon_{p_2}} \sum_{p_3 p_4} \frac{\langle p_1 p_2 | v | p_3 p_4 \rangle_{as} \langle p_3 p_4 | v | h_1 h_2 \rangle_{as}}{\epsilon_{h_1} + \epsilon_{h_2} - \epsilon_{p_3} - \epsilon_{p_4}} \right),$$

$$\Delta E_{2p-4h}^{(3)} = \frac{1}{8} \sum_{h_1 h_2 p_1 p_2} \left(\frac{\langle h_1 h_2 | v | p_1 p_2 \rangle_{as}}{\epsilon_{h_1} + \epsilon_{h_2} - \epsilon_{p_1} - \epsilon_{p_2}} \sum_{h_3 h_4} \frac{\langle h_1 h_2 | v | h_3 h_4 \rangle_{as} \langle h_3 h_4 | v | h_1 h_2 \rangle_{as}}{\epsilon_{h_3} + \epsilon_{h_4} - \epsilon_{p_1} - \epsilon_{p_2}} \right),$$

$$\Delta E_{3p-3h}^{(3)} = \sum_{h_1 h_2 p_1 p_2} \left(\frac{\langle h_1 h_2 | v | p_1 p_2 \rangle_{as}}{\epsilon_{h_1} + \epsilon_{h_2} - \epsilon_{p_1} - \epsilon_{p_2}} \left(\sum_{h_3} \sum_{p_3} \frac{\langle h_1 h_3 | v | p_1 p_3 \rangle_{as} \langle p_3 h_2 | v | h_3 h_2 \rangle_{as}}{\epsilon_{h_1} + \epsilon_{h_3} - \epsilon_{p_1} - \epsilon_{p_3}} \right) \right),$$

where the p_i denote the particle states, h_i the hole states, and ϵ_i the single particle energies of the corresponfing state.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Code implementation

Tools

- C++ language: for flexibility using classes and efficiency.
- BLITZ++ library: managing dense arrays.
- LPP / LAPACK library: (Fortran) routines for solving linear algebra problems.
- MESSAGE-PASSING INTERFACE: for parallel computing

Functionality

- Read parameters from a unique textual input file or command line.
- The simulator class performs the initialization and calls other objects.
- The orbitalsQuantumNumbers class: generates the harmonic oscillator states.
- The CoulombMatrix class generates the Coulomb interaction matrix outside Hartree-Fock.
- The HartreeFock class computes the HF energy and generates the interaction matrix in the HF basis.
- The PerturbationTheory class computes many-body perturbation corrections from 1st- to 3rd-order, either in the harmonic oscillator or in the HF basis set.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Model of a quantum dot
The method of Variational calculus
The Hartree-Fock method
The many-body perturbation theory
Implementation

Implementation issues

Difficulties encountered if not making use of symmetries

- Huge Fock matrix to diagonalize (grows exponentially with R^b).
- Huge Coulomb interaction matrix to store
  $V_{\alpha\beta\gamma\delta} = \langle (n_1, m_{l1})(n_2, m_{l2})|V|(n_3, m_{l3})(n_4, m_{l4}) \rangle$ is an 8-dimensional array.

Solutions implemented

By using the symmetries and properties of the Coulomb interaction:

- $V_{\alpha\beta\gamma\delta}$ does not act on spin: $m_{s1} = m_{s3}$ & $m_{s2} = m_{s4}$.
- $V_{\alpha\beta\gamma\delta}$ conserves the total spin and angular momentum: $m_{l1} + m_{l2} = m_{l3} + m_{l4}$.

By sorting the states of the basis by blocks of identical angular (m_l) and spin (m_s) quantum numbers:

- It allows to reduce the storage of the Coulomb interactions per blocks of couple of states by avoiding to store zeros's elements.
- The Fock matrix appears as block diagonal, allowing much smaller eigenvalue problems to solve.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Table of contents

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Validation of the simulator

Simple checks

- Reproduce the non-interacting ground state energies.
- Reproduce the two-body interaction matrix elements of OPENFCI almost with machine precision

Comparison of MBPT results with similar experiments

- Waltersson computed the open-shell 2nd-order MBPT correction.
- Our closed-shell 2nd-order MBPT correction shows close agreement.
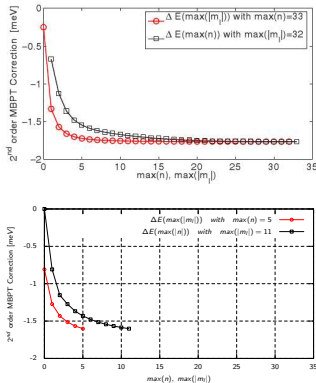
Figure 9: 2nd-order perturbation theory correction for the $2e^-$ QD. Comparison between results of Waltersson (top) [3] and our results (down).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

Fock-Darwin orbitals

When neglecting the repulsions between the particles, the eigenenergies $\epsilon_{n\,m_l}$ as a function of the magnetic field B can be solved analytically for a parabolic confining potential $V(r) = 1/(2m^*\omega_0^2 r^2)$ leading to a spectrum known as the Fock-Darwin states

Rewriting the eigenenergies in units of $\hbar\omega_0$, $\epsilon_{n\,m_l}$ becomes dimensionless and we obtain

$$\epsilon_{n\,m_l} = (2n + |m_l| + 1)\sqrt{1 + \frac{(\omega_c/\omega_0)^2}{4}} - \frac{1}{2}(\omega_c/\omega_0)\,m_l \qquad (32)$$

$$= (2n + |m_l| + 1)\sqrt{1 + (\frac{eB}{2m^*\omega_0})^2} - \frac{eB}{2m^*\omega_0}\,m_l. \qquad (33)$$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (2/3)
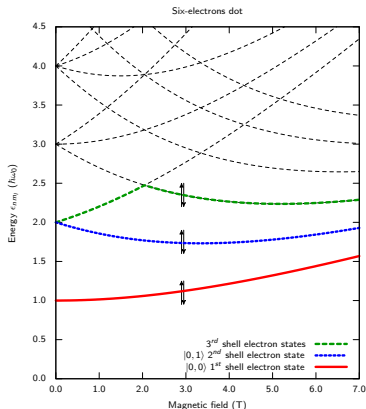
Figure 10: Spectrum of Fock-Darwin orbitals for 6 non-interacting particles (GaAs: $\hbar\omega_0 = 5meV, \epsilon_r = 12$).
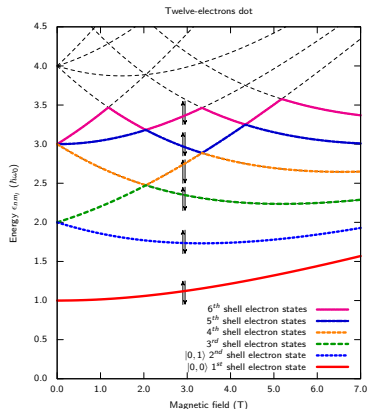
Figure 11: Spectrum of Fock-Darwin orbitals for 12 non-interacting particles (GaAs: $\hbar\omega_0 = 5meV, \epsilon_r = 12$).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
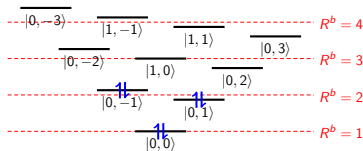
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
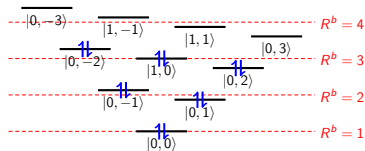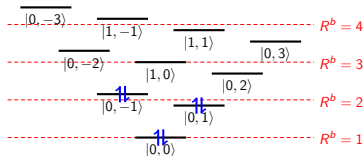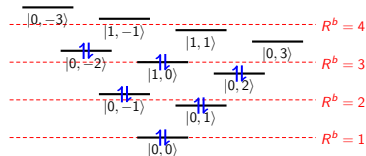Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
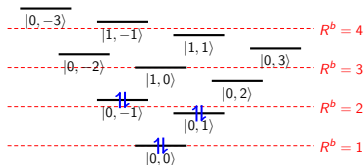
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles

Quantum Dots
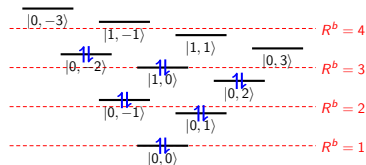Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
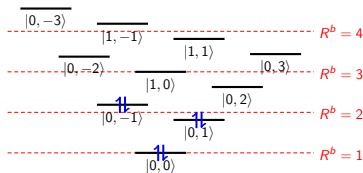
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
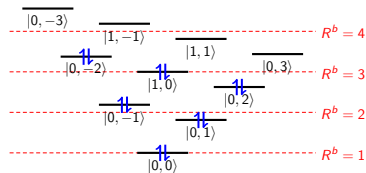Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
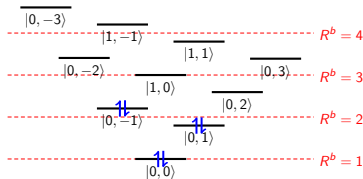
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
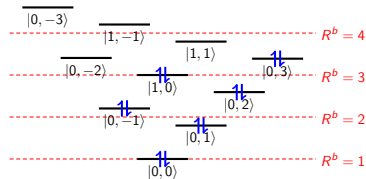Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
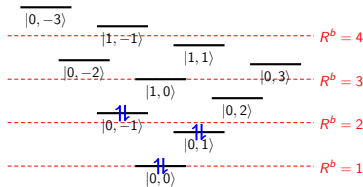
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles

Quantum Dots
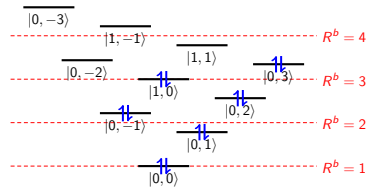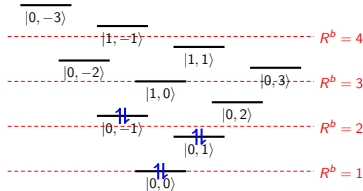Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles

Quantum Dots
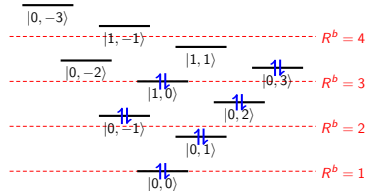Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
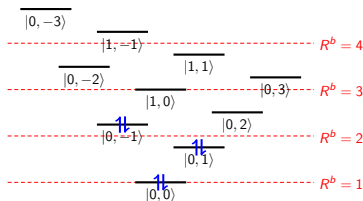
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
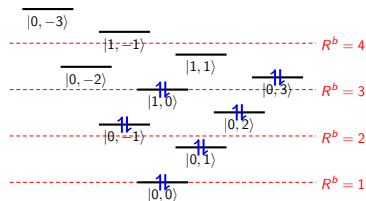Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
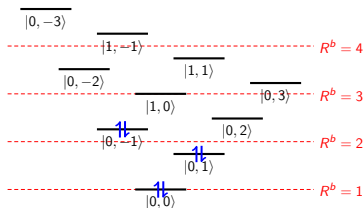
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles

Quantum Dots
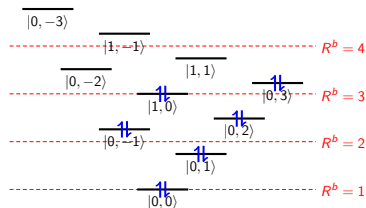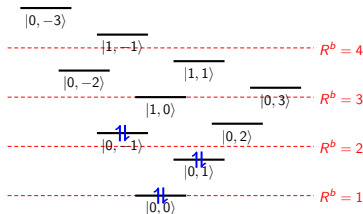Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
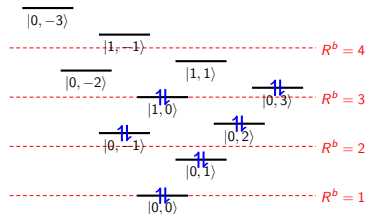Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
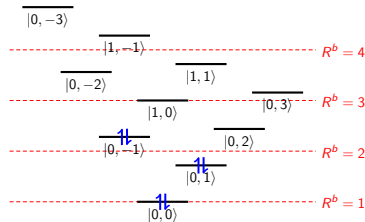
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
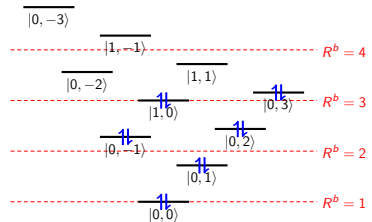Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
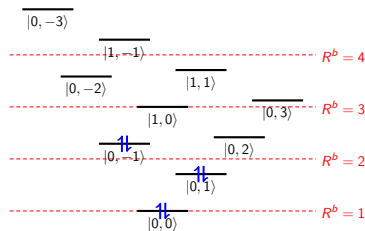Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles

Quantum Dots
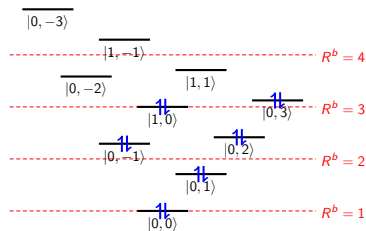Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
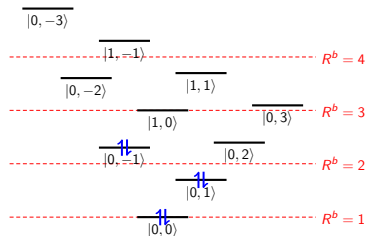
Level crossing as a function of B without interactions (1/3)

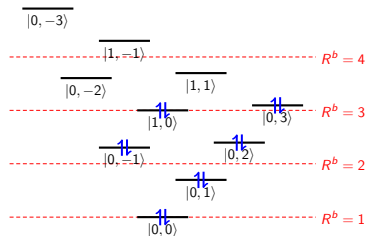6 non-interacting particles   12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
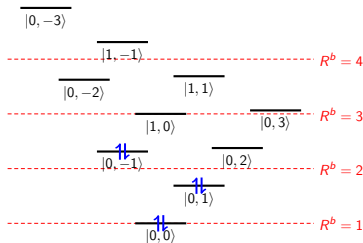
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
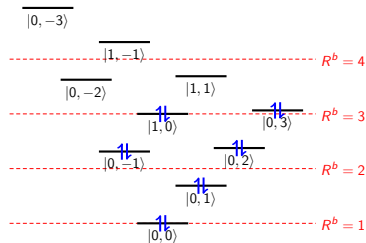Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
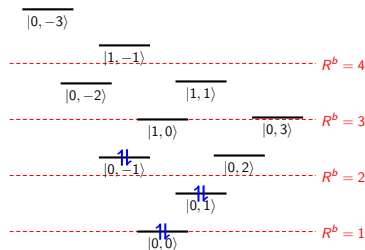
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
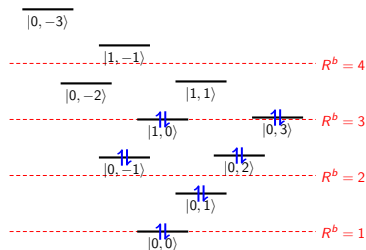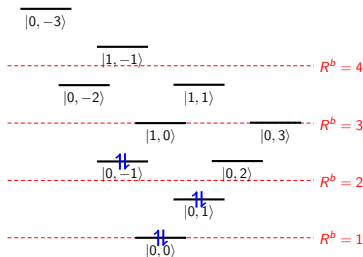Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
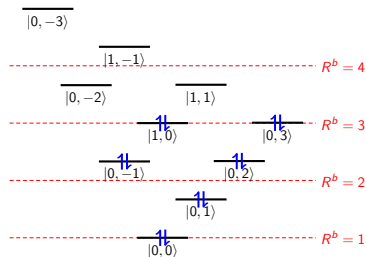Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
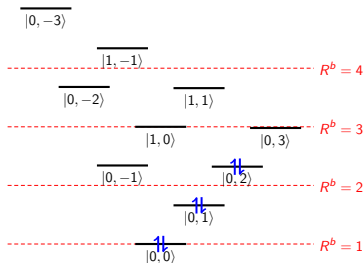Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles

Quantum Dots
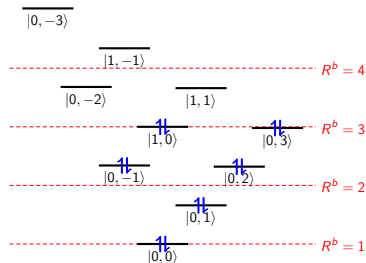Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
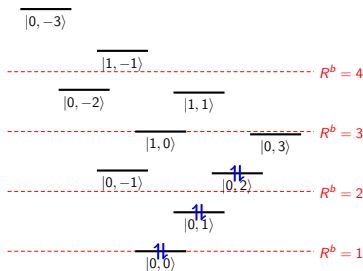
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
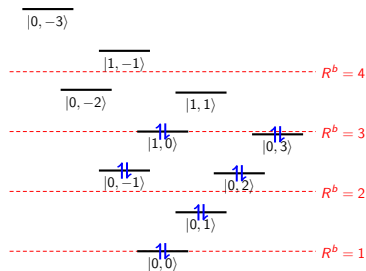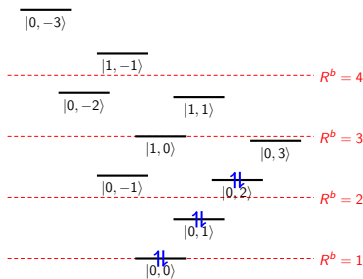Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles   12 non-interacting particles
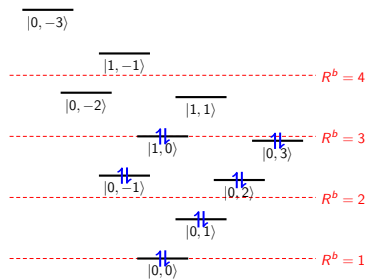
Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
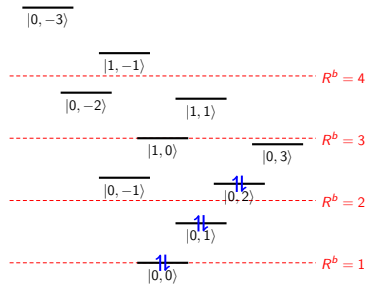
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
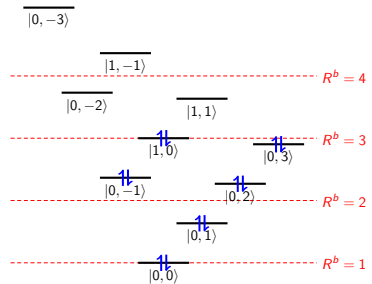Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
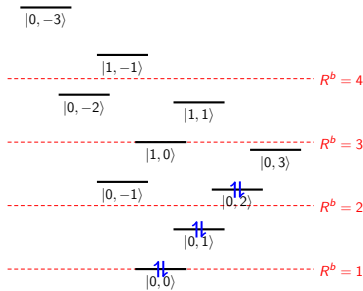
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

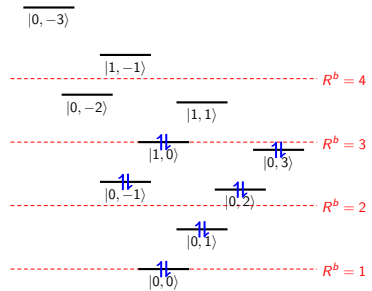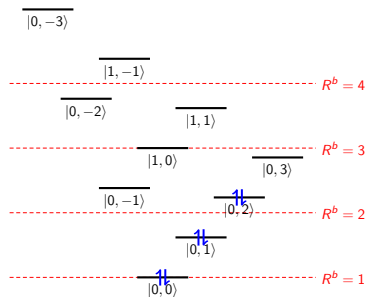6 non-interacting particles   12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
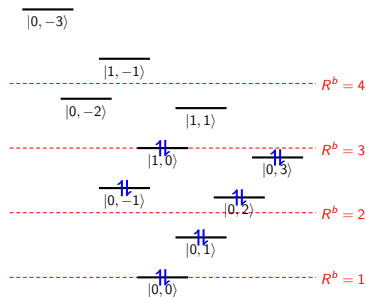Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
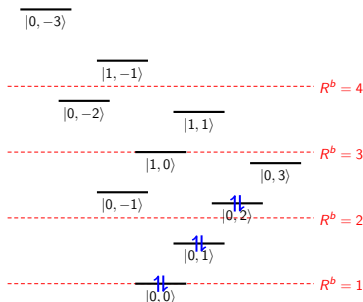Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

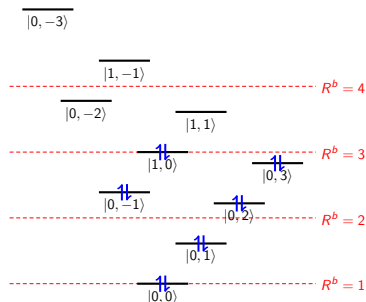6 non-interacting particles   12 non-interacting particles

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
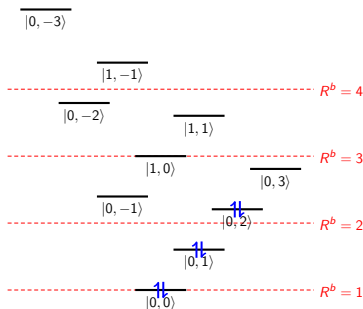
Level crossing as a function of B without interactions (1/3)

6 non-interacting particles

12 non-interacting particles

Quantum Dots
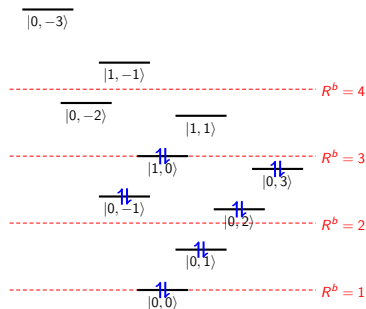Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of B without interactions (1/3)

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of λ with interactions (1/2)

Figure 12: FCI ground state energies for a 6-electron QD with $\lambda = \{0.1, 1\}$ (top) and $\lambda = \{15, 50\}$ (bottom).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Level crossing as a function of λ with interactions (2/2)
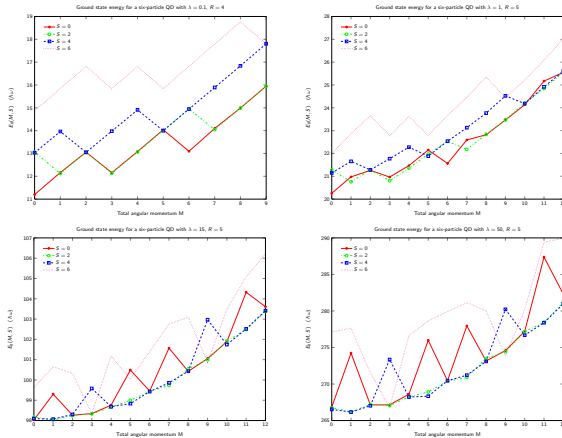
Summary of FCI results using OPENFCI

λ	FCI ground state energy ($\hbar\omega$) R=5	(M,S)
0.1	11.197	(0,0)
0.5	15.561	(0,0)
1	20.257	(0,0)
2	28.032	(0,0)
5	46.482	(0,0)
10	73.067	(0,0)
11	78.143	(0,0)
12	83.168	(0,0)
13	88.152	(0,0)
15	98.027	**(1,2)**
20	122.325	**(1,2)**
50	266.157	**(1,4)**

- For a 6-particle QD, break of the model of a single Slater determinant from $\lambda \simeq 13$.

- A similar study performed on a 2-particle QD indicates a break of the closed-shell model from $\lambda \simeq 150$.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Exponential convergence of HF as a function of R^b (1/3)

Definition

The size of the basis set characterized by R^b ($R^b \in \mathbb{N}$ $R^b \geq R^f$)
It defines the maximum shell number in the model space for our Hartree-Fock computation. It implies the number of orbitals in which each single particle wavefunction will be expanded, with spin degeneracy the number of states N_S is

$$N_S = (R^b + 1)(R^b + 2) \tag{34}$$

The bigger the basis set, the more accurate the single particle wavefunction is expected. In mathematical notation, R^b and the size of the basis set $\mathcal{B}$ are defined by

$$\mathcal{B} = \mathcal{B}(R^b) = \left\{ |\phi_{nm_l}(\mathbf{r})\rangle \quad : 2n + |m_l| \leq R^b \right\}, \tag{35}$$

where $|\phi_{nm_l}(\mathbf{r})\rangle$ are the single orbital in the Harmonic oscillator basis with quantum numbers n, m_l such that the single orbital energy reads: $\epsilon_{nm_l} = 2n + |m_l| + 1$ in two-dimensions.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Exponential convergence of HF as a function of R^b (2/3)
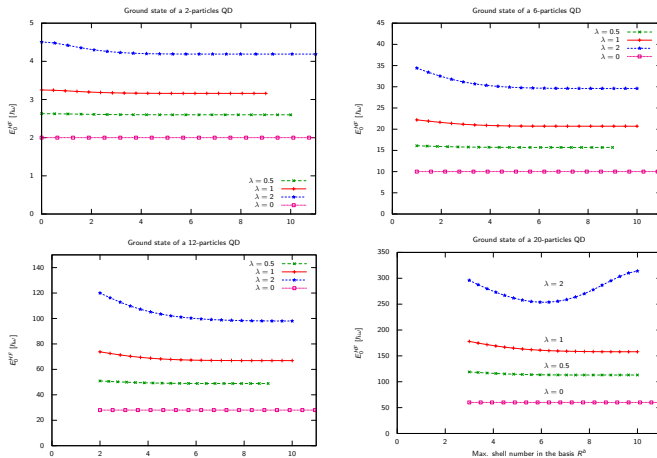

Figure 13: Hartree-Fock ground state (E^{HF} as a function of R^b for 2-,6-electron QD (top) and 12-,20-electron QD (bottom).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

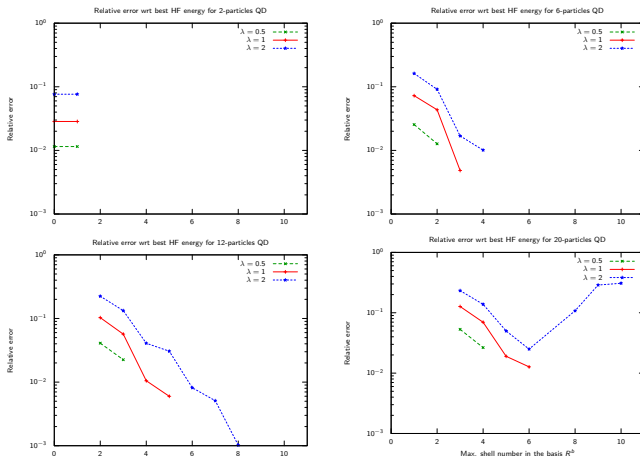# Exponential convergence of HF as a function of R^b (3/3)

Figure 14: Hartree-Fock relative error $(E^{HF}(R^b) - E^{HF}_{min})/E^{HF}_{min}$ as a function of R^b for 2-,6-electron QD (top) and 12-,20-electron QD (bottom).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

"Convergence history" as a function of λ (1/3)

Definition

The convergence history of a simulation shows how the convergence "improve" over iterations. We could plot

- the energy difference from one iteration to the next

$$\delta(iter) = |E^{HF}(iter) - E^{HF}(iter - 1)|.$$

- more intuitive on the form: $\delta(iter) \simeq 10^{-\beta \, iter}$.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

"Convergence history" as a function of λ (2/3)

- slower convergence as λ increases.
- much less impact due to R^b or to the nb.of particles, except when leading to unstability.
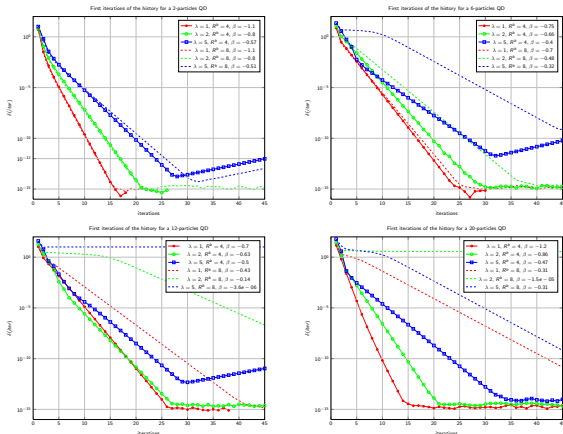
Figure 15: Convergence history of the Hartree-Fock iterative process for 2-,6-electron QD (top) and 12-,20-electron QD (bottom).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

"Convergence history" as a function of λ (3/3)

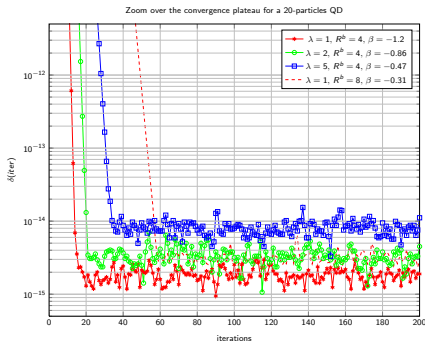Zoom over the convergence plateau for a 20-particles QD

Figure 16: Zoom over the limit of convergence of the Hartree-Fock iterative process for a 20-particle QD.

- expected "plateau" when reaching machine precision.

However it seems that increasing the interaction strength:

- slows down the convergence process, as adding error at each iteration,

- induces a lower accuracy, as if it could "decrease the machine precision",
  $\Rightarrow$ Phenomena maybe due to round-off error, proportional to λ, and entering the eigenvalue solver in a non-trivial way.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

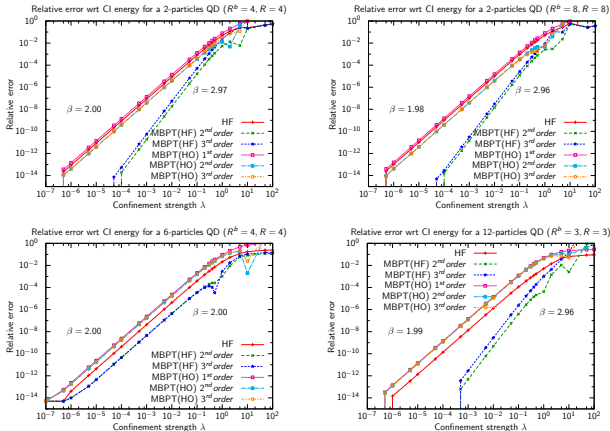# Quadratic error growth of HF/MBPT wrt FCI ground state

Figure 17: Comparison of HF/MBPT and HF corrected by MBPT up to 3rd-order wrt to the FCI ground state taken as reference for 2-electron QD (top) and 6-,12-electron QD (bottom).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations
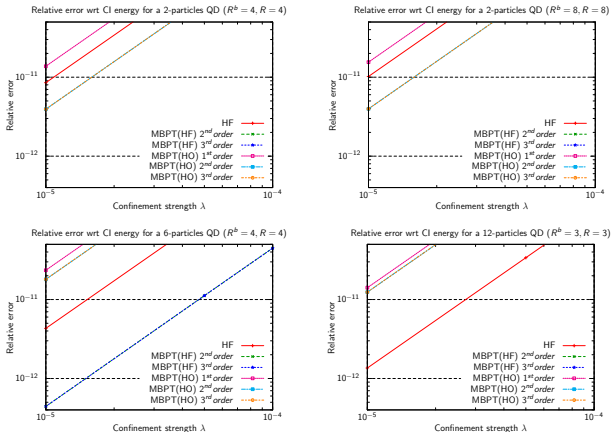
Respective accuracy of HF and MBPT (1/2)

Figure 18: Zoom on the quadratic growth of the error when λ is relatively small ($\lambda < 0.05$), showing different accuracies with respect to the method, the number of particles and the size of the basis. for 2-electron QD (top) and 6-,12-electron QD (bottom).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

$\sharp\, e^-$	R^{FCI}	R^{HF}	Relative error shift between each method for $\lambda = 10^{-3}$	
2	4	4	MBPT(HF)-2nd order	$\rightarrow \epsilon_{min}$
			MBPT(HF)-3rd order	$\rightarrow 3\,\epsilon_{min}$
			MBPT(H0)-2nd order and MBPT(H0)-3rd order	$\rightarrow 2.1 \times 10^3\,\epsilon_{min}$
			HF	$\rightarrow 4.6 \times 10^3\,\epsilon_{min}$
			MBPT(HO)-1st order	$\rightarrow 7.5 \times 10^3\,\epsilon_{min}$
	8	8	MBPT(HF)-2nd order and MBPT(HF)-3rd order	$\rightarrow \epsilon_{min}$
			MBPT(H0)-2nd order and MBPT(H0)-3rd order	$\rightarrow 2.7 \times 10^3\,\epsilon_{min}$
			HF	$\rightarrow 7 \times 10^3\,\epsilon_{min}$
			MBPT(HO)-1st order	$\rightarrow 10.7 \times 10^3\,\epsilon_{min}$
6	4	4	MBPT(HF)-2nd order and MBPT(HF)-3rd order	$\rightarrow \epsilon_{min}$
			HF	$\rightarrow 9.65\,\epsilon_{min}$
			MBPT(H0)-2nd order and MBPT(H0)-3rd order	$\rightarrow 40.3\,\epsilon_{min}$
			MBPT(HO)-1st order	$\rightarrow 52.55\,\epsilon_{min}$
	4	8	MBPT(HF)-2nd order and MBPT(HF)-3rd order	$\rightarrow \epsilon_{min}$
			HF	$\rightarrow 1.31\,\epsilon_{min}$
			MBPT(H0)-2nd order and MBPT(H0)-3rd order	$\rightarrow 5.47\,\epsilon_{min}$
			MBPT(HO)-1st order	$\rightarrow 8\,\epsilon_{min}$
12	3	3	MBPT(HF)-2nd order	$\rightarrow \epsilon_{min}$
			MBPT(HF)-3rd order	$\rightarrow 5.98\,\epsilon_{min}$
			HF	$\rightarrow 5.6 \times 10^3\,\epsilon_{min}$
			MBPT(H0)-2nd order and MBPT(H0)-3rd order	$\rightarrow 51.3 \times 10^3\,\epsilon_{min}$
			MBPT(HO)-1st order	$\rightarrow 58.4 \times 10^3\,\epsilon_{min}$

Table 1: Classification of the methods with respect to their relative accuracy in the range of λ that exhibits a quadratic error growth.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
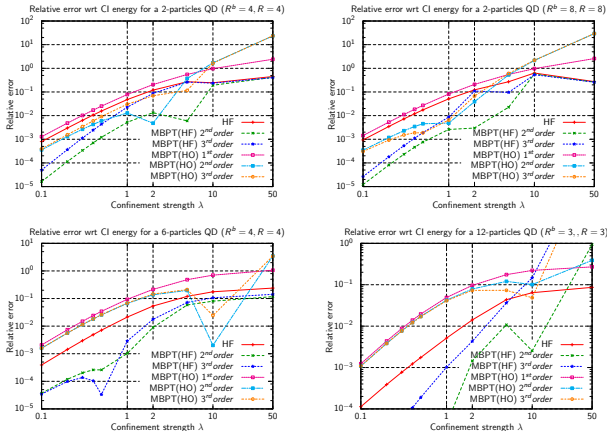**Comparison on HF/MBPT/FCI calculations**

Break of the methods

Figure 19: The plots display a zoom for λ approaching the limit of the closed-shell model for 2-electron QD (top) and 6-,12-electron QD (bottom).

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Summary of the results

- Exponential convergence of HF as a function of R^b.
- Increasing λ slows down the convergence of HF, and decreases its accuracy.
- Compared to FCI, HF and MBPT have a quadratic error growth wrt λ.
- Unstability of the 2nd- 3rd-order MBPT corrections before 1st-order MBPT and HF.

Break of the method before the closed-shell model

# particles	Break of the method	Break of the model
2	$\lambda \simeq 5$	$\lambda \simeq 150$
6	$\lambda \simeq 2$	$\lambda \simeq 14$
12	$\lambda \simeq 1$	$\lambda \simeq ???$

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

Thank you for your attention ;)

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

L. P. Kouwenhoven. *Electron transport in quantum dots*, Proceedings of the Advanced Study Institute, 1997.

X. Gao, Y. Cui, R. M. Levenson, L. W. Chung, and S. Nie. In vivo cancer targeting and imaging with semiconductor quantum dots. *Nat Biotech*, 22:969–976, 2004.

L. P. Kouwenhoven, D. G. Austing, and S. Tarucha. Few-electron quantum dots. *Reports on Progress in Physics*, 64:701–736, 2001.

S. Kvaal. Open source fci code for quantum dots and effective interactions. *arXiv.org*, 2008.

S. Kvaal. *Analysis of many-body methods for quantum dots*. PhD thesis, University of Oslo, 2009.

M. Moshinsky and Y. F. Smirnov. *The Harmonic Oscillator in Modern Physics: From Atoms to Quarks*. Taylor & Francis, 1996.

Quantum Dots
Model, Methods and Implementation
Results and Discussions

Validation of the simulator
Limit of the closed-shell model as a function of λ
Convergence/Stability/Accuracy of HF
Comparison on HF/MBPT/FCI calculations

D. Pfannkuche, V. Gudmundsson, and P. A. Maksym. Comparison of a hartree, a hartree-fock, and an exact treatment of quantum-dot helium. *Phys. Rev. B*, 47:2244–2250, 1993.

S. Raimes. *Many-electron Theory*. North-Holland Publishing, 1972.

E. Waltersson and E. Lindroth. Many-body perturbation theory calculations on circular quantum dots. *Phys. Rev. B*, 76:045314, 2007.

J. O. Winter. *Development and optimization of quantum dot-neuron interfaces*. PhD thesis, The University of Texas at Austin, 2004.