Computational Methods for Studies of Quantum Dots and Boson Condensates

Dissertation for the degree of Doctor Philosophiae

Tommy Vänskä
University of Helsinki
Faculty of Science
Department of Chemistry
Laboratory for Instruction in Swedish
P.O. Box 55 (A.I. Virtanens plats 1)
FI-00014 University of Helsinki, Finland

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In the beginner’s mind there are many possibilities, but in the expert’s mind there are few.

–Shunryu Suzuki-roshi
Abstract

This thesis presents *ab initio* studies of two kinds of physical systems, quantum dots and bosons, using two program packages of which the bosonic one has mainly been developed by the author. The implemented models, *i.e.*, configuration interaction (CI) and coupled cluster (CC) take the correlated motion of the particles into account, and provide a hierarchy of computational schemes, on top of which the exact solution, within the limit of the single-particle basis set, is obtained. The theory underlying the models is presented in some detail, in order to provide insight into the approximations made and the circumstances under which they hold. Some of the computational methods are also highlighted. In the final sections the results are summarized. The CI and CC calculations on multiexciton complexes in self-assembled semiconductor quantum dots are presented and compared, along with radiative and non-radiative transition rates. Full CI calculations on quantum rings and double quantum rings are also presented. In the latter case, experimental and theoretical results from the literature are re-examined and an alternative explanation for the reported photoluminescence spectra is found. The boson program is first applied on a fictitious model system consisting of bosonic electrons in a central Coulomb field for which CI at the singles and doubles level is found to account for almost all of the correlation energy. Finally, the boson program is employed to study Bose-Einstein condensates confined in different anisotropic trap potentials. The effects of the anisotropy on the relative correlation energy is examined, as well as the effect of varying the interaction potential.
List of Publications

List of original publications included in the thesis


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Of all the people I have had the pleasure of working with one stands above the rest, if not in stature then in importance. My supervisor Dage Sundholm is the initiator and mastermind behind the research projects I have been working on, and I can only hope that I haven’t done his original ideas too much injustice. I quite simply cannot imagine a better supervisor than Dage; his unwavering enthusiasm for what he does is only paralleled by his ability to find the time in an often very busy schedule to listen and to help. Innumerable are the occasions that I have been stuck with a problem, large or small, finally asked Dage for help, and witnessed him patiently scribble down the solution on the backside of some scrappy piece of paper salvaged from the mess of my desk.

Another person without whom this thesis certainly would not exist, in a very literal sense, is my old friend Jonas Jusélius, who introduced me to Dage and suggested I join his group at the University of Helsinki. At the time I was a few years into a PhD at the University of Technology, but having lost my supervisor and struggling with the funding, I was extremely grateful to have been given a chance at a fresh start. I have also engaged in quite a few scientific discussions with Jonas over the years, some of them at his summer cottage in the archipelago (a finer setting for any discussion is hard to find), and I hope that these, and other discussions not even remotely related to science have been mutually fruitful.

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something vastly more important, as this is where I met my wife!

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<tr>
<td>BEC</td>
<td>Bose-Einstein Condensate</td>
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<tr>
<td>CC</td>
<td>Coupled cluster</td>
</tr>
<tr>
<td>CDQR</td>
<td>Concentric double quantum ring</td>
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<tr>
<td>CI</td>
<td>Configuration interaction</td>
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<tr>
<td>EMA</td>
<td>Effective mass approximation</td>
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<tr>
<td>FCI</td>
<td>Full CI</td>
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<tr>
<td>GP</td>
<td>Gross-Pitaevskii</td>
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<tr>
<td>HF</td>
<td>Hartree-Fock</td>
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<tr>
<td>HH</td>
<td>Heavy hole</td>
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<tr>
<td>LH</td>
<td>Light hole</td>
</tr>
<tr>
<td>LK</td>
<td>Luttinger-Kohn</td>
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<tr>
<td>PL</td>
<td>Photoluminescence</td>
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<tr>
<td>SCF</td>
<td>Self consistent field</td>
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<tr>
<td>SE</td>
<td>Schrödinger Equation</td>
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<tr>
<td>QD</td>
<td>Quantum dot</td>
</tr>
<tr>
<td>SAQD</td>
<td>Self-assembled Quantum dot</td>
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<td>QW</td>
<td>Quantum well</td>
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1 Introduction

In the sciences a rather rigid division of labor is often encountered, in the sense that certain problems are assigned to certain disciplines or subdisciplines. This is, for the most part, a well-founded practise which ensures that research is focused and that the right kind of know-how is appropriately concentrated where needed. Indeed, in some cases the results of a less rigid compartmentalization could be nothing short of disastrous; few people in the dentist’s chair would appreciate a geologist entering the room with her nuclear physicist assistant. The historical development from the ancient philosophers – whose area of interest included everything! – through the scientific revolution to the compartmentalized, subcompartmentalized and subsubcompartmentalized (and so on) structure of the scientific community today has been largely beneficial to the advancement of science by practically any measure.

One might ask, however, whether there is a dark side to ultraspecialization. Is something lost when the overwhelming majority of scientists are focused on very specific problems in extremely narrow fields of investigation? Sidestepping the more philosophical aspects of the question, at least this may be said: in the murky borderlands between disciplines scientists from different compartments may be found, all investigating the same phenomena, using different instruments and – importantly – different language. They are also largely unaware of, or indifferent to, each other’s presence.

Someone who has attended conferences and meetings in both physics and (quantum) chemistry would certainly recognize this phenomenon. Although there are vast areas of study which are firmly, and with good reason, placed in either field, there is also a significant area of overlap. Physics has traditionally been concerned with atoms and subatomic phenomena, as well as the solid state. Chemistry, broadly speaking, operates in the intermediate realm, studying molecular systems: intermolecular interactions, chemical reactions, and the underlying molecular electronic structure. As no fundamental distinction exists between the atomic and the molecular, or the molecular and bulk material, it follows that there are systems for which it is not clear which group ought to be assigned the task to investigate.

In this thesis methods originally developed for modeling molecular electronic structure are applied to systems mainly investigated by the physics community, using different approaches. The studied systems are quantum dots and rings as well as Bose-Einstein condensates. The methods used for modeling these systems are ones which account for the correlated motion of the particles involved, fermionic electrons and holes in the former case, and bosonic atoms in the latter. In chemistry, correlation effects – while quantitatively small – play a crucial role in correctly describing electronic structure. An extensive toolbox of methods has therefore been developed over the past decades, providing a hierarchic structure of models which makes it possible to approach the exact solution in a systematic way. The theoretical work has been accompanied by an extensive development of computational methods, designed to make the most of hitherto ever growing but also always finite computational resources. While it is possible to solve the Schrödinger equation virtually exactly for systems of several interacting particles there is still a limit for even today’s most
powerful computers, and models further down in the hierarchy must be employed.

The configuration interaction method employed in this thesis is conceptually quite simple: given a single-particle basis the Hamiltonian matrix is constructed in a many-body basis spanned by that single-particle basis. If the whole many-body space is used the exact solution within the limits of the single-particle basis is obtained upon diagonalizing the Hamiltonian. If this task is computationally unfeasible the variational space is restricted. The coupled cluster model included in the quantum dot software is similar, but uses a different, more complicated way to limit the many-body space.

The aim of this work is to investigate the importance of correlation in the studied systems and to, hopefully, make the methods more widely known, so that they may be applied by others when deemed appropriate. To this end the quantum-dot program has been applied to calculate energies and recombination rates of multiexciton complexes in self-assembled strain-induced quantum dots as well as quantum rings and concentric double-quantum-ring systems. The boson software has been tested on fictitious atom-like systems and then applied to Bose-Einstein condensates with various trap geometries.
Part I

Theory
2 Approximate Solution of Schrödinger equations

The physical world consists of interacting many-particle systems. In principle, all possible information about a system of $N$ particles is contained in the many-body wave function $\Psi(x; t)$, where $x$ represents the coordinates, corresponding to the system’s degrees of freedom, e.g., the spatial coordinates $r_1, r_2, \ldots, r_N$ and the spins $\sigma_1, \sigma_2, \ldots, \sigma_N$. The wave function is determined by the time-dependent Schrödinger equation, which in general form reads (in SI units)

$$\hat{H}\Psi(x; t) = i\hbar \frac{\partial}{\partial t}\Psi(x; t), \quad (2.1)$$

where the Hamiltonian operator $\hat{H}$ takes the following form in almost all cases of interest:

$$\hat{H} = \sum_{p=1}^{N} \hat{T}(x_p) + \hat{V}_{\text{ext}}(x_p) + \sum_{p>q}^{N} \hat{V}(x_p, x_q), \quad (2.2)$$

where $\hat{T}$ is the kinetic energy operator, $\hat{V}_{\text{ext}}$ is the external potential and $\hat{V}$ is the potential energy operator of interaction between the particles. For all but a few systems of practical interest solving Eq. (2.1) or, indeed, even the time-independent version directly is an intractable task. Obtaining $\Psi$ or some more limited set of properties of the system therefore requires approximations. One may divide the approximations into two classes: first, the Hamiltonian (and thereby the SE) can be approximated by a simpler one that is more accessible to solution. Second, if the equations with the approximative Hamiltonian are not solvable analytically, some approximative, often numerical scheme needs to be employed. Typically, then, an approximate solution to an approximate SE is as good as it gets, and the quality and understanding of the approximations and the solution schemes are the decisive factors in modelling microscopic systems.

The appropriate approximation for the Hamiltonian obviously depends on the system under study. For example, in most cases it makes little sense to include relativistic effects in describing a system of particles whose velocities are much smaller than the speed of light. Likewise, it is usually wise to consider the motion of atomic nuclei separately from the motion of electrons, due to the difference in mass of several orders of magnitude (the Born-Oppenheimer approximation). Section 4.2 provides a rather detailed description of how the model employed for describing strain-induced semiconductor quantum dots is constructed.

Having settled for a Hamiltonian that captures the essential physics of the system, choosing an approximation scheme for treating the resulting equations is perhaps not as straight-forward. Several different approaches to solving the SE approximately

\cite{In a more fundamental formulation the external potential term might not be included, since any “external” field arises from interactions between particles.}

\cite{The systems studied in this thesis exhibit no explicit time dependence, and we are thus concerned with the time-independent Schrödinger equation.}
have been developed since the conception of quantum mechanics, each with its own advantages and shortcomings. Some of the commonly used general techniques include perturbation theory, quantum Monte Carlo schemes, density functional theory, coupled cluster theory, and variational approaches.\footnote{Except for the coupled cluster model which, in most implementations, is not variational.}

The method of choice depends at least partly on the scientific field within which the research is conducted. Although quantum chemistry and quantum physics in principle operate within the same realm it is clear that chemists and physicists have their own preferred set of tools. This can partly be explained by the fact that chemistry has traditionally been concerned with molecular systems, whereas the realm of quantum physics in a sense surrounds the molecular world, comprising the solid state on one flank and the atomic and subatomic world on the other. The advent of nano- and mesoscopic science, however, has rendered the historical separation of the two fields somewhat obsolete.

Thus the methods employed in this thesis are ones that have been primarily developed and used for modelling molecular systems, although the systems we study may be said to lie within the field of physics. The methods are based upon the variational principle\footnote{Except for the coupled cluster model which, in most implementations, is not variational.}, and rely upon expanding the wave function in a basis of many-body states generated by a finite basis of single-particle functions.

### 2.1 Many-body states in Fock space and symmetry

Expressing the many-body wave function in terms of single-particle states presents a notational problem, which is elegantly dealt with using the formalism of second quantization. Within this formalism states are represented as vectors in an abstract vector space $\mathcal{F}$, the Fock space. Let $\mathcal{H}$ be a Hilbert space spanned by the single-particle basis set $\{\phi_i\}$, which may be finite or infinite in size. The Fock space $\mathcal{F}$ generated by $\{\phi_i\}$ may then be defined as

$$\mathcal{F} = \bigoplus_{n=0}^{\infty} \hat{S}_n \mathcal{H}^\otimes n = \mathcal{F}_0 \oplus \mathcal{H} \oplus \hat{S}_2(\mathcal{H} \otimes \mathcal{H}) \oplus \hat{S}_3(\mathcal{H} \otimes \mathcal{H} \otimes \mathcal{H}) \oplus \ldots.$$  \hspace{1cm} (2.3)

The space $\mathcal{F}$ is thus a direct sum of $N$-particle subspaces (Hilbert spaces) $\mathcal{F}_N = \hat{S}_N \otimes_{n=0}^{N} \mathcal{H}$, including $\mathcal{F}_0$, which is the space containing no particles, i.e., the vacuum. The operator $\hat{S}_\nu$ either symmetrizes ($\nu = 1$) or antisymmetrizes ($\nu = -1$) the space $\mathcal{F}_N$.

All operators and states in Fock space can be constructed from a set of elementary creation and annihilation operators, the properties of which can be derived from their commutation or anticommutation relations. Particles for which the Fock space is symmetric are called bosons and the commutation relations for the bosonic operators $b_p$ and $b_p^\dagger$ and are

$$[b_p, b_q^\dagger] = b_p b_q^\dagger - b_q^\dagger b_p = \delta_{pq},$$  \hspace{1cm} (2.4)

$$[b_p, b_q] = [b_p^\dagger, b_q^\dagger] = 0,$$  \hspace{1cm} (2.5)

where the indices $p$ and $q$ refer to single-particle states, or modes. The operators $a_p$ and $a_q^\dagger$ for fermions, i.e., particles inhabiting the antisymmetric space, fulfill the following anticommutation relations:

$$\{a_p, a_q^\dagger\} = a_p a_q^\dagger + a_q^\dagger a_p = \delta_{pq},$$  \hspace{1cm} (2.6)

$$\{a_p, a_q\} = \{a_p^\dagger, a_q^\dagger\} = 0.$$  \hspace{1cm} (2.7)

The operator $\hat{n}_i = c_i^\dagger c_i$ (with $c = a, b$) is called the number operator for the mode $i$, and it follows from Eqs. (2.4) and (2.7) that $[n_i, n_j] = 0$. Therefore, there exist states...
which are the simultaneous eigenstates of the number operators for all modes. We denote these occupation number (ON) states, or configurations |n⟩ = |n₁, n₂, . . .⟩. Operating on |n⟩ with nᵢ yields:

\[ \hat{c}^\dagger \hat{c} |n₁, n₂, . . ., nᵢ , . . .⟩ = nᵢ |n₁, n₂, . . ., nᵢ , . . .⟩, \]

where the eigenvalue nᵢ is an integer corresponding to the number of particles occupying the single-particle state φᵢ. From the relations (2.13), (2.14) and (2.15) it can be shown that for bosons the spectrum of the number operator is nᵢ = {0, 1, 2, . . .} and for the fermionic case nᵢ = {0, 1}. This is an expression of the Fermi exclusion principle, which states that no two fermions can occupy the same single-particle state. For bosons no such restriction exists.

ON-states are not eigenstates of the creation and annihilation operators. Rather, denoting \( |n_p⟩ = |n₁, . . ., nᵢ , . . ., n_p , . . .⟩ \), we have for bosons

\[ b_p |n_p⟩ = (n_p)^{1/2} |n_p - 1⟩ \]
\[ b_p^\dagger |n_p⟩ = (n_p + 1)^{1/2} |n_p + 1⟩, \]

and for fermions

\[ a_p |n_q⟩ = \begin{cases} S_p(n_p)^{1/2} |n_p - 1⟩, & n_p = 1 \\ 0, & \text{otherwise} \end{cases} \]
\[ a_p^\dagger |n_p⟩ = \begin{cases} S_p(n_p + 1)^{1/2} |n_p + 1⟩, & n_p = 0 \\ 0, & \text{otherwise} \end{cases} \]

where the factor \( S_p \) is a phase factor, defined below. The operator \( \hat{c}^\dagger \) thus decreases the number of particles in the mode i by one and \( \hat{c}^\dagger \) increases the occupation by one particle in the corresponding mode. Using the relations (2.9)–(2.12) an ON-state may then be constructed from the vacuum state |0⟩ as follows:

\[ |n₁, n₂, . . ., n∞⟩ = \frac{1}{n₁!n₂! . . . n∞!} \left( \hat{c}^\dagger \right)ⁿ₁ \left( \hat{c}^\dagger \right)ⁿ₂ . . . \left( \hat{c}^\dagger \right)ⁿ∞ |0⟩, \]

where an infinite number of modes is assumed. In some cases – and certainly in practice, when working with finite-sized spaces – the number of modes is some finite \( n_{\text{max}} \). The prefactor in Eq. (2.13), which in the fermionic case is unity, ensures the proper normalization of the state |n⟩. From Eq. (2.13) and the fermionic commutation relations it follows that the phase factor in Eqs. (2.11)–(2.12) is \( S_p = (-1)^{\sum_{i=1}^p n_i} \). From Eq. (2.13) it can also be seen that permuting two particles \( p \leftrightarrow q \) in an ON-state has the following effect:

\[ |n₁, . . ., n_p , . . ., n_q , . . .⟩ = |n₁, . . ., n_q , . . ., n_p , . . .⟩ \text{ bosons} \]
\[ |n₁, . . ., n_p , . . ., n_q , . . .⟩ = -|n₁, . . ., n_q , . . ., n_p , . . .⟩ \text{ fermions} \]

The relations (2.14) and (2.15) express a fundamental property of many-body states, i.e., the bosonic (fermionic) states are symmetric (antisymmetric) under the permutation of two particles. The operator \( \hat{S}_b \) in Eq. (2.3) then symmetrizes (antisymmetrizes) the Fock space for bosons (fermions). It is instructive to see what this means within the formalism of first quantization. Consider, then, a state in which N particles occupy the N single-particle states \{ϕ₁, ϕ₂, . . ., ϕₐₙ \}. The symmetry conditions stated above then require that the bosonic state \( \Psi_b(x) = \Psi_b(x₁, x₂, . . ., x_N) \) is of the form

\[ \Psi_b(x) = \hat{S}_b^+ \left[ \prod_{i=1}^{N} \phiₐₜ(xᵢ) \right] = \frac{1}{\sqrt{N!}} \sum_P \prod_{i=1}^{N} \phiₐₜ(Pxᵢ) \]

\(^\dagger\)For fermions a ≠ b ≠ . . . ≠ aₙ.
where the sum runs over all permutations $P$ of the string $(123 \ldots N)$. The fermionic counterpart $\Psi_f(x)$ is written

$$\Psi_f(x) = \hat{S}_{-1} \prod_{i=1}^{N} \phi_{A_i}(x_i) = \frac{1}{\sqrt{N!}} \sum_{P} (-1)^P \prod_{i=1}^{N} \phi_{A_i}(P x_i),$$

(2.17)

where the factor $(-1)^P$ is positive if the permutation $P$ is even and negative if it is odd. Equation (2.17) may also be expressed in the form of a Slater determinant as follows:

$$\Psi_f(x) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \phi_a(x_1) & \phi_b(x_1) & \ldots & \phi_{A_N}(x_1) \\ \phi_a(x_2) & \phi_b(x_2) & \ldots & \phi_{A_N}(x_2) \\ & \vdots & \ddots & \vdots \\ \phi_1(x_N) & \phi_2(x_N) & \ldots & \phi_{A_N}(x_N) \end{vmatrix}. \quad (2.18)$$

As mentioned above, operators in Fock space are also constructed with creation and annihilation operators. For our purposes it is sufficient to state the main result, i.e., we express the generic many-body Hamiltonian in Eq. (2.2) in the second-quantized form, which reads

$$\hat{H} = \sum_{pq} c^+_p c_q T_{pq} + \frac{1}{2} \sum_{pqrst} c^+_p c^+_q c_s c_t V_{pqrst}. \quad (2.19)$$

The matrix elements $T_{pq}$ and $V_{pqrst}$ are plain numbers, and are defined as

$$T_{pq} \equiv \langle p|\hat{T}|q \rangle = \int \phi^*_p(x_1)\hat{T}(x_1)\phi_q(x_1)dx_1 \quad (2.20)$$

$$V_{pqrst} \equiv \langle pq|\hat{V}|rs \rangle = \int \phi^*_p(x_1)\phi^*_q(x_2)\hat{V}(x_1, x_2)\phi_r(x_1)\phi_s(x_2)dx_1dx_2. \quad (2.21)$$

where \{\phi_i(x)\} is a set of single-particle states. Note the ordering of the annihilation operators in the two-particle term in Eq. (2.21), which is opposite that of the last two wave functions in the matrix element (2.20). For bosons the order is arbitrary, but for fermions the ordering affects the overall sign. Note also that the two-particle matrix element in Eq. (2.21) is written in physicists’ notation; the chemist community uses the notation $V_{pqrst} = \int \phi^*_p(x_1)\phi_q(x_1)\hat{V}(x_1, x_2)\phi^*_r(x_2)\phi_s(x_2)dx_1dx_2$.

Summarizing: in this section the formalism of second quantization was briefly reviewed, allowing us to express the many-body Hamiltonian in terms of creation and annihilation operators in Fock space. The difference between fermionic and bosonic systems is contained in the algebra of the corresponding creation and annihilation operators expressed in Eqs. (2.4)–(2.7), which leads to the following properties:

- **Bosons**: the spectrum of the number operator is \{0, 1, 2, \ldots\} and the wave function is symmetric under permutation of two particles.
- **Fermions**: the spectrum of the number operator is \{0, 1\} and the wave function is antisymmetric under permutation of two particles.

### 2.2 The variational principle

The variational principle provides a simple and powerful procedure for generating approximate wave functions. Let $|\Psi\rangle$ be any normalizable wave function. The variational principle states that

$$E[\Psi] = \frac{\langle \Psi | \hat{H} | \Psi \rangle}{\langle \Psi | \Psi \rangle}. \quad (2.22)$$

The two-body matrix element is expressed in so-called “chemist’s notation”, with the first two indices referring to $x_1$ and the other two to $x_2$. 

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*The two-body matrix element is expressed in so-called “chemist’s notation”, with the first two indices referring to $x_1$ and the other two to $x_2.$
2.3 The Hartree-Fock approximation

provides an upper bound on the ground state energy $E_0$ of the system. Thus, taking $|\Psi\rangle$ as a trial wave function depending on a set of parameters $C = (C_1, C_2, \ldots)$ the optimal parameters may be found by minimizing the energy functional $E[\Psi] = E(C)$ in Eq. (2.22). A particularly simple and useful form for the trial wave function is one which depends linearly on the parameters, i.e.,

$$|\Psi\rangle = \sum_{i=1}^{m} C_i |i\rangle,$$

where $\{|i\rangle\}$ is an $m$-dimensional set of $N$-particle states. Finding the stationary points of $E(C)$ is in this case equivalent to solving the matrix equation $HC = E(C)SC$, where the elements of $H$ and the overlap matrix $S$ are given by $H_{ij} = \langle i|\hat{H}|j\rangle$ and $S_{ij} = \langle i|j\rangle$, respectively. If the set $\{|i\rangle\}$ is orthonormal the problem reduces to a standard $m$-dimensional eigenvalue problem:

$$HC = E(C)C.$$  

(2.24)

The Hermiticity of $H$ ensures that Eq. (2.24) has exactly $m$ orthonormal solutions and associated real eigenvalues. These represent the approximate wave functions $|\Psi_K\rangle = \sum_{i=1}^{m} C_{iK} |i\rangle$ and approximate energies $E_K$.

In summary, the variational principle in conjunction with a linear expansion of the wave function leads, in an orthonormal basis, to an eigenvalue problem. In an $m$-dimensional trial space the problem yields $m$ solutions, which are associated with the ground and $(m-1)$ excited states of the system. In the limit of a complete expansion (a basis of infinite size) the exact solutions are recovered. Thus, by systematically extending the basis of $N$-particle states it is possible to improve the approximate solutions to the Schrödinger equations in a controlled manner.

2.3 The Hartree-Fock approximation

A natural starting point in constructing a hierarchy of approximations is to constrain the variational space so that it consists of only one many-body state $|\Psi_0\rangle$, or in other words, to find the optimal way to describe a given system in terms of one ON-state as defined in Eq. (2.8). ON-states are constructed using products of single-particle states (see Eqs. (2.16)–(2.17)), and therefore the single-particle states themselves need to be optimized. This scheme, which is not only a reference point for more advanced approximation schemes, but a useful approximation in its own right, is called the Hartree-Fock (HF) approximation.

Employing the variational principle to the problem leads to the requirement that the expectation value $E$ of the Hamiltonian be stationary with respect to variations in the single-particle states, formally expressed as $\phi_i \rightarrow \phi_i + \delta \phi_i$. Since $E$ is a functional of the single-particle states the variational principle may be expressed as

$$\delta E[\psi_i] = \delta \langle \Psi_0 | \hat{H} | \Psi_0 \rangle = 0.$$  

(2.25)

Equation (2.25) supplemented by the requirement that $\langle \Psi_0 | \Psi_0 \rangle = 1$, i.e., the trial state is normalized to unity, leads to a set of effective single-particle equations for the $|\phi_i\rangle$'s, the Hartree-Fock equations. For both bosons and fermions the equations are of the following general form:

$$\hat{f}|\phi_i\rangle = \sum_{j=1}^{n} \epsilon_{ij} |\phi_j\rangle, \quad i = 1, \ldots, n,$$

(2.26)

In some cases a fixed linear combination of ON-states may be sought.
where $n$ is the number of single-particle states and $\hat{f}$ is the Fock operator. It can be shown that the expectation value $E$ is invariant with respect to unitary transformations of the single-particle states. It is thus possible to find a set of states for which the matrix $\epsilon_{ij}$ is diagonal. In this canonical basis $\{|\tilde{\phi}_i\rangle\}$ the HF equations take the form

$$\hat{f}|\tilde{\phi}_i\rangle = \epsilon_i|\tilde{\phi}_i\rangle.$$  \hfill (2.27)

At first glance Eq. (2.27) is of the form of an eigenvalue equation. The Fock operator $\hat{f}$, however, depends on the states $\{|\phi_i\rangle\}$, i.e., the solutions themselves:

$$\hat{f} = \hat{f}[\{|\phi_i\rangle\}].$$  \hfill (2.28)

The canonical HF equation (2.27) is thus a pseudo-eigenvalue problem, which must be solved iteratively. The explicit form of the Fock operator depends on whether the system under consideration is bosonic or fermionic, but in both cases it is of the general form

$$\hat{f} = \sum_{pq} f_{pq} c_p^\dagger c_q = \hat{h} + \hat{g},$$  \hfill (2.29)

where $\hat{h}$ comprises the one-body terms of the full Hamiltonian and $\hat{g}$ is an effective one-body operator that accounts for the interparticle interaction by considering the interaction of a particle with the average field created by the other particles. The Hartree-Fock approximation is thus a mean field theory. This means that the HF model does not take the correlated motion of the particles into account. Indeed, the correlation energy $E_{\text{corr}}$ of a system is commonly defined as the part of the energy that is not accounted for within the HF approximation:

$$E_{\text{corr}} = E_{\text{exact}} - E_{\text{HF}}.$$  \hfill (2.30)

Note that the HF energy is not equal to the sum of the eigenvalues of the Fock operator $\hat{f}$, i.e., $E_{\text{HF}} \neq \sum_{i=1}^n \epsilon_i$, but is the expectation value of the full Hamiltonian in the state $|\Psi_{\text{HF}}\rangle$:

$$E_{\text{HF}} = \langle \Psi_{\text{HF}} | \hat{H} | \Psi_{\text{HF}} \rangle.$$  \hfill (2.31)

Rather than employing some numerical scheme for solving the HF equations directly, the problem is often approached by expanding the unknown single-particle states, or orbitals as they are referred to in quantum chemistry, in a basis of known functions, leading to a matrix representation of the Fock operator. For bases larger than the required minimum the solution yields a set of unoccupied orbitals in addition to the ones which are used to construct the HF wave function. These virtual orbitals are not superfluous when going beyond the HF model, but play a crucial role in constructing more accurate descriptions.

While the HF model in many cases may capture the essential physics of the system, there are numerous cases in which its predictions are qualitatively incorrect. More than one ON-state is thus required in order to describe the correlated motion of the particles. This implies formulating a way to incrementally expand the variational space.

### 2.4 Configuration interaction

Within the configuration interaction (CI) model the $N$-body wave function is expanded as a linear combination of $N$-body states $|i\rangle$ and the linear variational principle is then employed to find the optimal expansion coefficients $C$. The CI wave

**In the case of fermions, the HF model does account for the so-called exchange correlation arising from the antisymmetry of the total wave function.
The function \(|\Psi_{\text{CI}}\rangle \equiv |C\rangle\) is thus written as
\[|C\rangle = \sum_i C_i |i\rangle,\] (2.32)
where the states \(|i\rangle\) are ON-states. It then follows from the variational principle that the coefficients that minimize the energy \(E = \langle C|\hat{H}|C\rangle\) may be determined by solving the matrix eigenvalue problem
\[\mathbf{H}C = EC,\] (2.33)
where it is assumed that \(\langle C|C\rangle = 1\). The matrix elements of the Hamiltonian \(\mathbf{H}\) are simply
\[H_{ij} = \langle i|\hat{H}|j\rangle.\] (2.34)

If the basis \(|i\rangle\) is complete the exact solution to the Schrödinger equation is recovered. Since a complete basis is never available in practice it is imperative to formulate a strategy for generating appropriate bases for the CI expansion which lead to good descriptions of the system while rendering the problems computationally tractable.

The most widely used scheme for generating a hierarchy of variational CI-spaces relies on first obtaining a reference state \(|\Psi_0\rangle\), which is a single configuration assumed to provide a reasonable description of the system by itself. The Hartree-Fock state provides the optimum single-configuration description of a system, as discussed in the previous section, and is thus the obvious choice for \(|\Psi_0\rangle\). New configurations may be generated from the reference state by removing, or “exciting”, particles from occupied orbitals to unoccupied ones. Thus, the exact wave function \(|\Psi_{\text{FCI}}\rangle\) expanded in the full many-body basis generated by a set of single-particle states may be written as
\[|\Psi_{\text{FCI}}\rangle = \left(C_{\text{HF}} + \sum_{ab} C_{ab}^{\text{HF}} c_a^\dagger c_b + \sum_{a>b,i>j} C_{ij}^{\text{HF}} c_a^\dagger c_b^\dagger c_i c_j + \ldots\right)|\Psi_{\text{HF}}\rangle,\] (2.35)
where the indices \(\{i,j,\ldots\}\) refer to states which are occupied and \(\{a,b,\ldots\}\) to virtual orbitals, i.e., states unoccupied in \(|\Psi_{\text{HF}}\rangle\).

The number of different \(N\)-body states that can be generated by a given set of \(K\) single-particle states, i.e., the number of terms in the expansion (2.35), is \(\binom{K}{N}\) for fermions and \(\binom{K+N-1}{N}\) for bosons, a number often too large for practical calculations. A scheme for reducing the size of the variational space is thus called for. A hierarchy of successively larger spaces may be produced by retaining only terms up to a given level of excitation in the expansion (2.35). The model which includes all single and double excitations is called “CI singles and doubles” (CISD), including triple excitations leads to “CI singles, doubles and triples” (CISDT) etc., with the process terminating in the complete expansion, known as “full CI” (FCI)\(^{\dagger}\).

### 2.5 Coupled cluster

The coupled cluster model discussed in this section considers fermionic systems only, since no bosonic CC models were employed in this work. For a treatment of bosonic coupled cluster theory see, e.g., Cederbaum et al.\(^6\) In the CI model the ansatz wave function may be formally written as\(^7\)
\[|\Psi_{\text{CI}}\rangle = (1 + \hat{C}_1 + \hat{C}_2 + \ldots \hat{C}_m)|\Psi_0\rangle,\] (2.36)
\(^{\dagger}\)In the case of \(N_\alpha\) electrons with spin \(\alpha\) and \(N_\beta\) electrons with spin \(\beta\) and \(k\) spatial orbitals the number of states is \(\binom{k}{N_\alpha}\binom{k}{N_\beta}\). For a system with total spin \(S\) the number of states is \(\frac{1}{2(N/2-S+1)}(\binom{n}{N_\alpha})^\beta\binom{n}{N_\beta}\).
\(^{7}\)Within the physics community FCI is often referred to as “exact diagonalization”.

---


where the operators $\hat{C}_i$ are $i$th order excitation operators and $m$ is the maximum excitation level. In the coupled cluster (CC) model the linear expansion of excitation operators is replaced with an exponential form:

$$|\Psi_{CC}\rangle = e^{\hat{T}}|\Psi_0\rangle,$$

where the \textit{cluster operator} $\hat{T}$ is defined as

$$\hat{T} = \sum_{n=1}^{m} \hat{T}_n,$$

with $\hat{T}_n$ being the $n$th order cluster operator:

$$\hat{T}_n = \left(\frac{1}{n!}\right)^2 \sum_{ij...ab...} t^{\text{ab}...\text{ij}...}_{i_j...a_b...a_i}.$$ (2.39)

The factor $t^{\text{ab}...\text{ij}...}_{i_j...a_b...a_i}$ in Eq. (2.39) is the \textit{cluster amplitude} associated with a configuration in which the particles in the occupied states $\{i,j,\ldots\}$ are excited to the virtual states $\{a,b,\ldots\}$. If the sum in Eq. (2.38) is extended to $m = N$ the full many-body space spanned by the single-particle basis is recovered, while a hierarchy analogous to that of the CI model described above is achieved by truncating the sum at a lower $m$. Truncating at $m = 2$ gives CC singles and doubles (CCSD), $m = 3$ includes triple excitations (CCSDT) and so on.

If the expansion (2.38) is truncated, for example, at the double excitation level the exponential operator becomes

$$e^{\hat{T}} = \left(1 + \hat{T}_1 + \frac{1}{2!} \hat{T}_1^2 + \frac{1}{3!} \hat{T}_1^3 + \hat{T}_2 + \frac{1}{2!} \hat{T}_2^2 + \frac{1}{4!} \hat{T}_1^4 + \hat{T}_2 \hat{T}_1 + \frac{1}{2!} \hat{T}_2 \hat{T}_1^2 + \ldots\right).$$ (2.40)

The CC wave function thus includes configurations of an arbitrary excitation level even if the cluster operator is truncated, due to the occurrence of operator products, as seen in Eq. (2.40). The amplitudes for such \textit{disconnected} terms are not independent, however, but are products of amplitudes for connected configurations.

Although it is possible, in principle, to obtain the CC amplitudes and energy variationally, \textit{i.e.}, by finding the stationary points of $E = \langle \Psi_0 | (e^{\hat{T}})^\dagger \hat{H} e^{\hat{T}} | \Psi_0 \rangle$, the resulting equations tend to be tedious to solve and require the truncation of the exponential series according to some criteria which are not well defined. Rather, most practical implementations of coupled cluster theory rely on a different, “projective” technique for finding the CC wave function. Consider, then, the Schrödinger equation for the CC state:

$$\hat{H} |\Psi_{CC}\rangle \equiv \hat{H} e^{\hat{T}} |\Psi_0\rangle = E e^{\hat{T}} |\Psi_0\rangle.$$ (2.41)

Multiplying this expression with $e^{-\hat{T}}$ and left-projecting with the reference state we obtain

$$\langle \Psi_0 | e^{-\hat{T}} \hat{H} e^{\hat{T}} |\Psi_0\rangle = E.$$ (2.42)

By projecting with an excited configuration $|\Psi_{ab...}^{\text{ab}...\text{ij}...\text{a}\text{b}...\text{a}}\rangle$ instead of the reference, an equation for the corresponding amplitude is obtained:

$$\langle \Psi_{ab...}^{\text{ab}...\text{ij}...\text{a}\text{b}...\text{a}} | e^{-\hat{T}} \hat{H} e^{\hat{T}} |\Psi_0\rangle = 0.$$ (2.43)

Eqs. (2.42) and (2.43) constitute the basis for solving the coupled cluster model. Note the occurrence of a non-Hermitian similarity-transformed Hamiltonian $\hat{H} = e^{-\hat{T}} \hat{H} e^{\hat{T}}$ in place of the original $\hat{H}$ in these equations.
To facilitate the derivation of explicit expressions for \( E \) and the amplitudes \( t \) it is useful to expand the similarity-transformed Hamiltonian \( \hat{H} \) as a series of commutators using the Campbell-Baker-Hausdorff formula:

\[
e^{-\hat{T}} \hat{H} e^{\hat{T}} = \hat{H} + [\hat{H}, \hat{T}] + \frac{1}{2!} [[\hat{H}, \hat{T}], \hat{T}] + \ldots
\]  

(2.44)

The fermionic anticommutation relations (2.6)–(2.7) and the fact that the cluster operators commute amongst themselves, along with the two-particle property of the Hamiltonian leads to the exact termination of the series (2.44) at the fifth term. This renders the derivation of expressions for efficient implementation on a computer considerably easier. The energy expression (2.42), for example, becomes

\[
E_{\text{CC}} - E_{\text{HF}} = \sum i a f_{ia} t_i^a + \frac{1}{4} \sum a i b j \langle ij || ab \rangle t_{ij}^{ab} + \frac{1}{2} \sum a i b j \langle ij || ab \rangle t_i^a t_j^b,
\]  

(2.45)

where \( f_{ia} \) is the matrix element of the Fock operator and the standard quantum chemical notation \( \langle ab || ij \rangle \) is used as shorthand for the two-body matrix element containing the Coulomb and exchange interaction terms. Note that the energy expression (2.45) holds for arbitrary CC levels, even if it only contains amplitudes corresponding to a maximum level of double excitations.

The simple expression for the CC energy is “compensated” by much more complicated equations for the amplitudes. The cumbersome task of deriving them is commonly accomplished using diagrammatic techniques yielding a set of coupled nonlinear equations for the amplitudes.

The fact that the CC energy (as obtained from the implementation described here) is not variational, i.e., it is not obtained as an expectation value of a Hermitian operator, raises questions about its physical reality. For one, it means that the energy does not represent an upper bound to the exact energy in the event that the cluster operator is truncated. However, if \( \hat{T} \) is not truncated then \( \hat{H} \) has an eigenvalue spectrum which is identical to the true Hamiltonian. In practice the CC energy tends to closely approximate the expectation value result even when \( \hat{T} \) is truncated. 


3 Computational methods

3.1 Configuration addressing

In a program which repeatedly refers to and manipulates configurations, or occupation number strings, it is of importance to implement an efficient way to map a string to an address in the computer memory. In other words, the task is to find a simple and efficient mapping accomplishing

\[ |n_1, n_2, \ldots, n_N \rangle \rightarrow I_{n_1, n_2, \ldots, n_N}, \]  

(3.1)

where \( I_{n_1, n_2, \ldots, n_N} \) is an integer. It is also desirable that the mapping (3.1) produces an ordering of the strings according to some logical scheme. The method presented here is based on representing the strings in graphical form as paths in a diagram and then introducing a scheme for ordering these paths.

Consider, then, the case with \( N \) particles (of arbitrary symmetry) and a single-particle space of \( n \) orbitals. Each string may be represented by a vector of \( n \) ordered pairs of integers \((k, m)\), where \( k \) refers to the orbitals and \( m \) is the number of particles in the orbitals up to \( k \). For example, for \((n, N) = (3, 2)\) the string \(|1, 0, 1\rangle\) corresponds to the set \{(1, 1), (2, 1)\}. These pairs of integers may then be taken to represent coordinates, or vertices, in a two-dimensional diagram. The lines, or arcs, connecting them then form a path.

To illustrate the principle we consider a system of 3 particles in 5 orbitals. The string \(|1, 1, 0, 1\rangle\) has the graphical representation shown in Fig. 3.1 (a). A vertex connected to the previous one with a diagonal arc thus represents an occupied orbital, whereas a vertical arc corresponds to an unoccupied one. For bosons horizontal arcs are also allowed, corresponding to multiply occupied orbitals, as illustrated in Fig. 3.1 (c) and (d) for the strings \(|3, 0, 0, 0\rangle\) and \(|0, 0, 2, 1, 0\rangle\). Each string is thus represented by a unique path in the diagram. The last vertex \((n, N)\) may thus be assigned a number, or weight \( W_{n,N} = M \), where \( M \) is the number of different allowed paths starting at the origin and ending at \((n, N)\), i.e., the total number of configurations. This number must be equal to the sum of the corresponding weights for the vertices to which it is connected. In other words, for fermions

\[ W_{k,m}^F = W_{k-1,m}^F + W_{k-1,m-1}^F. \]  

(3.2)

For bosons, each vertex may be connected to up to three previous vertices, but since paths with ninety-degree angles are not allowed these must be subtracted from the total:

\[ W_{k,m}^B = W_{k-1,m}^B + W_{k-1,m-1}^B + \left( W_{k,m-1}^B - W_{k-1,m-1}^B \right) = W_{k-1,m}^B + W_{k,m-1}^B. \]  

(3.3)

Using the relations (3.2) and (3.3) weights may be assigned to all vertices in the diagram, which is illustrated for the bosonic case in Fig. 3.2.

In order to assign a unique number, or weight, to each path the arcs must also be assigned weights. We wish to order the strings in such a way that the ones with
3.1 Configuration addressing

All the allowed vertices are included in the graphs.

Figure 3.1: Paths corresponding to the fermionic states $|1,1,0,0,0\rangle$ (a), $|1,1,0,1,0\rangle$ (b) and the bosonic states $|3,0,0,0,0\rangle$ (c) and $|0,0,2,1,0\rangle$ (d). All the allowed vertices are included in the graphs.

highest occupation indices have largest weight. The vertical arcs do not contribute, so their weight is set to zero. Consider, then, a subgraph of size $(k,m)$. The point $(k,m)$, or the tail, has the weight $W_{k,m}$ as noted above. Adding a diagonal arc expands the graph such that the tail is at $(k+1,m+1)$, and this extended graph will have $W_{k+1,m+1}$ paths with weights between 1 and $W_{k+1,m+1}$. Of these paths the one with the highest weight, i.e. $W_{k+1,m+1}$, should correspond to one having the orbital

Figure 3.2: Vertex and arc weights for a system of 3 bosons in 5 orbitals. The vertical arcs all have a weight of zero.
$k+1$ occupied and connected to the path with the highest weight $W_{k,m}$ in the smaller subgraph. In other words,

$$W_{k+1,m+1} = W_{k,m} + Y^1_{k+1,m+1} \iff Y^1_{k+1,m+1} = W_{k+1,m+1} - W_{k,m} \equiv W_{k,m+1}, \quad (3.4)$$

where we have introduced the arc weight $Y^m_{k,m}$.

For the horizontal arcs, which may occur in bosonic diagrams, an analogous argument gives the following relation for the arc weight:

$$W_{k,m+1} = W_{k,m} + Y^1_{k,m+1} \iff Y^1_{k,m+1} = W_{k,m+1} - W_{k,m} \equiv W_{k-1,m+1}. \quad (3.5)$$

Finally, the path weight is obtained by summing the weights of the individual arcs:

$$I_{m_1,m_2,...,m_N} = 1 + \sum_{k=1}^{n} Y^{m_k-m_{k-1}}_{k,m_k}, \quad (3.6)$$

with $m_k = \sum_{i=1}^{k} n_i$, and $Y^0_{k,m} = 0$, as stated above. The vertex and path weights for a bosonic system with $N = 3$ and $n = 5$ are shown in Figure 3.2. From the Figure it may be verified that the configuration $|3,0,0,0,0\rangle$ obtains the address $I_{2,3,0,0,0,0} = 1$ and the address of the configuration $|0,0,0,0,3\rangle$ is $I_{1,0,0,0,1,2.3} = 35$, i.e., the highest numerical value in this case. This illustrates the reverse lexical ordering of the strings which this scheme produces. In this ordering string $A$ comes before string $B$ if in the last occupation in which they differ $A$ has the lower occupation number.

### 3.2 Direct CI

Constructing the matrix elements $H_{ij}$ of the CI-matrix, storing them in the computer memory (or, worse, on disk) and performing a full diagonalization of the matrix is a task which stretches the capabilities of even the most powerful computers once the number of configurations becomes large. One way to avoid these computational bottlenecks is to employ iterative techniques for obtaining the lowest or few lowest eigenvalues and eigenstates of the system. The scheme described here, which is implemented in both the QD and boson code is an inverse-iteration-based generalization of the Davidson algorithm developed by Olsen et al.

Let, then, the current approximation for the solution of the CI problem $HC = EC$ be $C^{(0)}$, the corresponding energy being $E^{(0)} = C^{(0)\dagger}HC^{(0)}$. Furthermore, let the Hamiltonian be divided into a zeroth-order term and a correction term

$$H = H^{(0)} + H^{(1)}. \quad (3.7)$$

The CI equation for the next iterative step is then written as

$$(H^{(0)} + H^{(1)})(C^{(0)} + C^{(1)}) = (E^{(0)} + E^{(1)})(C^{(0)} + C^{(1)}), \quad (3.8)$$

where $C^{(1)}$ and $E^{(1)}$ are the sought-after corrections to the current approximation. Solving for $C^{(1)}$ and neglecting the terms which are quadratic in the correction terms one obtains

$$C^{(1)} = -(H^{(0)} - E^{(0)})^{-1}[(H - E^{(0)})C^{(0)} - E^{(1)}C^{(0)}]. \quad (3.9)$$

Requiring that the correction vector is orthogonal to the current approximation, i.e., $C^{(0)\dagger}C^{(1)} = 0$, the energy correction becomes

$$E^{(1)} = \frac{C^{(0)\dagger}(H^{(0)} - E^{(0)})^{-1}(H - E^{(0)})C^{(0)}}{C^{(0)\dagger}(H^{(0)} - E^{(0)})^{-1}C^{(0)}}. \quad (3.10)$$
In the iterative scheme defined by Eqs. (3.9) – (3.10), the most demanding computational step is carrying out the matrix-vector multiplications of the form

\[ \sigma = HC. \]  

(3.11)

The \( \sigma \)- and (a few) \( C \)-vectors are also the largest sets of data that need to be stored at any one time. For practical implementations it is important that the linear transformation \( (3.11) \) is performed efficiently in terms of the one-and two-body matrix elements. In the bosonic case the matrix-vector multiplication operation can be written as

\[ \sigma_I = \sum_J \left( \sum_{pq} \langle I|a_p^\dagger a_q|J \rangle + \frac{1}{2} \sum_{pqrs} g_{pqrs} \langle I|a_p^\dagger a_r^\dagger a_s a_q|J \rangle \right) C_J, \]  

(3.12)

where \( \sigma_I \) is the component of the product vector corresponding to the \( I \)th configuration. In the boson program a significant reduction in computational effort is achieved by summing, in Eq. (3.12), over the indices corresponding to the annihilation operators first, identifying cases in which the vacuum is annihilated, and then summing over the creation operator indices only when a nonvanishing contribution is at hand. In practice the vast majority of the contributions vanish, and these “Slater-Condor” rules effectively reduce the summations to a single summation for the one-body part and a double summation for the two-body part.

The correction vector obtained from Eqs. (3.9) and (3.10) can in principle be added to \( C^{(0)} \) to obtain an improved eigenvector. Usually, however, it is better to regard the vectors \( C^{(0)} \) and \( C^{(1)} \) as spanning a two-dimensional subspace and to then find the improved eigenvector by diagonalizing the Hamiltonian projected onto this subspace. The elements of the projected Hamiltonian are found from the \( \sigma \)-vectors which have been computed in generating the correction vector.

As seen from Eq. (3.9) the algorithm involves the use of a preconditioner in terms of the matrix \( H_0 \). In our implementation \( H_0 \) consists of a block of the Hamiltonian formed within a determinant space defined by the \( p \) lowest diagonal elements of \( H \). Outside this block we use the diagonal elements of \( H \) to define \( H_0 \).
Part II

The Physical Systems
4 Quantum Dots

A quantum dot (QD) is a system in which charge carriers are artificially confined in all three spatial dimensions on a scale comparable to their de Broglie wavelength, leading to quantized motion and corresponding discrete energy levels. Here, “artificial” is used to distinguish quantum dots from naturally occurring similar systems, such as atoms or impurities in bulk material.

The most obvious difference between QDs and their natural counterparts is size: atomic sizes are measured in Ångstroms, whereas the length scale of confinement potentials in QDs are typically tens of nanometers. The geometrical shape of the confinement potential in QDs also differs from the spherically symmetric Coulomb potential, which binds electrons to the atomic nucleus. A QD is often quasi two-dimensional, with a strong confinement in the z-direction and a lateral potential closely approximating a harmonic one. The $1/r$-singularity of the central Coulomb potential is therefore absent in QDs.

The properties of a specific quantum dot depend on several factors, such as the materials in which it is embedded, the details of the fabrication process and so on. This means that the structure and therefore the electronic properties of QD systems can be adjusted, or tuned. Quantum dots have therefore already been widely used in devices, such as light emitting diodes, lasers, and field effect transistors. The possibility to tune QDs into effective two-level systems (qubits) have made them a subject of interest in the realm of quantum information processing.

4.1 General Properties

4.1.1 Materials and fabrication

A semiconductor quantum dot can be viewed as a man-made, artificial environment in which single charges or elementary excitations may be trapped. How, then, is this achieved? Historically QDs can be seen as the culmination of a process of miniaturization and dimensionality reduction, perhaps logically starting from the advent of epitaxial growth techniques and the two-dimensional electron gas (2DEG). The quantum well (QW) confinement of the 2DEG is most commonly achieved by sandwiching a thin layer of semiconductor between layers of another semiconductor material with a larger bandgap, effectively confining the carriers to motion in two dimensions, provided the layer is thin compared to the wavelength (or mean free path) of the carriers. The reduced dimensionality compared to the bulk crystal is reflected in the density of states, which becomes step-like, as illustrated in Fig. 4.1. Further restriction of the carrier motion to one effective dimension, yielding a quantum wire (QW), may be achieved by, e.g. advanced etching techniques. Several quite different techniques for creating the lateral confinement potential have been implemented. The first QDs were realized by etching techniques, creating pillars of lithographically produced QW material. Another early confinement
method relies upon patterning the surface of a QW structure with miniature electrodes, with which a tunable, purely electrostatic lateral potential can be produced.\cite{22}

4.1.2 Strain-induced quantum dots

A set of techniques that avoid the precision work of depositing electrodes or high resolution etching are the ones based upon the phenomenon of self-organized growth, yielding self-assembled quantum dots (SAQD). This involves depositing monolayers of material on a substrate with a differing lattice constant. At first the deposited material crystallizes in the form of strained layers with a lattice constant equal to that of the substrate, but when a critical thickness is exceeded the structure spontaneously breaks down into islets of regular shape and similar sizes. This phase transition is called the Stranski-Krastanow (SK) transition\cite{22}

The QDs modeled in this thesis are fabricated by SK growth of InP on a \( \text{In}_{x}\text{Ga}_{1-x}\text{As/GaAs QW}\)\cite{22}. After a critical thickness of two mono-layers is exceeded the InP assembles into dislocation-free islands. Atomic force microscope images of the prepared samples show a typical island diameter of 60-80 nm and a height of 25 nm. Figure 4.2 features a schematic picture of the system.

4.1.3 Carriers in strain-induced quantum dots

In a bulk semiconductor electronic states form energy bands, which are continuous (but not unique) functions of the wave vector \( \mathbf{k} \). Semiconductors display a band gap, i.e., an energy interval between the edge of the highest occupied (valence) band and the corresponding point in the lowest unoccupied (conduction) band in which no electronic states exist. When an electron is excited from the valence band to the conduction band the unoccupied state it leaves behind may be treated as a quasiparticle of its own, i.e., a positively charged hole.

![Figure 4.1: Schematic illustration of the density of states of a semiconductor system with different degrees of freedom.](image-url)
4.2 The model

Figure 4.2: Schematic picture of the strain-induced GaAs/In\textsubscript{x}Ga\textsubscript{1-x}As/GaAs QD system. The parameters used in most of our calculations are \( x = 0.1, D = 5 \) nm, \( 2R = 80 \) nm, and \( W = 7 \) nm. Reprinted with permission from Braskén et al.\textsuperscript{87}

The electron and the hole interact via the Coulomb interaction, so that each pair contributes the interaction energy \( W_{eh} \) to the system:

\[
W_{eh}(\mathbf{r}_e, \mathbf{r}_h) = \frac{1}{4\pi\varepsilon_0\varepsilon_r} \frac{-e^2}{|\mathbf{r}_e - \mathbf{r}_h|},
\]

where the indices \((e, h)\) refer to the electron and the hole, respectively, and \( \varepsilon_r \) is the relative dielectric constant of the material. Due to the opposite charge of the particles the interaction lowers the energy of the system and an exciton is formed. In the bulk material an exciton can dissociate into a pair of free carriers in the conduction and valence bands, and in principle the exciton binding energy is measurable. In a quantum dot an uncorrelated ground state does not exist, and the definition of the binding energy as the difference between the eigenenergies of a Hamiltonian with and without the Coulomb interaction is mainly a theoretical concept\textsuperscript{24}.

For two excitons bound in the same structure, forming a biexciton \((2X)\), the binding energy, defined as \( E_{\text{bind}} = 2E_X - E_{2X} \), can be experimentally observed, e.g., in recombination spectra.

The Coulomb interaction leads to a correlation of the motion of the carriers, which leads to qualitative effects in the form of changes in the energy spectrum and – importantly – altered oscillator strengths of transitions. The importance of the Coulomb interaction in a given system depends on the interplay between the size of the exciton and the quantum dot radius.

4.2 The model

4.2.1 The \(k \cdot p\)-model

The starting point in constructing our model of strain-induced QDs is the \(k \cdot p\) model, which is a perturbative treatment for calculating the dispersion relations. Letting a Hamiltonian of the form \( H = p^2/2m + V(\mathbf{r}) \) act on a Bloch state \( u_{nk}(\mathbf{r}) = e^{i\mathbf{k} \cdot \mathbf{r}} u_{nk}(\mathbf{r}) \) one obtains the following equation for the cell-periodic part \( u_{nk} \):

\[
\begin{bmatrix}
\frac{p^2}{2m_0} + V(\mathbf{r}) + \frac{\hbar}{m_0} \mathbf{k} \cdot \mathbf{p} + \frac{\hbar^2 k^2}{2m_0}
\end{bmatrix} u_{nk}(\mathbf{r}) = E_n(\mathbf{k}) u_{nk}(\mathbf{r}).
\]

For \( \mathbf{k} = 0 \) the operator on the left hand side becomes \( H_0 = p^2/2m_0 + V(\mathbf{r}) \) (with solutions \( u_{mn} \)) and Eq. (4.2) may be written as

\[
[H_0 + H_k] u_{nk}(\mathbf{r}) = E_n(\mathbf{k}) u_{nk}(\mathbf{r}),
\]

where
where $H_k = \frac{\hbar}{m_0} k \cdot p + \frac{\hbar^2 k^2}{2m_0}$. The unperturbed states are thus the ones at the band edge ($k = 0$) and the $k$-dependent part of the Hamiltonian is treated as a perturbation. For a single band the dispersion is, to second order:

$$E_n(k) - E_n(0) = \sum_{\alpha, \beta = \{x, y, z\}} D^{\alpha\beta} k_\alpha k_\beta,$$  \hspace{1cm} (4.4)

where

$$D^{\alpha\beta} = \frac{\hbar^2}{2m_0} \delta_{\alpha\beta} + \frac{\hbar^2}{2m_0} \sum_{n \neq n'} p_{n\alpha}^0 p_{n'\alpha}^0 + p_{n\beta}^0 p_{n'\beta}^0 \frac{E_n(0) - E_{n'}(0)}{E_A - H_{00}},$$  \hspace{1cm} (4.5)

In Eq. (4.5) $(1/m^*)_{\alpha\beta}$ is the inverse effective mass tensor.

### 4.2.2 The Luttinger-Kohn Hamiltonian

At the $\Gamma$-point, the band structure of III-V-compounds (e.g. GaAs) exhibits a doubly degenerate conduction band and 3 valence bands, which are all doubly degenerate. Other bands lie energetically far enough above (below) these bands to motivate a treatment which only considers these bands explicitly, taking other bands into account perturbatively using L"owdin’s renormalization technique. In this approach, bands are divided into two classes: class A, which are the bands of interest, and class B, which includes all other bands. The ansatz wavefunction is then written as:

$$\Psi(r) = \sum_{n} a_n \phi_n^{(0)} + \sum_{m} b_m \psi_m^{(0)}.$$  \hspace{1cm} (4.6)

In the case of degenerate (or nearly degenerate) bands in the presence of a perturbation $H'$ L"owdin’s technique yields the matrix $U_{mn}^A$, which upon diagonalization gives the coefficients $a_n$ and the corresponding eigenenergies of the perturbed system. The form of this matrix is:

$$U_{mn}^A = H_{mn} + \sum_{\alpha} H'_{\alpha n} H'^{\alpha}_{mn} \frac{E_A - H_{00}}{E_A - H_{00}},$$  \hspace{1cm} (4.7)

where $E_A$ is the average energy of the unperturbed states in class A.

For compounds such as GaAs the spin-orbit coupling is significant enough to induce qualitative effects on the dispersion. The Hamiltonian for the cell-periodic part of the wave function including spin-orbit coupling is:

$$H = H_0 + \frac{\hbar^2 k^2}{2m_0} + \frac{\hbar}{4m_0 c^2} \nabla V \times p \cdot \sigma + H',$$  \hspace{1cm} (4.8)

where

$$H_0 = \frac{p^2}{2m_0} + V(r)$$  \hspace{1cm} (4.9)

$$H' = \frac{\hbar}{m_0} k \cdot \Pi, \quad \Pi = p + \frac{\hbar}{4m_0 c^2} \sigma \times \nabla V.$$  \hspace{1cm} (4.10)

The basis functions are conventionally chosen as the solutions to Kane’s model, e.g., near-band edge ($k \approx 0$) states for the system including spin-orbit interaction. These states are eigenfunctions of the total angular momentum $J$, and are labeled as $|J, J_z\rangle$, with $J = \{\frac{1}{2}, \frac{3}{2}\}$ and $J_z = \{\pm\frac{1}{2}, \pm\frac{3}{2}\}$ for these $4p$-like states. Below, these states are referred to as $u_0 n$, $n = \{1, 2, \ldots 6\}$. 


Applying Löwdin’s technique with the six valence bands belonging to class A the matrix elements in Eq. (4.7) become:

\[ H_{mn} = \langle u_m | H | u_n \rangle = \left[ E_m(0) + \frac{\hbar^2 k^2}{2m_0} \right] \delta_{mn} \] (4.11)

\[ H'_{mA} = \langle u_m | | u_\alpha \rangle \approx \sum_{i=x,y,z} \frac{\hbar k_i}{m_0} p_i \alpha, \ m \in A, \ \alpha \notin A, \] (4.12)

In Eq. (4.12) \( \Pi \) is approximated by \( p \), which is justified on the grounds that the crystal momentum \( \hbar k \) is much smaller that the particle momentum in the atomic orbit \( p \), i.e., the second to last term in Eq. (4.8) is much larger than the last term in the definition of \( \Pi \) in Eq. (4.10).

Letting \( U^A_{mn} \equiv D_{mn} \) one can write the Hamiltonian in the form

\[ D_{mn} = E_m(0) \delta_{mn} + \sum_{\alpha,\beta} D^\alpha_{mn} k_\alpha k_\beta, \] (4.13)

where \( D^\alpha_{mn} \) is defined as

\[ D^\alpha_{mn} = \frac{\hbar^2}{2m_0} \left[ \delta_{mn} \delta_{\alpha\beta} + \sum_\gamma \frac{p_{m\gamma} p_{\gamma n} + p_{\gamma m} p_{\gamma n}}{m_0(E_0 - E_\gamma)} \right], \] (4.14)

which is similar to the single-band case, Eq. (4.5), only generalized to include degenerate bands. Expressing the Hamiltonian (4.13) explicitly in the basis \( \{ u_j \}_{a=1}^6 \) a 6×6 matrix is obtained, which is referred to as the Luttinger-Kohn (LK) Hamiltonian \( H^{LK} \).

Summarizing, diagonalizing the LK Hamiltonian is equivalent to solving the equations

\[ \left[ \frac{p^2}{2m_0} + V(r) + \frac{\hbar}{4m_0^2 c^2} \nabla V \times \sigma + \frac{\hbar k}{m_0} \cdot p \right] \psi_{nk}(r) = E_{nk} \psi_{nk}(r) \] (4.15)

for the valence hole subbands, with

\[ \psi_{nk}(r) = e^{i\mathbf{k} \cdot \mathbf{r}} u_{nk}(r), \] and \[ u_{nk}(r) = \sum_{m=1}^6 a_m(k) u_{m0}(r). \] (4.16) (4.17)

### 4.2.3 The effective mass approximation

So far only the periodic external potential produced by the atoms in the crystal has been considered. When a non-periodic perturbation \( U(r) \) is present, such as a strain field or a heterojunction, the effective mass approximation (EMA), also called the envelope function approximation, may be employed under the assumption that the perturbation varies slowly on the interatomic scale. The main results of EMA as they apply to the problem at hand are presented below.

Assume that, for a single band, labeled by \( n \) near \( k_0 \) (assumed to be 0) the dispersion is of the form (see Eqs. (4.4) and (4.5)):

\[ E_n(k) = E_n(0) + \sum_{\alpha,\beta} \left( \frac{1}{m^*} \right) k_\alpha k_\beta \] (4.18)

for a Hamiltonian \( H_0 \) with a periodic potential \( V(r) \). In the presence of a non-periodic perturbation \( H = H_0 + U(r) \) the wave function within EMA is approximated by

\[ \psi(r) = F(r) u_{n0}(r), \] (4.19)
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where \( F(\mathbf{r}) \) is the *envelope function*. Within this approximation the envelope satisfies the following equation:

\[
\left[ \sum_{\alpha,\beta} \frac{\hbar^2}{2} \left( \frac{1}{m^*} \right)_{\alpha\beta} \left( -i \frac{\partial}{\partial x_\alpha} \right) \left( -i \frac{\partial}{\partial x_\beta} \right) + U(\mathbf{r}) \right] F(\mathbf{r}) = \left[ E - E_n(0) \right] F(\mathbf{r}). \tag{4.20}
\]

The periodic potential \( V(\mathbf{r}) \) thus determines the effective mass \((1/m^*)_{\alpha\beta}\) and the effective mass equation (4.20) only contains the perturbation \( U(\mathbf{r}) \).

In the case of degenerate bands the effective mass equation for the envelope is given, in analogy to the single band case above, by replacing \( k_\alpha \) by \(-i\partial/\partial x_\alpha\) in the LK dispersion relation. A set of coupled differential equations for the \( m \) envelope functions then needs to be solved.

### 4.2.4 Strain and the Pikus-Bir Hamiltonian

For a crystal under uniform strain the deformed unit vectors \( \{\mathbf{a}_i'\} \) can be expressed using the undeformed ones \( \{\mathbf{a}_i\} \) as

\[
\mathbf{a}_i' = \sum_j (\delta_{ij} + \epsilon_{ij}) \mathbf{a}_j, \tag{4.21}
\]

where \( \epsilon_{\alpha\beta} \) are components of the *strain tensor* \( \epsilon \). The effect of the strain on the system may be investigated by expressing the Hamiltonian in the deformed coordinate system:

\[
eq : H_{\text{strained coord}} \left[ \frac{\hbar^2}{2m_0} + V(\mathbf{r}') \right] \psi_n^{k'}(\mathbf{r}') = E_n(k') \psi_n^{k'}(\mathbf{r}'). \tag{4.22}
\]

By expressing the Hamiltonian above in the original unstrained coordinates with help of the relation (4.21) and defining \( \psi_n^{k'}[(1 + \epsilon) \cdot \mathbf{r}] = e^{i\mathbf{k}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r}) \) one obtains the equation for the strained Bloch periodic part \( u_{n\mathbf{k}}(\mathbf{r}) \):

\[
[H_0 + H'] u_{n\mathbf{k}}(\mathbf{r}) = \left( E - \frac{\hbar^2 k^2}{2m_0} \right) u_{n\mathbf{k}}(\mathbf{r})
\]

\[
H' = H_k + H_\epsilon + H_{\epsilon k}
\]

\[
H_k = \frac{\hbar}{m_0} \mathbf{k} \cdot \mathbf{p}
\]

\[
H_\epsilon = \sum_{\alpha,\beta} \hat{D}_{\alpha\beta} \epsilon_{\alpha\beta}
\]

\[
H_{\epsilon k} = -2 \frac{\hbar}{m_0} \sum_{\alpha,\beta} k_\alpha \epsilon_{\alpha\beta} p_{\beta}.
\]

The matrix \( \hat{D}_{\alpha\beta} \) in \( H_\epsilon \) is defined as:

\[
\hat{D}_{\alpha\beta} = \frac{1}{m_0} \rho_{\alpha\beta} + V_{\alpha\beta}, \quad \text{with}\quad V_{\alpha\beta} = \frac{\partial V}{\partial x_{\alpha\beta}} |_{\epsilon_{\alpha\beta} \rightarrow 0}.
\]

Applying the results above to the degenerate case one obtains the Hamiltonian for a lattice of diamond type including strain as:

\[
H = H^{LK} + H_\epsilon. \tag{4.25}
\]
Table 4.1: Deformation potential constants used in the strain calculations

<table>
<thead>
<tr>
<th>Parameter</th>
<th>InAs</th>
<th>GaAs</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_e$</td>
<td>-5.4 eV</td>
<td>-7.1 eV</td>
</tr>
<tr>
<td>$a_v$</td>
<td>0.66 eV</td>
<td>2.67 eV</td>
</tr>
<tr>
<td>$b_v$</td>
<td>-1.8 eV</td>
<td>-1.70</td>
</tr>
</tbody>
</table>

4.2.5 The single-particle model

In order to arrive at a single-particle model of the strain-induced quantum dot system shown in Fig. 4.2 the theory outlined in the previous subsections is applied. Since the size of the InP island is large compared to the interatomic distance, the effective mass approximation including strain according to the theory of Pikus and Bir is applied. As discussed above, this involves finding the strain tensor $\epsilon$.

In the work of Tulkki et al. the strain tensor for a strain-induced QD with a InP island on a In$_{1-x}$Ga$_x$As/GaAs QW is calculated by minimizing the total strain energy of the system using a finite element method. The strain interaction for the conduction band, which is considered independently, is given by

$$V_{eH} = a_c (\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}),$$

where $a_c$ is called the hydrostatic potential of the conduction band. For the valence band strain terms ought in principle to be added to the full six-dimensional LK Hamiltonian. In this treatment, however, the split-off band is ignored, leading to a four band strain Hamiltonian of the form

$$H_{e} = \left( \begin{array}{cccc} V_{H}^{h} - V_{S}^{h} & S^{*} & -R & 0 \\ S & V_{H}^{h} + V_{S}^{h} & 0 & -R \\ -R^{*} & 0 & V_{H}^{h} + V_{S}^{h} & -S^{*} \\ 0 & -R^{*} & -S & V_{H}^{h} - V_{S}^{h} \end{array} \right).$$

This strain Hamiltonian matrix is represented in the angular momentum basis $\{|\frac{3}{2}, \pm \frac{3}{2}\}, |\frac{1}{2}, \pm \frac{1}{2}\}$ described above and the matrix elements are

$$V_{H}^{h} = a_v (\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}),$$
$$S = \frac{\sqrt{3}}{2} b_v (\epsilon_{zx} - \epsilon_{zy}) - i d_v \epsilon_{xy},$$
$$R = \sqrt{3} b_v (\epsilon_{zz} - \epsilon_{yy}).$$

The deformation potential constants $a_c, a_v$ and $b_v$ are given in Table 4.1. The non-diagonal terms in the Hamiltonian (4.27) are non-zero except far away from the dot axis. However, the diagonal shear term $V_{S}^{h}$ decouples the heavy hole and light hole bands in the QD sufficiently as to consider the correction of the non-diagonal terms small near the band edge. The strain potential for the conduction ($e$) and heavy hole ($h$) bands may thus be expressed as

$$V_{e}^{\text{strain}}(z,r) = V_{H}^{h},$$
$$V_{h}^{\text{strain}}(z,r) = V_{H}^{h} + V_{S}^{h}. (4.29)$$

The total confinement potentials are obtained by adding the band-edge confinement potential $V_{QW}^{e,h}(z)$, which is an expression of the difference in the band-gap between the well and barrier materials, to the strain potentials above. The total confinement potential for the conduction and HH band for a QD with the parameters given in Fig. 4.2 is shown in Fig. 4.3.
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Figure 4.3: Numerical confinement potentials for holes (left) and electrons (right) for the QD shown in Fig. 4.2. Reprinted with permission from Braskén et al. [87].

\[ R^{(e,h)}_{n,m}(r,z)e^{im\phi}. \]

Using this functional form the effective mass equation for the electrons and holes in the presence of the strain potential is

\[
-\frac{\hbar^2}{2} \left( \frac{1}{m_{e,h}^r} \frac{\partial^2}{\partial z^2} + \frac{1}{m_{e,h}^r} \frac{\partial}{\partial r} \frac{r}{\partial r} - \frac{1}{m_{e,h}^z} \frac{m_e^2}{r^2} \right) + V^{e,h}_{\text{QW}}(z) + + V^{e,h}_{\text{strain}}(r,z) \times R^{e,h}_{n,m}(r,z) = E_{n,m} R^{e,h}_{n,m}(r,z),
\]

where the effective mass of the electron is \( m_e^r = m_e^z = m_e \) and for the holes \( m_h^r = 1/(\gamma_1 + \gamma_2) \) and \( m_h^z = 1/(\gamma_1 - 2\gamma_2) \).

4.2.6 The many-body model

In the previous subsections a single-particle model of a strain-induced QD and its foundations have been described, arriving at Eq. (4.30). In effect, two kinds of non-interacting particles are considered: electrons and heavy holes, distinguished by their opposing charges and differing masses. In order to describe the system realistically, however, the strong Coulomb interaction among and between the species must be taken into account. To simplify the notation the formalism of second quantization is employed for describing the many-body system.

The many-body Hamiltonian

The two species of particles considered in the model are represented by the creation (annihilation) operators \( c_{i\sigma}^\dagger \) \( (c_{i\sigma}) \) for electrons and \( h_{i\sigma}^\dagger \) \( (h_{i\sigma}) \) for holes. These operators create (annihilate) an electron/hole in the single-particle state \( i \) with spin \( \sigma \).
They fulfill fermionic anticommutation relations \( \{ a_{i\sigma}^\dagger, a_{j\sigma'}^\dagger \} = \delta_{ii'}\delta_{\sigma\sigma'}, \{ a_{i\sigma}, a_{j\sigma'}^\dagger \} = 0 \) (with \( a = (e, h) \)), and thus a state with \( N \) electron-hole pairs of proper statistics may be formed by acting with the creation operators on the vacuum state \( |\text{vac}\rangle \):

\[
|N\rangle = c_{1e}^\dagger c_{1h}^\dagger \cdots c_{N_e}^\dagger h_{1e}^\dagger h_{1h}^\dagger \cdots h_{N_h}^\dagger |\text{vac}\rangle, \tag{4.31}
\]

where the spin index \( \sigma \) has been suppressed. Within first quantization the state \( |N\rangle \) can, due to the fermionic statistics, be expressed as an antisymmetrized linear combination of states which are direct products of single-particle states. In the language of quantum chemistry the state \( |N\rangle \) is often referred to as a configuration.

The many-body Hamiltonian for the electron-hole system then has the form

\[
\hat{H} = \sum_{ij} E_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_{ij} E_{ij}^h h_{i\sigma} h_{j\sigma} - \sum_{ijkl\sigma\sigma'} (ij|V_{eh}|kl)c_{i\sigma}^\dagger c_{j\sigma'}^\dagger h_{k\sigma}^\dagger h_{l\sigma'} + \frac{1}{2} \sum_{ijkl\sigma\sigma'} (ij|V_{eh}|kl)c_{i\sigma}^\dagger h_{k\sigma}^\dagger h_{l\sigma'}c_{j\sigma'}^\dagger, \tag{4.32}
\]

where \( (ij|V|kl) \) is the Coulomb matrix element for the electron-electron (ee), hole-hole (hh) and electron-hole (eh) interaction, and \( E_{ij}^e \) and \( E_{ij}^h \) are the matrix elements of the single-particle Hamiltonian of Eq. (4.30). In Eq. (4.32) the electron-hole exchange term has been neglected, reflecting that we are considering Wannier excitons, as opposed to Frenkel excitons.\[30\]

### The Hartree-Fock equations

In the Hartree-Fock (HF) approximation the wavefunction \( |\Psi_0\rangle \) is a single configuration of the form (4.31). The optimal single-particle states are found by requiring that the expectation value of the Hamiltonian (4.32) to be stationary with respect to variations in these states. Here, the HF equations in the form relevant to the QD model are presented; for more on the HF approximation see Sec. 2.3.

The Fock operator \( \hat{f} \), defined here by its action on the single-particle state \( |\psi_i\rangle = a_i^\dagger |\text{vac}\rangle \), is of the form

\[
\hat{f}|\psi_i\rangle \equiv \left[ \hat{h} + \sum_j \hat{J}_j - \sum_j \hat{K}_j \right] |\psi_i\rangle = \epsilon_i |\psi_i\rangle, \tag{4.33}
\]

where \( \hat{h} \) is the one-body part (see Eq. (2.29)), and the Coulomb and exchange operators \( \hat{J} \) and \( \hat{K} \) are defined by

\[
\hat{J}_j|\psi_i\rangle = \langle \psi_j|V|\psi_i\rangle |\psi_i\rangle \tag{4.34}
\]

\[
\hat{K}_j|\psi_i\rangle = \langle \psi_j|V|\psi_i\rangle |\psi_j\rangle. \tag{4.35}
\]

Since we neglect the \( e - h \) exchange the term (4.35) will be zero if the the indices \( i \) and \( j \) refer to different species. In solving Eq. (4.33) we require that the \( z \)-component of the spin as well as the total spin are good quantum numbers, and furthermore that the HF state is a spin singlet (\( S = 0 \)), \( i.e. \), an equal number of electrons and holes are present in the dot.\[7\] The restricted set of spin-orbitals is then of the form

\[
\psi(r) = \chi(r) \begin{pmatrix} \alpha \\ \beta \end{pmatrix}, \tag{4.36}
\]

where the spinors \( (\alpha, \beta) = (1, 0) \) and \( (\alpha, \beta) = (0, 1) \) refer to spin up and down, respectively. Inserting \( \psi(r) \) in (4.36) into the Hartree-Fock equation (4.33) an equation for only the spatial part \( \chi(r) \) can be derived. Expanding \( \chi(r) \) in a finite basis consisting

---

*In the case of charged dots this requirement is relaxed, since the charged states are spin triplets.*
of $k$ (Gaussian) functions $\{\phi_{\nu}\}$ we have $|\chi_i\rangle = \sum_{\nu} c_{\nu i} |\phi_{\nu}\rangle$, and in this basis the HF equation becomes:

$$
\sum_{\nu=1}^{k} c_{\nu i} \langle \phi_{\nu} | \hat{f} | \phi_{\nu} \rangle = \epsilon_i \sum_{\nu=1}^{k} c_{\nu i} \langle \phi_{\nu} | \phi_{\nu} \rangle \\
\sum_{\nu=1}^{k} P_{\nu \mu} c_{\nu i} = \epsilon_i \sum_{\nu=1}^{k} S_{\nu \mu} c_{\nu i},
$$

(4.37)

where the Fock matrix $F_{\nu \mu}$ and the overlap matrix $S_{\nu \mu}$ have been introduced. In matrix notation Eq. (4.37) can be written compactly in the form

$$
\mathbf{F} = \epsilon \mathbf{S}. \quad (4.38)
$$

The explicit expressions for the matrix elements of $\mathbf{F}$ are

$$
F^{e}_{ij} = h^{e}_{ij} + \sum_{kl} P^{e}_{kl}(\langle ij | V | kl \rangle - \frac{1}{2} \langle il | V | kj \rangle) + \sum_{pq} P^{h}_{pq}(\langle ij | V | pq \rangle - \frac{1}{2} \langle ps | V |rq \rangle) + \sum_{ij} P^{e}_{ij} |pq|V|ij\rangle.
$$

(4.39)

The density matrix $P_{\mu \nu}$ introduced in Eq. (4.39) is defined as

$$
P_{\mu \nu} = \sum_{a} n_{a} c_{\mu a} c_{\nu a},
$$

(4.40)

where $a$ is an electron or hole orbital index and the occupation number $n_{a}$ takes the values $\{0, 1, 2\}$. Obtaining the HF solution within the chosen basis is thus equivalent to iteratively solving the pseudo eigenvalue problem (4.38) until self consistency is reached, as discussed in Sec. 2.3.

Configuration interaction

The Hartee-Fock model described above, while including interactions in a mean-field sense, does not describe the correlated motion of the carriers. The configuration interaction scheme, discussed in Sec. 2.4, on the other hand, provides the exact solution within the constraints of the single-particle basis set when carried out at the full CI level.

The QD CI program is implemented based on a string-based CI formalism, in which the Slater determinants are expressed as a product of $\alpha$ and $\beta$ spin strings. The $\alpha$ spin string is an ordered product of $n_{a}$ creation operators and a corresponding spin string contains the information about the particles with $\beta$ spin. For example, the string corresponding to three $\alpha$ electrons occupying the three lowest orbitals is written $\alpha(I_{\alpha}^{e} = (1, 1, 1, 0, 0, \cdots)) = a_{1}^{e}a_{2}^{e}a_{3}^{e}$. Since both electrons and holes are considered the $\alpha$ and $\beta$ strings are further subdivided into electron and hole spin strings, such that the whole string is expressed as $\langle \alpha(I_{\alpha}^{e})|\beta(I_{\beta}^{h})\rangle$. A given Slater determinant may then be written as a product of four $\alpha$ and $\beta$ spin strings operating on the vacuum state. A unique addressing of the determinants is achieved using the graphical representation scheme presented in Sec. 3.1, considering the $\alpha$ and $\beta$ spin strings of the electrons and holes separately. The CI coefficients are then expressed in terms of a four-index matrix, instead of the conventional vector form, and a given state $|CI\rangle$ for the electron-hole system is written as

$$
|CI\rangle = \sum_{I_{\alpha}^{e}, I_{\beta}^{h}, I_{\alpha}^{h}, I_{\beta}^{h}} C(I_{\alpha}^{e}, I_{\beta}^{h}, I_{\alpha}^{h}, I_{\beta}^{h})|\alpha(I_{\alpha}^{e})\beta(I_{\beta}^{h})\rangle.
$$

(4.41)

Since the electrons and holes are considered as different particles, the electron and hole spin strings commute. This, along with the omission of exchange interaction in the Hamiltonian (4.32), means that the electron and hole number operators (see Sec. 2.1) commute with the Hamiltonian and that, consequently, the number of electrons
Figure 4.4: Schematic picture of some configurations of a system with 3 excitons. The HF reference state is followed by configurations singly (S) and doubly (D) excited in the electron (e) and/or hole (h) spaces. Particles excited from the reference state are depicted by arrows with unfilled heads for clarity.

and holes constitute good quantum numbers. The CI-space is, in other words, divided into electron and hole subspaces and no excitations between these spaces are included in the computations. This is illustrated in Figure 4.4 in which a few configurations of a system of three excitons are schematically illustrated. Note that the doubly excited configurations of the type $D_{eh}$, consisting of single excitations in both the electron and hole spaces is included in the coupled cluster singles (CCS) model in the form of disconnected terms.

### 4.3 Carrier Relaxation

In a typical photoluminescence experiment electron-hole pairs are created deep in the substrate by laser excitation at energies above the band gap of the substrate. The charge carriers are transported to the QW region by drift and diffusion, while being cooled down to the band edge by emitting longitudinal optical (LO) phonons. After being confined in the QW the electron-hole pairs can either recombine radiatively or scatter into the quantum dot (or ring). These carriers can then relax either radiatively or non-radiatively, or by a longer relaxation chain involving both mechanisms. In this section the part of the relaxation process occuring in the QD is discussed.

#### 4.3.1 Radiative recombination rates

Within the EMA framework, the spontaneous photon-emission rate $\Gamma_{n-1,L\leftarrow N,R}$, for the transition from the $N$-exciton state $|N,R\rangle$ to the $(N-1)$-exciton state $|N-1,L\rangle$, can be written as:

$$\Gamma_{n-1,L\leftarrow N,R} = \frac{ne^2 E_p E_{ph}}{6\pi\hbar^2 c\epsilon_0 m} \sum_{\sigma} |\langle L, N-1 | P^-_{\sigma} | N, R \rangle|^2,$$  \hspace{1cm} (4.42)

where $n$ is the refractive index of the semiconductor, $E_p$ is the Kane matrix element and $E_{ph}$ is the emitted phonon energy. The operator $P^-_{\sigma}$ is the interband polarization operator which annihilates an electron-hole pair with a given spin projection ($\sigma = \uparrow$).
with Eq. (4.44) includes all configurations of the transitions between states with different numbers of charge carriers. The sum in the recombination density matrix contains the many-body information as an contribution when the electron in spin-orbital \( I \) and phonons:

\[
\gamma_\sigma^{\tau,\sigma} = \langle \mathcal{L}, N - 1 | c_i \hbar \sigma | N, \mathcal{R} \rangle = \sum_I c_i^{N-1,\mathcal{L}} c_i^{N,\mathcal{R}}.
\]

The elements of the recombination density matrix contain the many-body information about transitions between states with different numbers of charge carriers. The sum in Eq. (4.44) includes all configurations of the \( (\mathcal{L}, N - 1) \) state (with coefficients \( C_i^{N-1,\mathcal{L}} \)) and \( C_i^{N,\mathcal{R}} \) denotes the CI coefficients of the \( |N, \mathcal{R}\rangle \) state that give a non-vanishing contribution when the electron in spin-orbital \( i \) and the hole in spin-orbital \( r \) are annihilated. The computation of the recombination density matrix is accomplished by a generalization of computational methods for evaluating the transition density matrix for non-orthogonal states in atoms and molecules.

### 4.3.2 Phonon-relaxation rates

In bulk and QW’s the dominant mechanism of carrier relaxation is LO-phonon emission. In QD’s, the discrete energy levels render this process ineffective, and the intraband relaxation proceeds by the slower longitudinal acoustic (LA) phonon emission. In QD’s, the discrete energy levels render this process ineffective, and the dominant mechanism of carrier relaxation is LO-phonon emission.

In bulk and QW’s, the dominant mechanism of carrier relaxation is LO-phonon emission. In QD’s, the discrete energy levels render this process ineffective, and the intraband relaxation proceeds by the slower longitudinal acoustic (LA) phonon emission. In QD’s, the discrete energy levels render this process ineffective, and the dominant mechanism of carrier relaxation is LO-phonon emission.

In QD’s, the discrete energy levels render this process ineffective, and the intraband relaxation proceeds by the slower longitudinal acoustic (LA) phonon emission. In QD’s, the discrete energy levels render this process ineffective, and the dominant mechanism of carrier relaxation is LO-phonon emission.

In contrast to the radiative relaxation discussed above, the initial and final states in Eq. (4.47) have an equal number of excitons. The expression can therefore be cast

\[ H_{LA} = \sum_{ij} \sum_{q} \langle i | \psi^e(q) | j \rangle c_i^\dagger c_j \left( b_q^\dagger + b_{-q}^\dagger \right) + \sum_{rs} \sum_{q} \langle r | \psi^h(q) | s \rangle h_r^\dagger h_s \left( b_q^\dagger + b_{-q}^\dagger \right), \]

with

\[ w^{e,h}(q) = \sqrt{\frac{\hbar q}{2V\rho v}} D^{e,h}_{rs} e^{-i\mathbf{q} \cdot \mathbf{r}_{s,h}}, \]

where \( \rho \) is the density of the material, \( D^{e,h} \) are the deformation potentials for the electrons and holes, \( v \) is the longitudinal sound velocity (\( \hbar \omega_q = \hbar v q \)) in the material, and \( b_q \) annihilates an acoustic phonon with the momentum \( q \). The transition matrix element between the initial, \( \Psi_I \), and final, \( \Psi_F \), states can then be written as

\[ \langle \Psi_F | H_{LA} | \Psi_I \rangle = \sum_{ij} \langle \mathcal{L}, N | c_i^\dagger c_j | N, \mathcal{R} \rangle w_{ij}^e(q) + \sum_{rs} \langle \mathcal{L}, N | h_r^\dagger h_s | N, \mathcal{R} \rangle w_{rs}^h(q). \]

Here the initial phonon state is assumed to be the vacuum and the final state is the one with one phonon with momentum \( q \) with all other phonon states being empty. The process thus describes the electronic transition connected with a spontaneous emission of a phonon, which is the relevant kind of process at very low temperatures. In contrast to the radiative relaxation discussed above, the initial and final states in Eq. (4.47) have an equal number of excitons. The expression can therefore be cast

\[ \text{At room temperature a thermal (Bose) distribution for the phonons must be assumed for the initial state. A description of the complete dynamics of the multiexcitons must then take induced absorption and emission of phonons into account. The generalization to treat the thermal dependency of the rates induced by interaction with acoustic phonons is straightforward.} \]
into a more practical form by introducing the transition density matrix, expressed in terms of the CI coefficients for electrons as

$$\rho_{ij}^{e,LR} = \langle \mathcal{L}, N | c_i^\dagger c_j | N, \mathcal{R} \rangle = \sum_I C_{ij}^{N,L} C_{ij}^{N,R}. \quad (4.48)$$

The corresponding expression for holes is obtained by the obvious substitutions $e \rightarrow h$ and $c_i \rightarrow h_i$. In Eq. (4.48) the index $I$ includes configurations in which orbital $j$ is occupied, and $I_j^i$ refers to the configurations obtained from $I$ by replacing the spin orbital $j$ with $i$.

After summing over $i, j$ and $r, s$, Eq. (4.47) reduces to a function of $q$. Applying Fermi’s golden rule then gives

$$\Gamma_{N,\mathcal{L} \leftrightarrow N, \mathcal{R}} = \frac{2\pi}{\hbar} \int dq |F(q)|^2 \delta(E_{N,\mathcal{R}} - E_{N,\mathcal{L}} - \hbar \omega_q) \quad (4.49)$$

where

$$F(q) = \sum_{ij} \rho_{ij}^{e,LR} w_{ij}^{e}(q) + \sum_{rs} \rho_{rs}^{h,LR} w_{rs}^{h}(q) \quad (4.50)$$

and $\delta(E_{N,\mathcal{R}} - E_{N,\mathcal{L}} - \hbar \omega_q)$ ensures that the energy condition is fulfilled. The integration in Eq. (4.49) is most conveniently done in spherical coordinates. When Gaussian basis functions are employed, the integral can be evaluated analytically for one of the two angular variables, whereas the integration in the remaining angular direction is done using quadrature.
5 BECs

Bosons are particles with integer spin, and the wave function for a system of identical bosons is symmetric with respect to the interchange of any two particles, as shown formally in Eq. (2.14). As a consequence of this symmetry, there are no restrictions on the occupation number of bosonic single-particle states. It is therefore possible that, below a certain temperature, a finite fraction of the particles in a bosonic system occupy the lowest-lying single-particle state, a phenomenon which was first discussed by Einstein in 1924. Over 70 years elapsed between Einstein’s theoretical prediction and the first experimental realization of this phenomenon, dubbed Bose–Einstein condensation (BEC). In 1995 many years of experimental work on ultracold gases culminated in the observation by Anderson et al. of BEC in a dilute atomic vapor of $^{87}$Rb atoms. Within a few months other groups made similar observations in a gas of Na atoms and spin-polarized $^7$Li atoms.

What was behind this seemingly sudden success? Rather than some important single breakthrough these experiments may be seen as the result of a long line of work on trapping and cooling neutral atoms along with the identification of suitable atomic species. By combining laser and evaporative cooling for alkali atoms in magneto-optical traps the temperatures and densities required to observe BEC were finally reached.

In the 15 years since the first realizations of BEC the field has seen much development and diversification both in the experimental and theoretical realms. Some major experimental examples include the observation of interference of two overlapping matter waves, of long-range phase coherence, of quantized vortices and vortex lattices, and molecular condensates with bound pairs of fermions. Common to all these phenomena is the existence of a coherent, macroscopic matter wave in an interacting many-body system, a concept familiar from the phenomena of superfluidity and superconductivity. In the following sections the trapping, cooling and general physical properties of ultracold dilute gases of alkali atoms are briefly presented, followed by a description of the theoretical modeling of these systems.

5.1 General Properties

As long as an atom’s (thermal) deBroglie wavelength $\lambda_{\text{dB}} = \hbar/(2\pi mk_B T)^{1/2}$ (the uncertainty in position due to thermal motion) is small compared to interatomic distances its motion may be described by classical trajectories. When $\lambda_{\text{dB}}$ is of the same order as the interparticle separation $n^{-1/3}$ (with $n$ denoting the density) the atomic wave functions begin to overlap and the atomic gas starts becoming a “quantum soup” of indistinguishable particles. Bosons undergo a transition to a condensate state at a precise temperature, which for an ideal gas is related to the peak atomic density $n$.

*In 1998 the first experimental evidence of condensation in a superfluid was observed. Since signs of superfluidity in liquid He was observed in 1938 (and connected theoretically to BEC by Fritz London) one may argue that the experiments in 1995 signified the first realization of BEC in dilute atomic vapors, but were late in being the first to produce a system of bosons in the degenerate regime by some 57 years.
by $n\lambda_B^3 = 2.612$. In the case of fermionic atoms, cooling gradually brings the system towards a state where the $N$ lowest single-particle states are occupied, i.e., a Fermi sea.

The apparent simplicity of cooling an atomic gas into the desirable regime belies the fact that, in most cases, quantum degeneracy is pre-empted by a transition into a liquid or solid state. To avoid this the three-body collisions leading to molecule formation need to be suppressed. The timescale for molecule formation is inversely proportional to the square of the density, whereas the rate of binary collisions, which serve to equilibrate the gas, is proportional to the density\textsuperscript{24} In order to avoid the more conventional condensed states the gas must thus be sufficiently dilute. In practice densities some one hundred times lower than normal air are required, in conjunction with temperatures in the nanokelvin regime.

5.1.1 Trapping and cooling

The temperatures and densities required for BEC are achieved by the interactions between the atoms and magnetic and optical fields. A typical experiment might proceed as follows.\textsuperscript{53} A beam of alkali atoms emerges from an oven at a temperature of some hundreds of Kelvins. The beam is then passed through a Zeeman slower\textsuperscript{56} in which a laser beam propagates in the direction opposite to the atomic beam, and the radiation force produced by the absorption of photons retards the atoms to a temperature of the order of 1K. At this stage the atoms are slow enough to be captured by a magneto-optical trap (MOT)\textsuperscript{57,58} in which they are further cooled by interaction with laser light to the microkelvin range.\textsuperscript{59} The laser beams are now switched off and the atoms are trapped by a purely magnetic trap.\textsuperscript{60} By allowing the trap to gradually become “shallower” the most energetic atoms escape, lowering the average energy of the remaining vapor. This \textit{evaporative cooling}\textsuperscript{61} marks the final stage in reaching the degenerate regime.

The trap in which the condensation takes place, and which consequently forms the external potential in our model, confines the atoms through the interaction of an inhomogeneous magnetic field and the magnetic moment $\mu$ of the atoms, i.e., due to the Zeeman effect. The energy of an atom in a particular state $i$, assuming linearity in the magnetic field $B$, is

\begin{equation}
E_i = C_i - \mu_i B, \tag{5.1}
\end{equation}

where $C_i$ is a constant. If the magnetic moment is positive (negative) the atom will experience a force towards regions of higher (lower) field. A local maximum in $|B|$ is impossible in regions with no electrical currents, so \textit{low-field seeking} states with a negative magnetic moment in conjunction with a field possessing a local minimum is needed to achieve magnetic trapping. Several different magnetic field configurations have been implemented, such as the quadrupole trap,\textsuperscript{55} Ioffe-Pritchard trap\textsuperscript{57} and the time-averaged orbiting potential (TOP) trap, which was employed in the pioneering experiment by Anderson \textit{et al.}\textsuperscript{42} In the TOP trap a rotating, spatially uniform field is superimposed on a quadrupole field, in order to inhibit transitions from low-field seeking states to high-field seeking ones through inelastic scattering, a process which may occur in pure quadrupole traps due to a vanishing magnetic field at the origin.\textsuperscript{55} Consider, then, the case in which the quadrupole field is of the form $B = B' (x, y, -2z)$, and the oscillating field has the components $(B_0 \cos \omega t, B_0 \sin \omega t)$ in the $xy$-plane. The instantaneous field is thus given by

\begin{equation}
B = (B' x + B_0 \cos \omega t, B' y + B_0 \sin \omega t, -2B' z), \tag{5.2}
\end{equation}

where the frequency $\omega$ of the time-dependent field is chosen to be low compared with the frequency of transitions between the magnetic substates of the atoms, but large compared to the frequency of atomic motions. The atoms may then be described as
experiencing a time-averaged field which, close to the origin, is given by
\[ \langle B \rangle_t \approx B_0 + \frac{B'^2}{4B_0}(x^2 + y^2 + 8z^2). \] (5.3)

The field in Eq. (5.3) has a nonzero minimum, thus circumventing the “hole-in-the-trap” problem of quadrupole traps. The magnetic contribution to the energy of an atom in the magnetic substate \( i \) is thus given for small \( r \) by
\[ E_i(\langle B \rangle_t) \approx E_i(B_0) - \mu_i(B_0) \frac{B'^2}{4B_0}(x^2 + y^2 + 8z^2), \] (5.4)
where \( \mu_i = -\frac{\partial E_i}{\partial B} \bigg|_{B_0} \) is the projection of the magnetic moment in the direction of the magnetic field. The oscillating field thus converts the linearly varying field of the quadrupole trap to one which depends quadratically on the distance from the origin. For traps of this configuration the external potential thus corresponds to an anisotropic harmonic oscillator potential.

### 5.1.2 Interactions between alkali atoms

As a consequence of the low densities at which BEC is observed it is possible to reliably model the system considering only binary collisions between the atoms. Understanding the low-energy scattering properties of the atoms involved is therefore necessary in modeling BECs.

In general the scattering of cold alkali atoms is a multi-channel problem, due to the existence of several hyperfine states of the atoms involved. One practical consequence of inelastic scattering is the loss of atoms from the trap, as mentioned in Sec. 5.1.1. Although the atom-atom interaction potentials cannot be calculated to the required accuracy from theory alone, the cooling methods described in the previous section have greatly increased understanding of low-energy atomic collisions, thereby making it possible to successfully model the interactions as we shall see below.

The problem at hand is thus to formulate a description of the binary scattering of alkali atoms at very low energies and densities. Alkali atoms belong to group I of the periodic table, and therefore have one valence electron in an \( s \)-type shell (with angular momentum \( L = 0 \)). The electronic spin \( (S = \frac{1}{2}) \) is coupled to the nuclear spin \( I \) through the hyperfine interaction. The total spin of the system is thus \( F = I \pm 1/2 \), where isotopes with \( I \) of half-integer value are necessary for the total spin to be an integer and, consequently, the atom to obey bosonic statistics. The interaction potential is strongly dependent on the internal atomic states. For example, for two rubidium atoms in the ground state the potential corresponding to a singlet spin state has a depth of about 6000 K at a separation of \( 8 a_0 \) \((a_0 \approx 0.53 \text{ Å} \) is the Bohr radius), whereas the triplet potential has a minimum at \( \sim 11 a_0 \) and a depth of only a few hundred K. The short-range potential is thus composed of a strongly repulsive part arising from the core electrons followed by an attractive contribution from the valence electrons responsible for covalent bonding.

For large atomic separations the interaction is of the van der Waals form, which, although much weaker than the covalent bonding plays an important role in determining the effective range of the potential. The van der Waals potential has the form \( U(r) = -C_6/4r^6 \) (in atomic units), which introduces a characteristic length scale \( r_0 \), which by dimensional arguments may be estimated to be
\[ r_0 \approx (C_6 m/m_e)^{1/4} a_0, \] (5.5)
where \( m \) and \( m_e \) are the atomic and electronic masses, respectively. The van der Waals coefficients \( C_6 \) for alkali atoms are of the order of \( 10^3 \) to \( 10^4 \), and atomic masses of the order of \( 10^9 A \times \) the electronic mass, where \( A \) is the mass number. From Eq. (5.5) one thus finds that \( r_0 \) is of the order \( 10^2 \ a_0 \).
Two other pertinent length scales are the thermal de Broglie wavelength $\lambda_{\text{dB}}$, as defined above, and the average interparticle separation $n^{-1/3}$. In terms of the Bohr radius $a_0$ the average interparticle spacing is of the order $n^{-1/3} \sim 10^3 a_0$, whereas thermal wavelengths may be as large as $\lambda_{\text{dB}} \sim 10^4 a_0$.

A proper theoretical framework for studying interactions in dilute gases is provided by the formalism of scattering theory, in which the scattering of projectile particles off a target is “detected” at a distance much larger than the range of the scattering potential. In the time-independent formulation of scattering theory the wave function describing the relative motion of the particles is written as a sum of the incident plane wave and a scattered wave $\psi_{\text{inc}}(r)$:

$$\psi = e^{ikz} + \psi_{\text{sc}}(r),$$

(5.6)

where the incident beam is assumed to propagate in the $z$-direction, and $r$ is the coordinate of relative motion. For large $r$ and a spherically symmetric potential the scattered wave is of the form $\psi_{\text{sc}}(r) = f(\theta)e^{ikr/r}$, where $f(\theta)$ is the scattering amplitude. This asymptotic wave function is conveniently expanded as an infinite sum of spherical waves with angular momenta $l = 0, 1, 2, \ldots \infty$. A general result for scattering at low energies is that there is always an energy below which only $s$-waves contribute to the scattering. In this limit the scattering amplitude approaches a constant, the scattering length $a$, and the asymptotic wave function becomes $\psi = 1 - a/r$. The scattering length may thereby be identified as the point of intersection of the asymptotic wave function with the $r$-axis, with positive values corresponding to a repulsive potential and negative values to an attractive interaction. A qualitative picture of the physical effect of the scattering length is provided by imagining the relative wave function confined in a box, the length of which depends on the sign and magnitude of $a$. If for $a = 0$ the box is of size $L$ then a positive $a$ corresponds to changing the length to $L - a$, whereas for attractive interactions the wave function’s energy is lowered, corresponding to a larger box of size $L + a$.

A fundamental result in the theory of dilute gases for which the $s$-wave scattering approximation holds, is that the interaction may be described in terms of an effective interaction $U_0(r) \propto \delta(r)$. $U_0(r)$ describes the interactions among long-wavelength, low-frequency degrees of freedom of a system by taking into account coupling between them via interactions with higher-energy degrees of freedom. One way to derive this result is by formulating the two-particle scattering problem in terms of the Lippmann-Schwinger equation for the scattering matrix $T(k', k; E)$ in the momentum representation:

$$T(k', k; E) = U(k', k) + \frac{1}{V} \sum_{k''} U(k', k'') \left( E - \frac{\hbar^2 k''^2}{m} + i\delta \right)^{-1} T(k'', k; E),$$

(5.7)

where $U(k', k)$ is the Fourier transform of the atom-atom interaction, and the infinitesimal imaginary term $i\delta$ ensures the proper boundary condition for outgoing waves. By performing the summation in Eq. (5.7) first over wave vectors larger than some critical value $k_c$ (corresponding to an energy $\epsilon_c = \hbar^2 k_c^2/m$) one obtains a quantity $\tilde{U}(k', k; E)$, which is then used as the interaction in performing the second part of the summation, restricted to wave vectors with $k'' < k_c$. In this way it is seen that by using $\tilde{U}$ as the interaction in a scattering problem in which only intermediate states with energies lower than $\epsilon_c$ appear explicitly the correct scattering matrix is obtained. If only interactions between very long wavelength excitations are considered, i.e., for small $k_c$, the effective interaction becomes constant:

$$\tilde{U}(0, 0; 0)\bigg|_{k_c \to 0} = \frac{4\pi \hbar^2 a}{m} \equiv U_0.$$

(5.8)
5.2 The model

The Fourier transform of \( U_0 \) then gives the effective interaction in coordinate space:

\[
U_0(r) = \frac{4\pi\hbar^2a}{m}\delta(r),
\]

(5.9)

This use of the two-body \( T \)-matrix in an energy expression has been shown to be a rigorous procedure and fully justified as a valid approximation by Stoof et al.\(^{63}\)

An alternative way of illustrating the meaning of the effective interaction is in terms of the mean interaction energy of the many-body system, which becomes\(^{64}\)

\[
\langle E_{\text{int}} \rangle = \frac{1}{2} \cdot \frac{4\pi\hbar^2a}{m} \sum_{ij} |\Psi(r_{ij} \to 0)|^2,
\]

(5.10)

where \( \Psi \) is the many-body wave function and the limit \( r_{ij} \to 0 \) means, in this context, that the interatomic separation is large compared to the scattering length \( a \) but small compared to all other characteristic lengths; equivalently one may understand this as taking the average of \( |\Psi|^2 \) over a volume \( V \gg a^3 \).

Under the conditions typical for BEC in dilute alkali gases the interatomic interaction may thus be described by a single parameter, the \( s \)-wave scattering length \( a \), which enters the problem in the form of an effective interaction. The actual value of the scattering length depends sensitively on the details of the short-range part of the potential, and thereby on the hyperfine states of the atoms involved, external fields \( \text{etc.} \) This sensitivity is readily demonstrated by analytic calculations of \( a \) using simple models, in which the short-range part of the potential depends on some parameters, \( \text{e.g.} \), the radius of a hard-sphere potential, which is varied to produce scattering lengths ranging between \( +\infty \) and \( -\infty \).\(^{55, 65}\) It is, in fact, possible to “tune” the value of the scattering length for a system from positive to negative (or vice versa) by applying an external magnetic or optical field, thereby changing the effective interaction from repulsive to attractive. This is possible due to the occurrence of Feshbach resonances\(^{66}\), the description of which requires a multi-channel approach to the scattering problem and will not be discussed here.

Scattering lengths for alkali atoms have been determined both experimentally and theoretically. It has been shown by a semiclassical treatment that in a cooled gas sample a repulsive interaction is statistically three times as likely as an attractive one\(^{67}\) and other levels of theory have been applied in calculating interaction potentials and scattering lengths for different species.\(^{55, 65, 59}\) A number of experimental techniques have been developed for measuring scattering lengths, notably photoassociative spectroscopy.\(^{70}\) Experimental values from several groups for Li, Na, K, Rb and Cs are tabulated in a review article by Weiner et al.\(^{65}\)

5.2 The model

5.2.1 The Hamiltonian

The many-body Hamiltonian considering one- and two-body interactions expressed within second quantization is

\[
\hat{H} = \sum_{pq} h_{pq}a_p^\dagger a_q + \frac{1}{2} \sum_{pqrs} g_{pqrs}a_p^\dagger a_r^\dagger a_s a_q,
\]

(5.11)

where the creation and annihilation operators obey the bosonic algebra in Eqs. \((2.4-2.5)\) and \( h_{pq} \) and \( g_{pqrs} \) represent the one- and two-body matrix elements, respectively. The one-particle operator contains kinetic and potential energy contributions, and the matrix elements are written as

\[
h_{pq} = \int \phi_p^*(r) \left( -\frac{1}{2} \nabla^2 + V_{\text{ext}}(r) \right) \phi_q(r) \, dr,
\]

(5.12)
where the $\phi(r)$ are orthonormal single-particle states. The two-body matrix element is, in turn

$$g_{pqrs} = \int \int \phi^*_p(r_1)\phi^*_r(r_2)V(r_1,r_2)\phi_q(r_1)\phi_s(r_2)dr_1dr_2. \quad (5.13)$$

For the external potential $V_{\text{ext}}(r)$ we consider two cases, corresponding to alkali atoms in a parabolic trap as well as a fictitious model system of bosonic particles in an atomic Coulomb field. In the parabolic case the potential may be anisotropic and is thus of the form

$$V_{\text{BEC}}^\text{ext}(r) = \frac{1}{2}(\omega_x^2 x^2 + \omega_y^2 y^2 + \omega_z^2 z^2), \quad (5.14)$$

with the $\omega_i$ representing the trapping frequencies in the three Cartesian directions. In Eq. (5.14) the length scale $L$ is defined by

$$[L] = \sqrt{\frac{\hbar}{m\omega}}, \quad (5.15)$$

where $m$ is the mass of the trapped particles and $\omega$ refers to the lowest trapping frequency in the case of an anisotropic potential. In addition energies are measured in units of $\hbar\omega$ yielding dimensionless equations in harmonic oscillator (HO) units. In the latter case the confining potential is (expressed in atomic units)

$$V_{\text{ext}}^\text{at}(r) = -Z|\mathbf{r}|, \quad (5.16)$$

where $Z$ is the nuclear charge.

The boson-boson interaction term in the alkali atom model is, as was elaborated in Sec. 5.1.2, of the form

$$V_{\text{BEC}}(r_1,r_2) = 4\pi a\delta(r_1-r_2), \quad (5.17)$$

where $a$ is the s-wave scattering length. For reasons to be discussed in Sec. 7.2 we have also implemented an interaction potential of the form

$$V_{\text{BEC}}(r_1,r_2) = N_\theta e^{-\theta(r_1-r_2)^2}, \quad (5.18)$$

where the normalization factor $N_\theta = 4\pi a(\theta/\pi)^{3/2}$, such that this s-type Gaussian function reduces to the form (5.17) in the limit $\theta \to \infty$.

In the atomic case the interaction term describes the electrostatic two-body interaction between bosons of charge $e$ and is therefore written as

$$V^\text{at}(r_1,r_2) = \frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}. \quad (5.19)$$

### 5.2.2 The mean-field model

The mean-field model most commonly employed for modeling dilute systems of interacting bosons is the Gross-Pitaevskii-model. The ansatz many-body wave function is one in which all particles occupy the same one-body state $\phi_0(r)$:

$$\Psi(\mathbf{r}) = N\prod_{i=1}^{N} \phi_0(\mathbf{r}_i). \quad (5.20)$$

Minimizing the expectation value of the Hamiltonian in the state (5.20) under the constraint that $\langle \phi_0 | \phi_0 \rangle = 1$ yields, in terms of the order parameter $\psi(\mathbf{r}) \equiv \sqrt{N}\phi_0(\mathbf{r})$:

$$-\frac{\hbar^2}{2m}\nabla^2\psi(\mathbf{r}) + V_{\text{ext}}(\mathbf{r})\psi(\mathbf{r}) + U_0|\psi(\mathbf{r})|^2\psi(\mathbf{r}) = \mu\psi(\mathbf{r}), \quad (5.21)$$
where \( U_0 = 4\pi\hbar^2 a/m \) and \( \mu = \delta(H)/\delta N \) is the chemical potential. In deriving the Gross-Pitaevskii (GP) equation (5.21), the approximation \( N - 1 \approx N \) has been made, which is the only distinction between the GP model and a Hartree-Fock model with an interaction term of the form (5.17).

In our approach the Hartree-Fock scheme (see Sec. 2.3) is employed in order to obtain a reference state for our CI model. We solve the HF-equations by expanding the single-particle states in a finite set of non-orthonormal basis functions \( \{\chi_i(r)\} \):

\[
\phi_i(r) = \sum_{j=1}^{n_b} c_{ij} \chi_j(r),
\] (5.22)

where \( n_b \) is the number of basis functions. Quantities represented in a non-orthonormal basis will henceforth be denoted with a tilde. The Fock operator is thus represented in matrix form, and the HF equation becomes

\[
\tilde{F}(\tilde{D})C = \tilde{S}CE,
\] (5.23)

where \( \tilde{F} \) is the Fock matrix, which is a function of the one-particle density matrix \( \tilde{D} \). The overlap matrix \( \tilde{S} \) serves as the metric, and \( C \) is the matrix containing the eigenvectors. The Fock matrix is constructed as

\[
\tilde{F}_{pq}(\tilde{D}) = \delta_{pq} + \frac{N-1}{2} \left( \sum_{rs} \tilde{g}_{pqrs} \tilde{D}_{rs} + \sum_{rs} \tilde{g}_{psrq} \tilde{D}_{rs} \right).
\] (5.24)

Since the HF ansatz is of the form (5.20), the density matrix is obtained from the expansion coefficients of the lowest-lying orbital as

\[
\tilde{D}_{rs} = c_{r1}c_{s1}.
\] (5.25)

In Eq. (5.24) the first term in the sum is the classical interaction term and the second the exchange interaction. In the case of an interaction potential proportional to \( \delta(r_1 - r_2) \) these terms are identical. This is the case when solving for the energetically lowest state for an arbitrary interaction potential. The unoccupied orbitals, however, are affected by the exchange term, which in turn may modify the results at a restricted CI level. The exchange interaction is therefore always included in our calculations.

### 5.2.3 The CI model

The configuration interaction scheme has been presented generally in Sec. 2.4 and for a system of electrons and holes in Sec. 4.2.6. In the case of bosons, the main difference compared to fermions is that the number of configurations at a restricted CI level is independent of the number of particles \( N \). For \( K \) single-particle states the number of configurations at the \( M \)th CI level is

\[
N_{\text{conf}} = \sum_{i=0}^{M} \binom{K+i-1}{i}.
\] (5.26)

The number of basis functions may need to be increased as a function of \( N \), but not out of necessity, as for fermions, but rather out of need to better describe the interparticle correlations.

As in the fermionic case, an efficient addressing scheme of the configurations is needed. The graphic addressing strategy for fermions has been generalized for bosons, as described in Sec. 3.1. Since full diagonalization of the Hamiltonian is unfeasible beyond a certain number of included configurations the direct CI algorithm described in Sec. 3.2 for finding the lowest few eigenstates and eigenenergies has been implemented in the boson program, whereby calculations larger by several orders of magnitude are possible.
Part III

Results
Studied systems

The theoretical models and computational methods described in the previous sections have been applied to a variety of physical systems. The quantum dot program has been used for calculating total energies of multiexciton complexes at the CI and CC levels as well as radiative recombination rates and energies for various neutral and charged exciton systems. Phonon-relaxation rates for a few low-lying states of the exciton were also computed. Systems with different confinement potentials have also been studied, namely single and double quantum rings. In the latter case experimental and theoretical results in the literature indicating recombination from states located in either one of the rings are re-examined at the CI level.

The first application for the software for bosonic systems was a study of fictitious atomic systems with bosonic electrons in place of the fermionic ones found in nature. The purpose of this work was to test the software on a well-defined problem for which computational elements such as basis sets and matrix element integrals were readily available. Furthermore, the first system in the series studied, “helium”, is equivalent to the fermionic one, thus providing a benchmark to compare against. In the second work, we apply the program on BECs at different densities, particle numbers and trap geometries.
6 Quantum Dots

6.1 CC and CI energies and recombination rates

The CI and CC methods described in Secs. 2.4, 2.5 and 4.2.6 have been employed in calculating the ground-state energies of a biexciton (2X) and a complex of six excitons (6X) in a InGaAs/GaAs QD sample, described in detail in Sec. 4.1. The single-particle basis set employed in these calculations consists of anisotropic Gaussian functions of the form

\[ \phi(x, y, z) = x^{l_x} y^{l_y} z^{l_z} e^{-a(x^2+y^2)-bz^2}. \]  (6.1)

The calculations presented here use a basis set consisting of 4s, 4p, and 3d functions, in the standard notation in which the letters s, p, d, ··· correspond to the angular momenta \( l_x + l_y + l_z = 0, 1, 2, \cdots \). In the z-direction a single Gaussian s function is used to describe the variation of the orbitals.

The ground-state energies obtained at excitation levels ranging from singles and doubles (SD) up to singles, doubles, triples and quadruples (SDTQ) are compared in Table 6.1. The exponential form of the CC wave function (2.37) means that some configurations with higher excitations than the coupled cluster level are included in the expansion; e.g., the CCSD expansion includes some triple and quadruple excitations within the CI framework. This is readily reflected in the obtained energies: the CC energies are consistently lower, and thus closer to the FCI limit, than the CI energies obtained at the same level. For the 6X system the CCSDTQ energy is assumed to be close to the FCI limit, i.e. \( E_{\text{CCSDTQ}} \approx E_{\text{FCI}} \). This assumption rests upon molecular electronic structure calculations which show that, indeed, CCSDTQ energies are close to the FCI limit.\(^{26, 77}\)

Table 6.1: Ground-state multiexciton energies (in meV) calculated for the biexciton (2X) and for a multiexciton complex consisting of six excitons (6X) at different CI and CC levels. The relative correlation energy (in %) as compared to the CCSDTQ values are given within parentheses. The corresponding HF SCF energies are -314.960 and -905.054 meV, respectively.

<table>
<thead>
<tr>
<th>Multiexciton</th>
<th>Method</th>
<th>SD</th>
<th>SDT</th>
<th>SDTQ</th>
</tr>
</thead>
<tbody>
<tr>
<td>2X</td>
<td>CI</td>
<td>-319.587 (93.6%)</td>
<td>-319.688 (95.6%)</td>
<td>-319.906(^a)</td>
</tr>
<tr>
<td>2X</td>
<td>CC</td>
<td>-319.733 (96.5%)</td>
<td>-319.833 (98.5%)</td>
<td>-319.906(^b) (100%)</td>
</tr>
<tr>
<td>6X</td>
<td>CI</td>
<td>-913.377 (84.1%)</td>
<td>-913.603 (86.4%)</td>
<td>-914.787 (98.4%)</td>
</tr>
<tr>
<td>6X</td>
<td>CC</td>
<td>-914.570 (96.1%)</td>
<td>-914.855 (99.0%)</td>
<td>-914.948 (100%)</td>
</tr>
</tbody>
</table>

\(^a\) A FCI calculation.
\(^b\) A FCC calculation.

Radiative recombination rates for the exciton, biexciton as well as the positively and negatively charged trions have been calculated using the method described in Sec. 4.3. The recombination energies and rates are reported in Table 6.2. Also given...
Table 6.2: The recombination energies (in meV) and radiative recombination rates ($\Gamma_{N-1,L\leftarrow N,R}$ in ns$^{-1}$) of charged and neutral exciton complexes calculated at the FCI level. The shifts of the corresponding recombination energy (in meV) relative to the recombination peak of the exciton are also given.

<table>
<thead>
<tr>
<th>Exciton</th>
<th>$E_{\text{recomb.}}$</th>
<th>$E_{\text{shift}}$</th>
<th>$\Gamma_{N-1,L\leftarrow N,R}$ (ns$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1X</td>
<td>1361.1</td>
<td></td>
<td>4.05</td>
</tr>
<tr>
<td>2X</td>
<td>1359.0</td>
<td>-2.17</td>
<td>3.02</td>
</tr>
<tr>
<td>1X$^+$</td>
<td>1359.1</td>
<td>-2.02</td>
<td>3.02</td>
</tr>
<tr>
<td>1X$^-$</td>
<td>1359.3</td>
<td>-1.81</td>
<td>3.16</td>
</tr>
</tbody>
</table>

$^a$ A band gap of 1520 meV is assumed.

are the stabilization energies $E_{\text{shift}}$ of the biexciton and charged trions with respect to the exciton. The negative energies imply that all the systems studied are bound with respect to the exciton and that the photoluminescence peaks of these are expected to be redshifted by about 2 meV with respect to the exciton transition.

Table 6.3 displays the calculated phonon relaxation rates for a few low-lying $\Delta_g$-states. By expressing the phonon relaxation rate in Eq. (4.49) in the atomic orbital (AO) basis, and performing the integration over $\mathbf{q}$ in spherical coordinates it can be shown that the relaxation process is strongly dependent on the width of the QW, such that the narrower the well the lower the relaxation rate. As seen from the table all the transitions correspond to an energy difference of less than 2 meV between the initial and final states. Since the relaxation rate also depends on the energy difference, states separated by $\sim 3$ meV or more are long-lived enough to contribute to the photoluminescence spectrum. A small energy difference does not, however, guarantee a large transition rate. Consider, for example, the transition $3\Sigma^+_g \rightarrow 3\Pi_g$, which has an energy separation of 0.068 meV. The $3\Sigma^+_g$ state is mainly composed of configurations in which both the electron and the hole occupy the first p-shell, whereas in the $3\Pi_g$ state the electron is predominantly in the s-shell and the hole in the second p-shell. At the HF level, the phonon transition moment between these states would be zero, and it is only the mixing of states due to electron-hole correlation that leads to a small, but non-zero transition rate of 32.38 ms$^{-1}$.

Table 6.3: Energy differences ($E_{\text{phonon}}$ in meV) and the phonon relaxation rates ($\Gamma_{N,L\leftarrow N,R}$ in ns$^{-1}$) from a few low-lying $\Delta_g$ states of the exciton confined in the InGaAs/GaAs quantum dot.

<table>
<thead>
<tr>
<th>Transition</th>
<th>$E_{\text{phonon}}$ (meV)</th>
<th>$\Gamma_{N,L\leftarrow N,R}$ (ns$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1\Delta_g \rightarrow 2\Sigma^+_g$</td>
<td>0.642</td>
<td>25.2</td>
</tr>
<tr>
<td>$2\Delta_g \rightarrow 3\Sigma^+_g$</td>
<td>1.582</td>
<td>15.0</td>
</tr>
<tr>
<td>$3\Delta_g \rightarrow 4\Sigma^+_g$</td>
<td>0.511</td>
<td>13.6</td>
</tr>
<tr>
<td>$4\Delta_g \rightarrow 5\Sigma^+_g$</td>
<td>0.599</td>
<td>14.5</td>
</tr>
<tr>
<td>$5\Delta_g \rightarrow 5\Sigma^+_g$</td>
<td>1.917</td>
<td>3.1</td>
</tr>
<tr>
<td>$2\Delta_g \rightarrow 3\Pi_g$</td>
<td>1.515</td>
<td>5.3</td>
</tr>
<tr>
<td>$3\Delta_g \rightarrow 4\Pi_g$</td>
<td>0.914</td>
<td>5.8</td>
</tr>
<tr>
<td>$5\Delta_g \rightarrow 4\Delta_g$</td>
<td>1.318</td>
<td>7.6</td>
</tr>
</tbody>
</table>
6.2 Quantum rings

Systems in which electrons and holes are confined in ring-like structures have been experimentally realized. By adding a repulsive center to the numerical confinement potentials described in Sec. 4.2.5, a ring-like potential is obtained. The potentials obtained by adding a repulsive Gaussian function of varying height to the center of the dot are shown in Fig. 6.1. The parameter $\lambda$ scales the height of the central repulsive part such that $\lambda = 0$ gives the original quantum dot potential and $\lambda = 1$ a quantum ring with a central barrier of maximum value equal to that of the QW potential. The single-particle basis set used in these calculations consists of 6$s$6$p$3$d$ functions of the form in Eq. (6.1) in the lateral direction and one $s$ function in the $z$-direction.

![Figure 6.1: The radial part of the electron (left) and hole (right) confinement potentials.](image1)

![Figure 6.2: The ground state energy of the biexciton confined in different ring-shaped potentials and the corresponding occupation of the HF reference state.](image2)

![Figure 6.3: Excitation energies for the first few singlet states of the biexciton for three different ring potentials.](image3)

One effect of the ring potential compared to a quantum dot is that the energy of the $\sigma$ states is increased, whereas states of $\pi, \delta, \ldots$-symmetry with a node at the origin are less affected. The energy spacing between the one-particle states becomes smaller, thus enabling a strong mixing of the states for the interacting many-body system. Correlation effects would thus be expected to be stronger for ring systems than for QDs.

The effect of the increased confinement is seen from the results of FCI calculations on the biexciton complex for various values of the scaling factor $\lambda$. The ground state energy increases by about 10% as $\lambda$ is increased from 0 to 1, as shown in Fig. 6.2.
In the same figure the relative weight of the HF reference state is also displayed. For the quantum dot the occupation is nearly unity, whereas for the ring with $\lambda = 1$ the reference state contributes only 53% to the total wave function, indicating strong correlation effects.

Full CI calculations of the lowest excited states of the quantum dot and quantum ring systems have been carried out, showing that the splitting between the first excited state and the ground state decreases with increasing $\lambda$. In Fig. 6.3 the excitation energies of the first few singlet states of the biexciton confined in quantum dot and the quantum rings with $\lambda = 0.4$ and $\lambda = 0.8$ are shown. The excitation energies of the $\Pi$ and $\Delta$ states decrease faster as a function of $\lambda$ than for the $\Sigma$ states. The small splitting between the ground and excited states in the quantum rings indicate that the wave function can be drastically influenced by external perturbations.

6.3 Double quantum rings

![Figure 6.4](image)

Figure 6.4: (a) Electron (left) and hole (right) densities obtained from a single-particle model. (b) FCI densities for the electron (left) and hole (right) part of the exciton. The six lowest lying $\Sigma$-states are shown.

Concentric double quantum rings (CDQRs) are systems in which charge carriers are confined in a cylindrically symmetric potential with two local minima. An example of such a potential is shown in Figures 6.4 and 6.5. The optoelectronic properties of these and related systems make them potential building blocks for nanoscale devices and a thorough understanding of their electronic structure and recombination dynamics is therefore called for. Photoluminescence (PL) experiments carried out on CDQR structures have indicated that the main features of the PL spectrum stem from recombination from states localized either in the inner (IR) or outer ring (OR) of the structure. The theoretical arguments for this interpretation rest on single-particle models, but as correlation effects are strong for single quantum ring as seen from the results above it is expected that the same holds for CDQRs as well. The calculations reported here employ the CI method to investigate the nature of the states involved in the radiative recombination processes observed in
6.3 Double quantum rings

The lateral confinement potential used in the calculations is based on an atomic force microscope (AFM) profile obtained from a CDQR sample. The confinement potential has the same form as the AFM profile with the minima normalized to 91.9 eV for the electrons and 25.9 eV for the holes. The effective masses used in the calculations are $m_{eij} = 0.065$, $m_{eiz} = 0.143$, and $m_{hiz} = 0.341$ with a dielectric constant equal to 13. The basis set used to describe the single-particle orbitals is of the same form, Eq. (6.1), as in the QD and quantum ring calculations above. For the lateral part of the wave functions an even-tempered basis consisting of 8 $s$, 7 $p$, and 5 $d$ functions is used, and in the $z$-direction a single $s$ function is included.

To compare our model with the experimental PL spectra we solve the FCI problem for a system consisting of one exciton in a confinement potential whose radial form is shown in Figures 6.4 and 6.5. Single particle densities plotted at $z = 0$ obtained from the non-interacting single-particle model are shown in Fig. 6.4 (a). At this level of theory, one of the two lowest-lying stated is centered (mainly) on the outer ring and the other on the inner ring both for the electron and the hole, which is in qualitative agreement with the calculations by Kuroda et al. In contrast to these calculations, we find the lowest-lying state localized in the IR and the second lowest in the OR. Including correlated interactions provides a different picture. The densities shown in Figure 6.4 (b) are obtained at the FCI level and do not show states mainly localized in the outer ring, at least not among the six energetically lowest-lying states in each of the irreducible representations of the symmetry group. Some states do indeed have a considerable density in the outer ring, notably ones with II- and $\Delta$-symmetry, shown in Fig. 6.5, but these do not contribute to the luminescence for symmetry reasons.

To investigate if the detailed form of the potential has qualitative effects on the densities a series of calculations were carried out in which the height of the barrier separating the IR and OR was varied. No significant changes in the character of the lowest states was found. Considerable effort was also expended to ensure that the

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* S. Sanguinetti (private communication).
basis sets employed were capable of describing the states located in the OR. Only by completely removing the inner potential minimum – in effect creating a single quantum ring – were solutions located in the OR obtained, showing the basis set to be adequate.

The low part of the calculated energy spectrum for the exciton is depicted in Fig. 6.6 in which the three luminescent Σ states are highlighted with thick lines. The majority of the states are dark, including one lying between the ground and two luminescent excited states.

One way to appreciate the importance of charge-carrier correlations in the system is by examining the chemical potential for multiexciton complexes at different levels of theory. The chemical potential is defined as \[ \mu(N) = E_g(N) - E_g(N-1), \]
where \( E_g(N) \) is the ground state energy for a system of \( N \) excitons. The calculated chemical potentials \( \mu(N) \) for 1–6 excitons are shown in Fig. 6.7. The importance of correlation is clearly seen: the chemical potential at the CISD and CISDT levels does not exhibit a clear shell structure, nor is the biexciton bound. Indeed, quadruple excitations are required for the picture in Fig. 6.7 (c) to emerge. This strong dependence on
correlation effects is to be expected based on the studies on single quantum rings reported above. The qualitative behaviour of the chemical potential is similar to that of “ordinary” quantum dots, but the energy difference between the $\sigma$ and $\pi$ shells is an order of magnitude smaller, i.e., around 1 meV.

Radiative recombination rates for the exciton (1X) and biexciton (2X) have been calculated within the FCI scheme using the expression (4.49). The low-energy peak in Fig. 6.8 contains contributions from the recombination of ground-state exciton as well as the biexciton. The other peak, which lies some 9 meV higher contains contributions from the biexciton as well as one or two of the lowest bright excited states of the exciton.

### 6.4 Summary and discussion

The importance of correlation effects in systems consisting of excitons bound in strain-induced quantum dots has been studied previously mainly by Braskén et al.\cite{33,87–90} and Corni et al.\cite{29,91,92} using earlier versions of the same software as used in the work reported here. Other groups have also employed different kinds of CI-approaches, albeit on different types of SAQDs and generally with much smaller CI spaces.\cite{93–96}

From these studies it is clear that correlation can have qualitative effects on properties of the QD systems. It has been found, for example, that the dot-size dependence of the radiative recombination rates of the ground state transition of the exciton is quite different at correlated and uncorrelated levels of theory.\cite{92} At the noninteracting and SCF level the recombination rate is independent of the diameter of the QD, whereas at the FCI level the rate exhibits a monotonic increase as a function of the diameter.

Also, at the HF level, the biexciton is not bound, whereas at the FCI level a binding energy of a few meVs is obtained.\cite{90} The chemical potential, defined as in Sec. 6.3, does not exhibit a well-defined shell structure at the HF level, but at the CISD level a shell structure in good agreement with experimental PL spectra emerges.\cite{87}

In the QD study included in this thesis the coupled cluster model is introduced. The calculations on multie exciton complexes show that the disconnected terms included in the CC wave function (see Eq. (2.40) and Fig. 4.4) lead to more correlation energy taken into account at a given level of theory as compared to the CI scheme. To date the code cannot calculate recombination rates at the CC level, although considering the importance of correlation in recombination it would seem to be an advantageous extension of the program.\cite{†}

When going from quantum dots to ring-shaped systems correlation effects become more pronounced. The FCI calculations reveal that the occupation of the HF reference state drops from close to unity to around 0.5 as the confinement potential is changed from a QD to a ring with a strong central barrier. The low end of the singlet excitation spectrum also shows a decrease in the splitting between the ground and excited states, indicating the presence of near-degeneration correlation effects.

In the CDQR study the focus was to investigate the possibility of recombination from states spatially located on either the inner or outer rings of the structure, as suggested by experiments and single-particle calculations. The calculations show that in order to obtain a shell structure in the chemical potential quadruple excitations need to be included at the CI level. The structure is similar to the QD case, but with a significantly reduced spacing of only $\sim$1 meV between the $\sigma$ and $\pi$ shells. These results on their own indicate strong correlation effects. Single particle densities obtained at different levels of theory reveal that at the noninteracting level states located mainly on the IR and OR are indeed found. At the FCI level, however, the states involved in the recombination processes are all located on the inner ring. The two main peaks characteristic to CDQR PL spectra are nevertheless reproduced by

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\footnote{\textsuperscript{†}The implementation is not, regrettably, entirely straight-forward.}
considering recombination from the ground and the first excited states, which can be expected to be sufficiently long-lived due to a suppression of the phonon-mediated intraband relaxation process. This phonon bottleneck effect has been predicted for zero-dimensional semiconductor systems \cite{38,39,97} and also experimentally observed \cite{98}. The phonon matrix elements \( w_{e,h}^{i,j}(q) \) occurring in the expression for the transition rate in Eq. (4.50) reveal, when evaluated in the AO basis as in the appendix of Braskén et al. \cite{33}, that the more extended the basis functions are in coordinate space (corresponding to small Gaussian exponents and a narrow distribution in momentum space) the less they contribute to the relaxation rate. The shape of the double ring potential with a maximum at the center suppresses the contributions of basis functions with small exponents; a phonon bottleneck could thereby explain the long excited-state lifetime.

It is to be noted, however, that time-resolved PL experiments \cite{99,100} indicate that there are other relaxation mechanisms that circumvent the bottleneck caused by the reduction in the LA-phonon emission rate. For example, an Auger-like process, in which an electron intraband transition to a lower level is followed by the emission of a hole from the QD, is suggested by Braskén et al. in order to explain the temperature dependence of the carrier relaxation \cite{100}. A rigorous study of the relaxation process should therefore take all plausible mechanisms into account. The relaxation process can then be simulated using a master equation approach \cite{100,101}. Since this kind of analysis has not been carried out for the CDQR system, the phonon bottleneck effect remains a tentative explanation for the occurrence of a PL signal from an excited state.

In summary it may be concluded that the studies reported here support the notion that carrier-carrier correlation plays an important role in the studied systems. In particular, as the excitons become more confined in ring or double ring structures the correlation effects become more prominent, as illustrated by the character of the states involved in recombination in CDQRs.
7 Bosons

7.1 Atomic test system

In order to assess the reliability of the configuration interaction program for bosons a model system of spinless charged bosonic particles in a central Coulomb field was constructed and investigated. The system is equivalent to an atom in which the electrons have been replaced by bosonic counterparts. The systems are thus referred to by the conventional atomic names “He”, “Li” et. c., according to the charge of the central Coulomb field.

The results of CISD calculations on a series of neutral atomic systems with atomic numbers from 2 to 10 are summarized in Table 7.1, and for comparison energies for the corresponding fermionic atoms are shown in Table 7.2. The total energy of the neutral bosonic atoms decreases much faster with increasing atomic charge than for fermionic atoms, a trend which is easily understood since in the bosonic systems mainly the 1s orbital is occupied. The absolute and relative correlation energies are also larger in the bosonic case, even though the basis sets used for bosons are not as close to the basis set limit as the fermionic ones. The gap between the highest occupied and lowest unoccupied orbital (the HOMO-LUMO gap) increases with increasing nuclear charge; for Ne it is about 3.3 a.u., suggesting that the wavefunction is dominated by a single configuration. Indeed, the relative contribution of the HF reference state to the wavefunction is over 99% in all cases studied.

The ground state energy as a function of the basis set and CI level is shown in Fig. 7.1 for selected systems. The Gaussian basis sets employed for He and Ne are listed in Table 7.3. The energy as a function of the basis set decreases monotonically, as seen in Fig. 7.1 (a). For He a clear saturation is observed, whereas for Ne a larger single-particle basis set would be needed for the basis set limit to be reached. It

Table 7.1: Ground-state energies (in a.u.) of He–Ne calculated at the HF and CISD levels. The last column lists the relative weight of the reference (HF) state in the CI calculation.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(a.u.)</td>
<td>(a.u.)</td>
<td>(a.u.)</td>
<td>(%)</td>
<td>(%)</td>
<td>(%)</td>
</tr>
<tr>
<td>He (2)</td>
<td>0.90453</td>
<td>-2.85990</td>
<td>-2.89953</td>
<td>-0.03962</td>
<td>1.37</td>
<td>99.21</td>
</tr>
<tr>
<td>Li (3)</td>
<td>0.89226</td>
<td>-8.54595</td>
<td>-8.65006</td>
<td>-0.10410</td>
<td>1.20</td>
<td>99.28</td>
</tr>
<tr>
<td>Be (4)</td>
<td>1.17565</td>
<td>-19.01726</td>
<td>-19.22035</td>
<td>-0.20308</td>
<td>1.06</td>
<td>99.30</td>
</tr>
<tr>
<td>B (5)</td>
<td>1.48540</td>
<td>-35.74211</td>
<td>-36.11439</td>
<td>-0.37228</td>
<td>1.03</td>
<td>99.37</td>
</tr>
<tr>
<td>C (6)</td>
<td>1.81432</td>
<td>-60.17947</td>
<td>-60.737501</td>
<td>-0.55802</td>
<td>0.92</td>
<td>99.43</td>
</tr>
<tr>
<td>N (7)</td>
<td>2.16450</td>
<td>-93.79480</td>
<td>-94.57573</td>
<td>-0.78093</td>
<td>0.83</td>
<td>99.50</td>
</tr>
<tr>
<td>O (8)</td>
<td>2.53604</td>
<td>-138.05206</td>
<td>-139.09254</td>
<td>-1.04048</td>
<td>0.75</td>
<td>99.54</td>
</tr>
<tr>
<td>F (9)</td>
<td>2.92903</td>
<td>-194.41461</td>
<td>-195.75217</td>
<td>-1.37555</td>
<td>0.68</td>
<td>99.58</td>
</tr>
<tr>
<td>Ne (10)</td>
<td>3.34349</td>
<td>-264.34620</td>
<td>-266.01824</td>
<td>-1.67204</td>
<td>0.63</td>
<td>99.61</td>
</tr>
</tbody>
</table>

* The number of bosons is given within parentheses.
Figure 7.1: (a) The ground-state energy as a function of basis set for He (thick line) and Ne (thin line) calculated at the CISD level. (b) The ground-state energy for Li (thin line) and Be (thick line) calculated at different CI levels. The FCI calculation on Be was performed using the direct iterative CI method.
Figure 7.2: Timings for the direct iterative CI algorithm. (a) FCI keeping the number of particles constant ($N_{\text{bosons}}=3$) and increasing basis-set size. The number of single particle levels is 24, 32, 40, and 48. (b) FCI keeping the basis set constant (17 single-particle levels) and increasing the number of particles (3, 6, 7, and 8).
Table 7.2: Estimated ground-state and correlation energies (in a.u.) of neutral fermionic He–Ne atoms calculated by C. Froese Fischer et al.\textsuperscript{102,103}

<table>
<thead>
<tr>
<th>Atom</th>
<th>Total Energy (in a.u.)</th>
<th>Correlation Energy (in a.u.)</th>
<th>Correlation Energy (in %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>-2.903724</td>
<td>-0.042044</td>
<td>1.47</td>
</tr>
<tr>
<td>Li</td>
<td>-7.47806</td>
<td>-0.04533</td>
<td>0.61</td>
</tr>
<tr>
<td>Be</td>
<td>-14.66736</td>
<td>-0.09434</td>
<td>0.65</td>
</tr>
<tr>
<td>B</td>
<td>-24.65391</td>
<td>-0.12485</td>
<td>0.51</td>
</tr>
<tr>
<td>C</td>
<td>-37.8450</td>
<td>-0.15640</td>
<td>0.42</td>
</tr>
<tr>
<td>N</td>
<td>-54.5892</td>
<td>-0.18831</td>
<td>0.35</td>
</tr>
<tr>
<td>O</td>
<td>-75.0673</td>
<td>-0.25794</td>
<td>0.35</td>
</tr>
<tr>
<td>F</td>
<td>-99.7339</td>
<td>-0.32453</td>
<td>0.33</td>
</tr>
<tr>
<td>Ne</td>
<td>-128.9376</td>
<td>-0.39047</td>
<td>0.30</td>
</tr>
</tbody>
</table>

Table 7.3: Gaussian basis sets used in the He and Ne calculations, respectively\textsuperscript{104}

<table>
<thead>
<tr>
<th>Basis set</th>
<th>Number of basis functions</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>SV\textsuperscript{a}</td>
<td>4</td>
<td>19</td>
</tr>
<tr>
<td>SVP\textsuperscript{b}</td>
<td>7</td>
<td>25</td>
</tr>
<tr>
<td>TZP\textsuperscript{c}</td>
<td>8</td>
<td>28</td>
</tr>
<tr>
<td>TZVPP\textsuperscript{d}</td>
<td>17</td>
<td>41</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Uncontracted split valence basis sets.
\textsuperscript{b} Uncontracted split valence basis sets augmented with one polarization function.
\textsuperscript{c} Triple zeta valence basis sets augmented with one polarization function.
\textsuperscript{d} Triple zeta valence basis sets double polarization functions.

is to be noted that the basis sets employed are optimized for atoms with fermionic electrons, and may therefore not be ideal for the systems studied here.\textsuperscript{101}

Figure 7.1 (b) shows the ground state energy as a function of the CI excitation level for Li and Be. In both cases it is clear that almost all of the correlation energy is taken into account at the CISD level; only 1% of the correlation energy is contained within the higher excitations.

Some results on the performance of the iterative direct CI technique described in Sec. 3.2 are shown in Fig. 7.2. FCI calculations were carried out on two different systems and the number of configurations was increased in two different ways: first by keeping the number of particles constant and increasing the basis-set size and then by keeping the basis set constant and increasing the number of particles. In the latter case the two-body interaction was scaled by the number of particles in order to enforce system stability. The scaling is not strictly linear in either case, a fact which can be anticipated from the form of the matrix-vector multiplication operation in Eq. 3.12: an increase in the number of particles or in the number of single-particle states leads to additional computations in the innermost loops over the basis functions, even when ‘Slater-Condon’ rules are used for selecting the contributions. Linear regression fits yield a leading power for the number of configurations of 1.96 for (a) and 1.25 for (b).

*For He the bosonic and fermionic systems coincide, which is reflected by the observed saturation at the TZVPP level
7.2 BECs

The direct CI program for bosonic systems has been developed for studies of Bose-Einstein condensates. The model underlying the implementation is described in Sections 5.1 and 5.2. The single-particle basis sets employed in the BEC calculations consist of anisotropic Cartesian Gaussian functions of the form

\[ \chi_i(r) = x^{l_i}y^{m_z}z^{n_z}e^{-\alpha_{r,i}x^2}e^{-\alpha_{y,i}y^2}e^{-\alpha_{z,i}z^2}, \]  

(7.1)

which is a natural choice due to the shape of the confinement potential. The anisotropic confinement potentials considered here are of the form \( \omega_z > \omega_x = \omega_y \equiv \omega_r \), where \( r \) denotes the \( xy \)-plane (see Eq. (5.14)). The degree of anisotropy is described by the parameter \( \lambda \equiv \omega_z/\omega_r \). The basis sets used for calculations with anisotropic potentials are chosen to be of a restricted tensorial form, such that for every exponent \( \alpha_{r,i} \) there are three scaled exponents in the \( z \)-direction: \( \alpha_{z,i-1}, \alpha_{z,i}, \) and \( \alpha_{z,i+1} \), giving a basis three times the size of the corresponding isotropic one. The reason for this is that simply scaling the exponents, i.e., \( \alpha_{z,i} = \lambda \alpha_{r,i} \) does not result in a satisfactory basis set.

One aim of this work was to investigate the interplay between particle density

**Figure 7.3:** Normalized radial densities for a trap with anisotropy \( \lambda = \sqrt{8} \), calculated at the CISD level for (from top to bottom) \( N = 100, 200, 500, 1000, 2000 \) and 5000.

Table 7.4: Calculated energies for a BEC with anisotropy \( \lambda = \sqrt{8} \). \( N \) is the number of particles, \( \epsilon_0 \) the lowest HF orbital energy and \( E_{HF}/N \), \( E_{CISD}/N \) and \( E_{CISDT}/N \) are the total HF, CISD and CISDT energies per particle, respectively.

<table>
<thead>
<tr>
<th>( N )</th>
<th>( \epsilon_0 )</th>
<th>( E_{HF}/N )</th>
<th>( E_{CISD}/N )</th>
<th>( E_{CISDT}/N )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.41421</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>100</td>
<td>2.87096</td>
<td>2.65819</td>
<td>2.65647</td>
<td>2.65647</td>
</tr>
<tr>
<td>200</td>
<td>3.20716</td>
<td>2.85296</td>
<td>2.85043</td>
<td>2.85043</td>
</tr>
<tr>
<td>500</td>
<td>3.92964</td>
<td>3.29756</td>
<td>3.29411</td>
<td>3.29411</td>
</tr>
<tr>
<td>1000</td>
<td>4.76010</td>
<td>3.83371</td>
<td>3.82998</td>
<td>3.82998</td>
</tr>
<tr>
<td>2000</td>
<td>5.91778</td>
<td>4.60445</td>
<td>4.60085</td>
<td>4.60084</td>
</tr>
<tr>
<td>5000</td>
<td>8.12345</td>
<td>6.11003</td>
<td>6.10700</td>
<td>6.10699</td>
</tr>
</tbody>
</table>
and correlation effects. It was found, however, that the self consistent Hartree-Fock procedure, which yields the reference state used in the CI scheme, diverges as the density parameter $N|a|$ becomes sufficiently large, despite the use of the convergence-improving DIIS algorithm. It is possible to go beyond this limit by performing a series of HF calculations incrementally increasing the value of the density parameter and using the converged HF solution for a given value of $N|a|$ as the initial guess for a calculation with a larger density parameter. Even this procedure will eventually encounter a limit beyond which the HF program does not converge. The reason for this is not fully understood. For example, near-degeneracy, manifested by a narrowing gap between the highest occupied orbital and the lowest unoccupied orbital, does not occur. This computational bottleneck occurs only at the HF stage, and does not affect the solution of the CI equation as long as the excitation level is unaltered.

The calculated HF energies obtained for a system consisting of a variable number of $^{87}\text{Rb}$ atoms are listed in Table 7.4. The scattering length used in the calculations is $a = 100a_0^{(52)}$ and the trap anisotropy is chosen as $\lambda = \sqrt{8}$, in accordance with the JILA TOP traps. The radial trapping frequency is $\omega_r/2\pi = 76$ Hz, yielding a scattering length expressed in radial harmonic oscillator units of $a/a_r = 0.0043$, where $a_r = \sqrt{\hbar/m\omega_r}$. The results are in agreement with those obtained at the GP level by Dalfovo and Stringari (109) apart from a small difference due to the use of slightly different scattering lengths.

The results of the CI calculations carried out with the model parameters listed above are displayed in Table 7.4. For the anisotropic 4$s$9$d$ basis the CISDT space is of dimension 4656 while the CISDT space is spanned by 152096 configurations. The radial single-particle densities obtained at the CISD level are shown in Fig. 7.3; the HF densities closely follow the CI ones and are not displayed. As was the case for the atomic systems discussed in the previous section, restricting the variational space to that obtained by single and double excitations from the reference state accounts for nearly 100% of the correlation energy, as illustrated in Fig. 7.4, although the triple excitations become more important with increasing particle number. A useful measure of correlation effects is the relative correlation energy, defined as $E_{c,\text{rel}} = (E_{\text{HF}} - E_{\text{CI}})/E_{\text{CI}}$. Figure 7.5 shows the dependence of $E_{c,\text{rel}}$ on the density parameter, which is varied in two ways: varying the scattering length $a$ keeping $N$ fixed and

![Figure 7.4: Relative contribution from triple excitations $E_c(SDT)/E_c(SD)$ as a function of the number of particles for traps with anisotropies $\lambda = \sqrt{8}$ (dotted line), $\lambda = 1.9142$ (dashed line) and $\lambda = 1.0$ (solid line).]
reversely. In the former case a monotonic increase of $E_{C,\text{rel}}$ is observed, whereas fixing $a$ at the value 0.0043 and varying $N$ from 100 to 5000 yields a different behaviour: a maximum of the relative correlation energy occurs at $N = 500$, corresponding to a density parameter of $N|a| = 2.15$.

The effect of the trap anisotropy on the correlation energy was also studied using a trap geometry with $\lambda = 1.001$ as an isotropic reference\footnote{This is due to the restricted tensorial basis sets employed in order to provide flexibility to account for correlation.}. The results for trap geometries with $\lambda$ ranging from 1.001 to $2\sqrt{3}$ and $N = 500, 1000$ and 2000 are displayed in Fig. 7.6. For the particle numbers studied, $E_{C,\text{rel}}$, perhaps surprisingly, decreases with increasing anisotropy. This result, along with the maximum in relative correlation energy seen in Fig. 7.5, are not intuitive.

It has been argued that a $\delta$-function interaction potential should not be used in CI calculations\cite{109,110}. Esry and Greene have found a linear decrease in the total energy of the condensate as a function of the largest angular momentum value included in the basis set\cite{109}, reflecting an anticipated divergence of the energy. In order to investigate whether the observed behaviour is indeed due to the form of the interaction potential we introduce a tunable interaction potential in the form of a Gaussian $s$-function. As seen from Eq. (5.18) this form reduces to a $\delta$-function in the limit of an infinite exponent, $\theta \to \infty$. Using two basis sets (14$s$3$d$ and 14$s$10$p$3$d$2$f$) the increase in correlation energy in going from the smaller basis set to the larger one was investigated. Figure 7.7 shows the difference in correlation energy $\Delta E_c \equiv E_c(\text{large}) - E_c(\text{small})$ as a function of the interaction for three different particle numbers. As expected, for an interaction deviating sufficiently from the $\delta$-function $\Delta E_c$ is smaller since the interaction itself is weaker, whereas when the interaction approaches the $\delta$-function comparison is appropriate, as illustrated by the nearly identical HF-energies listed in Table 7.5. In going from a Gaussian $s$-type interaction to the $\delta$-function pseudopotential no significant increase in $\Delta E_c$ was found. In a basis set study using the Gaussian interaction potential a very slow convergence of the correlation energy as a function of the largest angular momentum number included in the basis was observed, i.e. the same qualitative behaviour as for the $\delta$-function. The reason for the slow convergence is not fully understood, but a possible cause may be found in the abrupt change in the wave function in the vicinity of the collision point between two bosons. These local perturbations in the wave function due to short-range dynamical correlation effects are extremely difficult to describe using one-particle expansions.

Table 7.5: Effect of interaction for $N = 2000$. Tabulated values for the Hartree-Fock energy per particle and the difference in relative correlation energy calculated with two basis sets: 14$s$3$d$ (S) and 14$s$10$p$3$d$2$f$ (L).

<table>
<thead>
<tr>
<th>Interaction</th>
<th>$E_{HF}/N$</th>
<th>$E_{C,\text{rel}}^{L} - E_{C,\text{rel}}^{S}$ (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^2$</td>
<td>2.931891</td>
<td>483</td>
</tr>
<tr>
<td>$5 \cdot 10^2$</td>
<td>2.933857</td>
<td>516</td>
</tr>
<tr>
<td>$10^3$</td>
<td>2.934103</td>
<td>520</td>
</tr>
<tr>
<td>$10^4$</td>
<td>2.934324</td>
<td>524</td>
</tr>
<tr>
<td>$10^5$</td>
<td>2.934347</td>
<td>524</td>
</tr>
<tr>
<td>$10^6$</td>
<td>2.934349</td>
<td>524</td>
</tr>
<tr>
<td>$\infty$</td>
<td>2.934350</td>
<td>525</td>
</tr>
</tbody>
</table>
7.3 Summary and discussion

The CI approach for bosonic systems has been used in other studies, albeit differing in the details. Hauget and Haugerud expand the single-particle functions in harmonic oscillator eigenfunctions and employ an energy cutoff to truncate the CI expansion. In the approach used by Esry, the HF equations are numerically solved using finite elements and the most important virtual orbitals are obtained by solving the linear SE for the optimized interaction potential of the mean-field solution. The Hamiltonian matrix, constructed in the basis obtained by performing single and double

Figure 7.5: Relative correlation energy as a function of the density parameter $Na_s$ at the CISD level. The dashed line corresponds to a fixed scattering length ($a_s$) of 0.0043 and a variable number of particles in the range of $N = 100 \ldots 5000$, whereas for the solid line the number of particles is 2000 and the scattering length varied from 2 to 25.

Figure 7.6: The relative correlation energy $E_{c,rel} = (E_{HF} - E_{CI})/E_{CI}$ as a function of trap anisotropy calculated at the CISD level for 500 (top curve), 1000 (middle) and 2000 (bottom) particles. The most isotropic geometry corresponds to $\lambda = 1.0001$. 
excitations from the HF orbital to the optimized virtual orbitals is then explicitly diagonalized. Streltsov et al. have implemented a few-level multiconfigurational HF model, which simultaneously optimizes the CI coefficients and the single-particle orbitals. In addition, Cederbaum et al. have reported calculations using a coupled cluster approach.

The functioning of our CI program was tested on fictitious atom-like systems in which the electrons are replaced by bosonic counterparts. The case of Helium, which is equivalent to ordinary Helium with electrons, shows that the program functions as intended since the numbers agree. Timings of the computations show that the direct CI algorithm scales nearly linearly as a function of the number of configurations when keeping the number of particles constant and increasing the size of the basis set and approximately quadratically when using a fixed basis set and increasing the number of particles.

In the BEC study the radial condensate densities obtained at the CISD level agree with corresponding ones found in the literature. CISD is found to account for nearly 100% of the correlation energy, as in the atomic case, although the relative contribution of triple excitations grows with increasing particle number. Two counterintuitive results are found: a maximum of the relative correlation energy as a function of the density parameter and a decrease in the relative correlation energy with increasing anisotropy.

An obvious question, given the behaviour of the importance of triple excitations as the particle number is increased, is whether some significant qualitative or quantitative correlation effects arise as the density is increased further. Unfortunately, the unsolved convergence problem encountered in the HF calculations has not allowed this to be investigated; the same holds for the interesting case of increasing the anisotropy until the system becomes effectively one-dimensional. Another computational phenomenon encountered in the study is the basis-set sensitivity, manifesting as rapid oscillations in the wave function.

The issue of the use of an interaction potential proportional to $\delta(r_1 - r_2)$ is also interesting. In our study no evidence of unsuitability was found: replacing the interaction with an adjustable Gaussian function and letting it smoothly approach the $\delta$-function did not reveal any dramatic effects. Nevertheless, as discussed by Huang,
a Hamiltonian with this pseudopotential should not be diagonalized exactly, since the exact eigenvalues are the same as those for a free-particle system\textsuperscript{110}. If this holds, then one would expect to find the correlation energy decreasing with, e.g., increasing basis set size, or increasing CI level, neither of which is observed in our calculations.

It is clear from our results, as well as from ones in the literature, that correlation does not play a pivotal role in dilute BEC systems in traps with a single potential minimum. Recent experimental development has, however, made it possible to construct systems in which correlations \textit{do} play a large role. The tuning of interatomic interactions through Feshbach resonances, the possibility of changing the dimensionality of the system with optical potentials, and generating a strong periodic potential through optical lattices opens up a strongly correlated regime\textsuperscript{113}. It is in this area that future applications of the boson CI software may be found.
References


References


