Quantum engineering with ultracold atoms

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Quantum Engineering with Ultracold Atoms

Rick van Bijnen
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PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de Technische Universiteit Eindhoven, op gezag van de rector magnificus, prof.dr.ir. C.J. van Duijn, voor een commissie aangewezen door het College voor Promoties in het openbaar te verdedigen op donderdag 20 juni 2013 om 16.00 uur

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Front cover: The figure represents the ground state configuration of 131 Rydberg atoms, confined to a spherical volume. The image itself is a direct camera capture of a 1mm × 1mm hologram created with 780 nm laser light, using techniques described in this thesis. The title text is written with ultracold ions, created by selectively ionising atoms inside a magneto optical trap with a shaped excitation laser. Each character is approximately 900µm × 900µm in size in the atomic cloud. The resulting ions are accelerated onto a detector, with each character corresponding to a single measurement. Smaller characters appear brighter, since all the excitation laser light is diffracted into a smaller area.

Back cover: Lattice of ions, spaced approximately 35µm apart, single measurement.

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Introduction

1.1 The quantum world

The world around us consists of atoms. A typical atom is around $10^{-10}$ m in size, i.e., one tenth of a millionth of a millimeter. To give an idea of this size: a grain of sand contains in the order of $10^{18}$ atoms. If we were to enlarge each atom in this single grain of sand to the size of a pingpong ball, the resulting volume of balls would cover an area the size of the Netherlands with a layer of pingpong balls of more than a hundred meters deep. This example serves to illustrate that the things we experience in our daily life are all the result of vast congregations of atoms interacting with other vast congregations of atoms.

We all have an intuition based on this macroscopic world. Nobody is surprised about what happens when two billiard balls collide, or when water is flowing from the tap, or when our grain of sand is blown away by the wind. However, if we look at what the individual atoms are doing in their daily life, we will find that they behave very differently. At the scale of single atoms, the laws of quantum mechanics take control. Light and matter become intimately connected, and both are found to exist in discrete, quantised states.

In the quantum world, we find new phenomena such as superposition and entanglement, and these are fundamental quantum effects without counterpart in our macroscopic world. Quantum mechanics is therefore often described as ‘spooky’ and ‘weird’, but the confusion only arises when we attempt to capture quantum effects in the language of our macroscopic world. Over the past century, a rigorous and exceptionally accurate formalism has been developed which has never deceived us. The 2012 Nobel prize has been awarded to S. Haroche and D. Wineland for pioneering studies that test the basis of quantum mechanics \[1\]. They probed the boundaries between the microscopic and macroscopic worlds, and provided experimental realisations of the thought experiments instrumental in developing the quantum theoretical framework.

The fact that deep down at the atomic scale things obey different laws than those we
are used to, is not only interesting from a purely philosophical point of view: the quantum world is brimming with possibilities for new technologies. To quote the legendary physicist Richard Feynman, in his famous 1959 talk "There’s plenty of room at the bottom" [2],

"When we get to the very, very small world we have a lot of new things that would happen that represent completely new opportunities for design. [...] we are working with different laws, and we can expect to do different things."

Indeed, since their discovery, the laws of quantum mechanics have brought us many new technologies. To name a few applications, semiconductors, lasers, atomic clocks, and NMR are all heavily dependent on quantum mechanical principles.

Currently, in the search for new applications a lot of research is directed towards realising a quantum computer, which can do calculations no other classical computer can. Among other envisioned applications are quantum cryptography, interferometric devices and spintronic circuits. Additionally, exotic new materials are investigated in which quantum properties of electrons and their spins govern their conductance, which may lead to superconductivity at temperatures which will hopefully some day be as high as room temperature.

These envisioned new technologies and materials typically rely on quantum effects appearing in interacting, many-body systems. However, as the systems under study grow in complexity, we might need some additions to the toolbox of the quantum physicist. Namely, our ability to simulate quantum many-body systems using conventional computers is only limited to a few tens of particles. The underlying problem is the scaling of the number of quantum states a system can assume, which is exponential in the number of constituents. For instance, to simulate the spins of $N$ electrons, we need to store and manipulate $2^N$ coefficients. For $N = 50$ this is already a number too large to handle for most computers. Even the famous law of Moore [3] governing our increase in computer power can not provide much relief: we can only add about 5 additional electrons to our simulation per decade. And even this slow progress has to come to an end soon: for $N = 300$ we would have to keep track of more states than there are atoms in the universe!

Obviously, a novel way of doing simulations of quantum systems is required. Once again, a revolutionary idea was proposed by Feynman. In his 1982 lecture "Simulating physics on computers" [4], he suggested that in order to simulate a quantum system efficiently, one should use another quantum system. Blurring the boundaries between theory and experiment, such a quantum simulator would consist of a system of particles, carefully engineered such that their interactions mimic those of the system under study. The values of physical constants can then be obtained by performing measurements on the quantum simulator.

The quantum computer mentioned earlier is taking the idea of quantum simulators a step further: it might be possible to use quantum many-body systems not only to simulate
complex materials, but also to perform general, arbitrary calculations. For a certain class of problems, with quantum simulations being a subset, quantum computers can provide exponential speedups in calculation time. The problems in question are typically in the form of a search for an optimum in an exponentially large dataset, with no alternative but to evaluate all possible entries. A quantum computer would be able to do such an evaluation in parallel, by exploiting the superposition principle \[5\]. Examples are integer factorisation, for which Peter Shor showed that it can be performed in polynomial time on a quantum computer \[6\], or Grover’s database search \[7\]. Ever since the discovery of these algorithms, quantum computers are the focus of an immense research effort.

The quantum computer and quantum simulator are only two examples of the many new technologies under development, and ongoing research will most certainly produce new ideas and opportunities. In any case, quantum many-body physics will likely play a role in many of these ideas. It is for this reason that scientists are interested in further exploring the laws of the quantum world, and learning how to quantum engineer the technology of the future.

1.2 Ultracold atoms

Perhaps the best way to study, engineer and control quantum many-particle systems is using ultracold atomic gases, the subject of this thesis. In this highly active field of research, atoms are cooled to temperatures in the micro- or nano-Kelvin regime, colder than anything in the known universe. Even the outer space background radiation at 3 K is millions of times hotter, hence the name \textit{ultracold}. When the atoms reach these temperatures, they reveal their quantum nature in ways directly observable to us. Moreover, ultracold atoms provide very clean and controllable quantum systems.

Firstly, ultracold atomic gases are \textit{clean} in the sense that they are typically very accurately described by simple theoretical models, starting from first principles, allowing effects of interest to be studied and understood in isolation from other complicating factors. Theory and experiment grow simultaneously as the complexity of the systems under study is increased \[8\]. This is to be contrasted with the research in real solids, where theory and experiment approach each other from two opposite ends on the complexity scale, with highly simplified models attempting to describe highly complex experimental situations.

Secondly, ultracold atomic gases are \textit{controllable} in the sense that experimentalists, using a combination of lasers and electromagnetic fields, can freely modify system parameters such as trapping geometries, dimensionality and even interaction strengths. Theorists can put their models to the test over wide parameter ranges, and exotic regimes can be probed. Again, this is to be contrasted with the situation in real solids where many parameters are immutable, and pre-determined by Mother Nature.

These two properties, coupled with good observability and the close agreement of
theory and experiment, make ultracold atoms very natural candidates for realising a quantum simulator. Indeed, experimentalists are already able to create extremely close physical realisations of some of the simplified, textbook, theoretical models of condensed matter.

The best example is perhaps given by optical lattices: these are artificial crystals made of light, filled with cold atoms emulating the behaviour of electrons in a solid [9–12]. A thousand times enlarged, the lattice provides a magnified version of the solid, free of defects and with controllable parameters such as dimensionality, lattice spacing and lattice depth, and the interactions between the particles [8]. State of the art quantum-gas microscopes allow for a close comparison with theory through the observation of individual atoms [13, 14].

Another example where theory and experiment go hand in hand is in the creation of Bose-Einstein condensates (BECs) [15–17], where bosonic particles all condense into a single quantum state, forming a giant matter wave. BECs provide a unique window on the quantum world, where for instance the phenomenon of superfluidity is clearly observed through the formation of vortices [18–20]. The first BEC experiments are widely regarded as the breakthrough moment for ultracold atomic gases, bringing together atomic physics and quantum optics with condensed matter [21].

Rapid advances followed those early experiments, for example in demonstrating that the interactions between particles could be controlled using Feshbach resonances [22–27]. Employing this technique in systems of ultracold fermionic atoms led to a realisation of the paradigmatic Bardeen-Cooper-Schrieffer (BCS) transition underlying superconductivity [28]. Here, the fermionic atoms form weakly-bound Cooper pairs which condense into a superfluid BEC. Using Feshbach resonances, experimentalists were able to explore a wide parameter regime and tune smoothly between a BCS superfluid and a BEC of molecules [29–31].

A large part of current research is focused on extending the quantum simulation toolbox, and engineering new forms of interaction between atoms. For instance, the dominant interactions between ultracold atoms have traditionally been short-range, s-wave contact interactions. That is, particles can only influence each other if they happen to localise at the same position. Taking a step further, Griesmaier et al. [32, 33] realised a chromium BEC with dipolar interactions between the atoms, endowing the system with true long-range interactions [34]. Here, every particle in the system influences every other particle, introducing interesting new physics and applications, such as new states of matter, quantum information processing and quantum simulation [35, 36].

Another recently added ingredient to the quantum simulation of charged particles is the Lorentz force. This force is notably absent when neutral atoms are used to simulate the behaviour of electrons in solid, making it a challenge to include the effect of magnetic fields on the ’electrons’. In 2009 Y.-J. Lin et al. [37] have overcome this challenge. By exploiting Berry’s geometric phase [38] they engineered a vector potential equivalent to that experienced by electrons in a magnetic field [39, 40]. These ’synthetic’ magnetic
fields open the door to explore physically interesting regimes, such as the Fractional Quantum Hall regime \[41\]. In the latter regime quasi-particles emerge that are neither fermions or bosons, but rather anyons \[42\].

A final promising avenue for quantum simulation and quantum information to be discussed here are Rydberg atoms \[43\]. These atoms have their valence electron excited to an orbital with very high principal quantum number \(n\). Such states are highly polarisable, making them sensitive to external fields and giving rise to extremely strong and highly tunable interactions \[44\]. Due to these interactions, Rydberg atoms can influence each other over distances of micrometers, which is simply enormous in the world of ultracold atoms. Moreover, these interactions can be switched on and off by exciting or de-exciting a Rydberg atom. These attractive features have lead to an explosion of theoretical and experimental interest in the past decade \[43, 45–47\], with applications such as, e.g., quantum gates \[48\], single photon sources \[49\], quantum simulators \[50\], nonlinear optics \[51\], and much more.

The above examples highlight only a small subset of the many directions of ultracold atom research. In the past two decades, remarkable progress has been made in trapping, cooling, and manipulating ultracold quantum systems. This is perhaps best illustrated by several Nobel prizes being awarded for ultracold atom research \[1, 52, 53\]. Using lasers and electromagnetic fields, researchers have taken the engineering of quantum many-particle systems to another level.

Ultracold atoms are now at the forefront of modern physics, where questions about the nature of quantum matter are investigated and new applications are developed. Some prominent applications are getting within reach, such as quantum simulators and quantum computers, which for instance could play a key role in unraveling the mechanisms behind High-\(T_c\) superconductivity \[54\]. Judging from the rate of progress of the past 10-20 years, the future of ultracold atoms is going to be very exciting!

1.3 This thesis

In this thesis we explore theoretically, but also experimentally (Chapter 4), aspects of several ultracold quantum systems, each of which representing a promising direction in the science of ultracold atomic gases. The thesis can roughly be divided into two parts. The first part details the creation of quantum many-body states of interacting Rydberg atoms, with applications in quantum simulation of lattice systems. The second part is concerned with understanding internal and external forces on Bose-Einstein condensates, relevant for quantum simulation of superfluid phenomena. In more detail, the topics in question are:

- **Rydberg Atoms and their Interactions**
  The strong interactions between Rydberg atoms give rise to a wealth of interesting physics. In Chapter 2 the precise form of these interactions is calculated, in the
van der Waals regime, for atoms occupying either of two possible Rydberg states, and in the presence of electric fields. These interacting Rydberg atoms are then combined with optical lattices to show how several elementary condensed matter models can be realised.

- *Adiabatic Formation of Rydberg Crystals with Chirped Laser Pulses*
  In Chapter 3 we continue on the topic of Rydberg atoms and theoretically investigate the creation of crystalline Rydberg states with carefully crafted laser pulses. We show how these crystals can be created in disordered clouds of atoms, and consider several experimentally relevant systems in a 1D geometry. Finally, we discuss the structures of these crystals as they would occur in higher dimensions.

- *Shaping Laser Light Fields with a Spatial Light Modulator*
  With increasing complexity of the experiments in the field of ultracold atoms comes an increasing demand for complex optical potentials. Liquid crystal-based Spatial Light Modulators (SLMs) can cater to these demands. In Chapter 4 we theoretically and experimentally investigate the creation of arbitrary optical potentials in an existing ultracold experiment with an SLM. The spatial control of the excitation laser is a first step towards experimentally realising the Rydberg systems discussed in Chapters 2 and 3.

- *Bose-Einstein Condensates with Long-Range Dipolar Interactions*
  In Chapter 5 we investigate the equilibrium shape of a BEC in the presence of long-range dipolar interactions. This is done in the so-called Thomas-Fermi limit, valid for large particle numbers. We extensively explore the parameter space for all possible interaction parameters and trap configurations. Additionally, we calculate the collective excitations of the system, using a semi-analytic approach making use of 19th century astrophysics.

- *Synthetic Magnetic Fields*
  The landmark experiment by Lin et al. successfully demonstrated the creation of synthetic magnetic fields, introducing a Lorentz-like force for neutral atoms. Evidence for their success was the appearance of vortices. In Chapter 6 we analytically establish the hydrodynamical behaviour of a BEC in a uniform synthetic magnetic field. We find that the onset of vortex nucleation, as observed by Lin et al., corresponds to a dynamical instability in the hydrodynamical solutions. Using knowledge from rotating systems, we reveal other routes to instability and anticipated vortex nucleation.
Introduction

Bibliography

[1] The Nobel Prize in Physics 2012 was awarded jointly to S. Haroche and D. J. Wineland "for ground-breaking experimental methods that enable measuring and manipulation of individual quantum systems". https://www.nobelprize.org/.


Chapter 1.

[53] The Nobel Prize in Physics 2001 was awarded jointly to E. A. Cornell, W. Ketterle and C. E. Wieman "for the achievement of Bose-Einstein condensation in dilute gases of alkali atoms, and for early fundamental studies of the properties of the condensates". http://www.nobelprize.org/.

Interacting Rydberg Atoms as Quantum Spin Models

Abstract - Rydberg atoms are atoms with one electron excited to an orbital with very high principal quantum number, giving rise to exaggerated properties such as high sensitivity to external fields and extremely strong and tunable interactions. After reviewing the relevant single particle properties of rubidium Rydberg atoms, we calculate the interaction strengths between atoms occupying either of two Rydberg S-states. Interactions in the van der Waals regime are considered, and in the presence of electric fields. When the principal quantum numbers of the two atoms differ by one, an interaction process can occur in which the two atoms swap states. It is shown how quantum simulations of several paradigmatic spin systems known from condensed matter physics can be realised, by arranging many interacting Rydberg atoms on a lattice. Besides a previously known quantum Ising model, novel Rydberg implementations are found of a spin-1 system and the famous Heisenberg XXZ model. External electric fields allow tuning through a large parameter regime. The inclusion of a second Rydberg level is essential; it provides a key ingredient in the form of the swapping interaction.

The work presented in this Chapter has been performed in collaboration with R. Nath and T. Pohl. Parts of Sec. 2.4 of this Chapter will be submitted for publication.
2.1 Introduction

Rydberg atoms are atoms in which one electron is excited to a state with very high principal quantum number $n$, such as $n = 20$, or higher. These high values of $n$ are associated with an extremely large orbital radius for the electron, scaling as $n^2$. Quite rapidly with increasing $n$, the electron will orbit at distances of thousands of Bohr radii from the core. When the electron is so far removed from its core, the atom as a whole acquires some extreme properties.

For instance, the polarisability of a Rydberg atom scales as $n^7$, making it very sensitive to external fields. This opens the door to various ways of controlling the properties of the atoms. But the most spectacular scaling law is perhaps achieved by the van der Waals interaction strength. Here, the polarisability and energy level spacings all conspire to produce a scaling of no less than $n^{11}$ for the associated $C_6$ interaction coefficients. Consequently, two Rydberg atoms can influence each other significantly over a distance of several micrometers, which is simply enormous in the realm of ultracold atoms. Finally, Rydberg atoms have extremely long lifetimes, scaling as $n^3$.

Although Rydberg atoms have been studied for many years [55], the rapid advances in the field of ultracold atoms have lead to a renewed surge of interest [56–59]. With modern techniques, the atoms can be cooled down to near absolute zero, to a regime where the interaction energy by far exceeds the kinetic energy. In this regime, the atoms stand practically still with only the internal states undergoing dynamics, and this regime has been aptly named the 'frozen gas' regime [60–62]. In addition to the inversion of energy scales, the interactions between Rydberg atoms are also highly tunable: using external fields, and / or by choosing particular values of $n$, the strength and character of the interactions can be manipulated with great precision. In what can be viewed as an extreme case of tunability, the interaction can even be switched on and off altogether, by either exciting or de-exciting an atom from the ground- to the Rydberg state.

One of the most striking consequences of the strong interactions is the so-called blockade effect. Here, out of two (or more) closely spaced atoms only one of them can be excited to the Rydberg level, while the strong interactions shift the multiply excited state out of resonance. The blockade effect has been subject of intense experimental investigation [63–69]. This interest was in great part triggered by two seminal papers, Refs. [70] and [71], where proposals were made to exploit the blockade for realising a CNOT gate [70] and storing and retrieving quantum information [71].

Further applications of the blockade effect and Rydberg atoms in general are now found in single-photon sources [69, 71, 72], nonlinear optics [73–75], quantum simulators [76–77], and quantum computers [56, 70, 71, 78]. Great progress has been made in recent times in manipulating and controlling Rydberg atoms, e.g., in observing Förster resonances in cold gases [60, 61, 79], spatially resolved resonant energy transfer [80], and ultracold chemistry, where long-range molecules are assembled from two Rydberg atoms [62, 81], or one Rydberg atom and a ground state atom [82–84].
In a many-body context, systems of interacting Rydberg atoms can be used for studying exotic quantum many body phases \[85\mathbf{88}\], and provide an ideal tool to implement strongly correlated quantum systems \[89\mathbf{93}\]. In particular, Rydberg atoms can provide a very close experimental realisation of interacting spin-1/2 systems \[89\mathbf{94}\mathbf{95}\]. So far, most of the studies involve only a single Rydberg state, typically an S-state. But, in this case, the quantum correlations between the atoms are absent once the laser coupling is switched off, and the system is characterized by classical Rydberg-crystals \[91\], and the phase diagram can be understood from classical statistical mechanics \[94\].

In this Chapter we show that non-classical ground states of a Rydberg lattice gas can be achieved even in the absence of an excitation laser, by including the possibility of excitation to a second Rydberg S-state. In this situation the system is described by a spin-1 Hamiltonian whose physics is known to be vastly different from that of spin 1/2 systems. In particular in 1D, integer and half-integer spins are known to differ fundamentally \[96\]. Moreover, we show how a near-perfect, and highly tunable realisation of the paradigmatic XXZ spin-1/2 model can be realised with our system. As such, Rydberg atoms arranged on a lattice can serve as highly flexible quantum simulators of quantum magnetism.

All results depend strongly on the interactions between Rydberg atoms, which in turn depend on the single particle properties of the Rydberg state. Therefore we will review those two topics first, before moving on to many-particle physics. The structure of this chapter therefore follows the old physics mantra: one, two, many.

- **One:** Rydberg states. In Section 2.2 of this chapter we review how the electronic state of rubidium Rydberg atoms can be calculated. Furthermore, the dipole moments are discussed, and their important role in determining the response of a Rydberg atom to external electric fields, and we discuss the optical excitation of Rydberg atoms.

- **Two:** Interactions. In Section 2.3 we discuss how two Rydberg atoms interact with each other. The mutual dipole-dipole interaction gives rise to van der Waals interactions, and we investigate its magnitude for interactions between two S states, possibly different, and how the interaction strength can be tuned using external fields.

- **Many:** Spin Hamiltonians. Finally, piecing all the ingredients together, we present the main results of this Chapter in Section 2.4. There, we show how a 1D chain of interacting Rydberg atoms can be mapped onto spin-1/2, and spin-1 systems, with highly tunable parameters, and we explore some of the resulting ground states.
2.2 One atom: single particle properties

2.2.1 Rydberg states

In this Section we will review some basic properties of Rydberg states, which will serve as ingredients to the calculations performed in the later Sections. In particular, we investigate the calculation of Rydberg states in rubidium.

Rubidium belongs to the group of alkaline atoms which have only a single valence electron. When the valence electron is in a Rydberg state, it is on average very far away from the nucleus and the inner electrons (we will refer to this combination as the ‘core’) and it will mostly experience a purely coulombic $1/r$ potential. Consequently, the rubidium Rydberg states are very similar to the states of the hydrogen atom.

The core electrons fill up the lowest 4 shells of the nuclear potential and remain almost unperturbed by the valence electron, which spends most of its time away from the core. Therefore, instead of having to solve the full many-body problem for the 37 electron wavefunction, we can already remove 36 bodies from the equation, and to good approximation write down the Hamiltonian for the single valence electron wavefunction $\psi(r, \theta, \phi)$:

$$\hat{H}_a = -\frac{1}{2} \nabla^2 + V_{\text{core}}(r) + \frac{\alpha^2}{r^3} \hat{L} \cdot \hat{S}, \quad (2.1)$$

where the first term is simply the kinetic energy, and $V_{\text{core}}(r)$ is the core potential that is experienced by the valence electron, due to interactions with the nucleus and the 36 inner electrons. The third term is the spin-orbit coupling, where $\alpha$ is the fine structure constant and $\hat{L}$ and $\hat{S}$ are the orbital and spin angular momentum operators of the valence electron. We use Hartree atomic units, where all length scales are in units of the Bohr radius $a_0 = 5.29 \cdot 10^{-11}$m, and the energies in units of the Hartree energy $E_H = m\alpha^2c^2 = 4.3597 \cdot 10^{-18}$J, with $m$ the electron mass, $\alpha$ the fine structure constant, and $c$ the speed of light.

Angular part

The core potential in Eq. (2.1) is spherically symmetric, decoupling the radial degree of freedom from the orbital and spin degrees of freedom. The angular part of the wavefunction is identical to that of hydrogen. The only point of attention here is the spin-orbit coupling term, $\propto \hat{L} \cdot \hat{S}$, which couples the angular and spin degrees of freedom. Due to this term, the eigenstates of the angular part of the Hamiltonian are no longer product states of the form

$$Y_l^m(\theta, \varphi)|s, m_s\rangle,$$  \quad (2.2)

in which the electron is in a definite angular momentum state, described by a spherical harmonic $Y_l^m(\theta, \varphi)$, and in a definite spin state, $|s, m_s\rangle$.

Instead, the angular eigenstates become superpositions of such product states, called
generalised spherical harmonics \[97\], or sometimes spin spherical harmonics \[98\],
\[ Y_{l,j,m}^{j}(\theta,\phi) = \sqrt{\frac{j + m}{2j}}|y^{+}\rangle + \sqrt{\frac{j - m}{2j}}|y^{-}\rangle \quad (j = l + \frac{1}{2}) \]  
(2.3)

\[ Y_{l,j,m}^{j}(\theta,\phi) = \sqrt{\frac{j + 1 + m}{2j + 2}}|y^{+}\rangle - \sqrt{\frac{j + 1 - m}{2j + 2}}|y^{-}\rangle \quad (j = l - \frac{1}{2}), \]  
(2.4)

with
\[ |y^{\pm}\rangle = Y_{l}^{m+\frac{1}{2}}(\theta,\phi)|\frac{1}{2}, \pm\frac{1}{2}\rangle. \]  
(2.5)

The orbital quantum number \( l \) remains a good quantum number, and it takes on integer values \( l = 0, \ldots, n - 1 \). The newly introduced quantum numbers \( j \) and \( m_{j} \) are associated with the total angular momentum operator \( \hat{J} = \hat{L} + \hat{S} \). The quantum number \( j \) is associated with the eigenvalue \( j(j+1) \) of \( \hat{J}^{2} \), and can consequently take on the values of \( l + \frac{1}{2} \) and \( l - \frac{1}{2} \) for \( l > 0 \), and \( j = +\frac{1}{2} \) for \( l = 0 \). The quantum number \( m_{j} \) is the projection of \( \hat{J} \) onto the z-axis, and takes on integer values ranging between \(-j\) and \(+j\).

In the remainder of this text, we will often use the spectroscopic nomenclature for designating the low-\( l \) orbitals, where \( l = 0, 1, 2, 3 \) corresponds to \( S, P, D, \) and \( F \), respectively. In particular, when the quantum number \( j \) and \( m_{j} \) are irrelevant, we will use the label \( nS, nP, \) etc., to indicate the \(|n, l = 0, \ldots\rangle, |n, l = 1, \ldots\rangle, \) etc., states. When only \( m_{j} \) is irrelevant, we will append the value of \( j \) as a subscript, e.g. writing \( nP_{3/2} \) for the \(|n, l = 1, j = 3/2, \ldots\rangle \) state.

**Radial part**

Continuing with the radial degree of freedom, we note that it is not coupled to either the angular or spin degree of freedom, such that the eigenstates of the total Hamiltonian \( \hat{H}_{a} \) of Eq. (2.1) can be written as
\[ \psi_{nljm}(r, \theta, \varphi) = R_{nlj}(r)Y_{l,j,m}(\theta, \varphi). \]  
(2.6)

The radial wavefunction \( R_{nlj}(r) \) can be obtained by working out the Schrödinger equation, using the above form of \( \psi_{nljm} \),
\[ \hat{H}_{a}\psi_{nljm}(r, \theta, \varphi) = E_{nlj}\psi_{nljm}(r, \theta, \varphi), \]  
(2.7)

which leads to the radial Schrödinger equation for \( R_{nlj} \):
\[ E_{nlj}R_{nlj} = \left\{ -\frac{1}{2} \left[ \frac{d^{2}}{dr^{2}} + \frac{2}{r} \frac{d}{dr} \right] + \frac{l(l + 1)}{2r^{2}} + V_{\text{core}}(r) \right\} R_{nlj}(r). \]  
(2.8)
The second term on the right hand side is the so-called centrifugal barrier, which becomes stronger for states with higher angular momentum \( l \). The fourth term follows from working out the spin-orbit term of \( \hat{H}_a \) in Eq. (2.1) for the generalised spherical harmonic of Eq. (2.3).

Compared to the Hydrogen case, there are two important differences to be noted in the radial Schrödinger equation. Firstly, the energies \( E_{nlj} \) are altered, and have to be determined empirically. Obtaining analytical expressions for the energies would involve solving the full many-body problem after all. Luckily, various high precision experiments have been performed which determined that the energies for the rubidium Rydberg states can be efficiently described by an altered Rydberg formula, very similar to that of hydrogen:

\[
E_{nlj} = -\frac{\text{Ry}^*}{n^2} = -\frac{\text{Ry}^*}{(n - \delta_{nlj})^2},
\]

(2.9)

where \( \text{Ry}^* = 0.4999673250 \) is the Rydberg constant for Rubidium (to be compared with \( \text{Ry} = 1/2 \) for Hydrogen). The quantity \( \delta_{nlj} \) is the so-called quantum defect, which acts as a correction to the principal quantum number, changing it from \( n \) to an effective value \( n^* \).

The quantum defects \( \delta_{nlj} \) are calculated with the Rydberg-Ritz formula [62]:

\[
\delta_{nlj} = \delta_0 + \frac{\delta_2}{(n - \delta_0)^2} + \frac{\delta_4}{(n - \delta_0)^4} + \frac{\delta_6}{(n - \delta_0)^6} + \ldots
\]

(2.10)

The parameters \( \delta_0, \delta_2, \ldots \) have been determined by experimental studies, and are listed for instance in Ref. [99]. The quantum defect depends not only on the principal quantum number \( n \), but also on the angular quantum numbers \( l \) and \( j \), with the most prominent dependence being that on \( l \). Ignoring the \( j \) dependence, then to first order we have

\[
\delta_0(l = 0) = 3.13, \quad \delta_0(l = 1) = 2.65, \quad \delta_0(l = 2) = 1.35 \quad \text{and} \quad \delta_0(l = 3) = 0.016, \quad \text{and} \quad \delta_0(l > 3) \approx 0.
\]

For \( l = 0 \), the centrifugal barrier of Eq. (2.8) is absent, allowing the valence electron to penetrate the core. Consequently, two effects take place. Firstly, the valence electron is exposed to more of the unshielded nuclear charge and becomes more attracted to the nucleus as compared to the Hydrogen case. Secondly, the valence electron polarises the core, attracting positive charges and repelling negative ones. Both effects result in a lowering of the energy, as reflected by the quantum defects for \( l = 0 \).

As \( l \) is increased, the centrifugal barrier becomes stronger and the electron spends less time in the core. Hence, the quantum defects decrease in magnitude as \( l \) is increased, and the wavefunction becomes more hydrogenic. For \( l > 3 \), the valence electron experiences only the \( 1/r \) part of the core potential and the defects become negligible. The corresponding high angular momentum states are identical to those of the Hydrogen atom.
To compute the actual wave functions of the $S, P, D,$ and $F$ states, we need to specify the core potential $V_{\text{core}}(r)$. We use a model potential as proposed by Marinescu et al. [100]:

$$V_{\text{core}}(r) = \frac{Z_l(r)}{r} - \frac{\alpha_c}{2r^4} \left[ 1 - e^{-\left(r/r_c\right)^6} \right],$$  \hspace{0.5cm} (2.11)

where the first term represents the potential due to the unshielded nuclear charge, $Z_l(r)$, to which the valence electron is exposed. The unshielded charge depends on the radial distance, and is modeled as

$$Z_l(r) = 1 + (z-1)e^{-a_1 r} - r(a_3 + a_4 r)e^{-a_2 r},$$  \hspace{0.5cm} (2.12)

where the parameters $a_1, ..a_4$, as well as $\alpha_c$ in Eq. (2.11), are determined from experiment [100].

The second term in Eq. (2.11) models the potential due to the core polarisability. As the valence electron sets up an electric field $\vec{E} \propto 1/r^2$, it induces a similarly proportional dipole moment $\mu_c \propto 1/r^2$ in the core. The interaction energy of a dipole with a field is $\vec{E} \cdot \mu_c$, leading to the $1/r^4$ dependence on the distance. The factor $\alpha_c = 9.0760$ is the core polarisability, and the quantity between square brackets serves simply to truncate unphysical short-range behaviour of the polarisation potential, with $r_c$ the truncation distance.

Inserting the core potential (2.11) into the radial Schrödinger equation we can, for given quantum numbers $n, l, j$, integrate the equation numerically. To this end, we employ the Numerov scheme [101], which is a fourth order integration scheme specialised for second-order differential equations in which the first-order term is absent. At very small $r$, the core potential is no longer accurate, leading to an unphysical divergence of the wavefunction. Therefore, the integration is performed radially inward, starting at $r = 2n(n+15)$ and ending a little bit before $r = 0$, at $r = \alpha_c^{1/3}$ [102]. We follow Bhatti et al. [103], and first transform $x = \sqrt{r}$, and $X = r^{3/4}R(r)$ and integrate $X(x)$. The resulting wavefunctions contain several nodes, which are spaced as $\sqrt{r}$, and the transformation ensures a near-constant number of gridpoints between each pair of nodes.

Fig. 2.1 shows the probability densities $|rR_{nlj}|^2$ of several Rydberg states. Panel (a) shows the $30S, 40S$, and $50S$ states, exhibiting the increase in spatial extent ($\propto n^2$) as the principal quantum number $n$ is increased. Panel (b) shows the same states, but with orbital angular momentum $l = 25$ ($j = l - 1/2$). The high centrifugal barrier for this value of $l$ is clearly seen, pushing the electron well away from the core region, justifying our earlier discussions on core penetration. The number of nodes in the wavefunction is $n - l - 1$, and panel (c) of Fig. 2.1 shows the case $l = n - 1$, where the wavefunctions contain only a single lump of probability density. For appropriate $j, m_j$, these states correspond to near-circular electron orbits [104].

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Figure 2.1: Radial probability densities $|rR_{nl}(r)|^2$ for numerically calculated Rydberg wavefunctions for $n = 30, 40, \text{ and } 50$. (a): $S$ states with $l = 0$, (b): states with a high value of $l$, exhibiting the effects of the centrifugal barrier which pushes the valence electron away from the core, and (c): $l = n - 1$ states, for which the wavefunction has no nodes anymore.

2.2.2 Dipole moments

Many important properties of Rydberg atoms follow from the matrix elements of the dipole moment operator

$$\hat{\mu} = -e\hat{\mathbf{r}},$$

(2.13)

where $\hat{\mathbf{r}}$ is the position operator that measures the displacement of the valence electron from the nucleus, and $e$ is the elementary charge, with $e = 1$ in atomic units. The transitions between states induced with laser light, the response to electric fields, and the van der Waals interaction between Rydberg atoms are all calculations that depend heavily on the dipole moments (2.13), and the corresponding matrix elements:

$$\langle n, l, j, m_j | \hat{\mathbf{r}} | n', l', j', m_{j}' \rangle.$$  

(2.14)

The separability of the wave function carries over to the dipole matrix elements, as they are products $R_{nlj}^{n'l'j'} A_{ljm_j}$ of a radial matrix element.
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\[ R_{nlj}^{n'l'j'} = \int_0^{\infty} R_{nlj}(r)r R_{n'l'j'}(r)r^2 dr, \]  

(2.15)

and a matrix element for the angular + spin component

\[ A_{ljm_j}^{l'j'm'_j} = \langle ljm_j | (\hat{e}_x + \hat{e}_y + \hat{e}_z) | l'j'm'_j \rangle, \]  

(2.16)

where \( \hat{e}_x, \hat{e}_y, \hat{e}_z \) are the operators that measure the component of the state \( |l'j'm'_j\rangle \) along the unit vector in the \( x, y, \) and \( z \) directions, respectively. It should be noted that \( A_{ljm_j}^{l'j'm'_j} \) is therefore a three-component vector. The radial matrix elements should in principle be evaluated numerically, as we only know the wave functions numerically.

The components of the angular + spin dipole matrix element \( A_{ljm_j}^{l'j'm'_j} \) are best evaluated in the basis

\[ e_1 = \frac{e_x + i e_y}{\sqrt{2}}, \quad e_2 = \frac{e_x - i e_y}{\sqrt{2}}, \quad e_3 = e_z, \]  

(2.17)

corresponding to the components of \( \sigma^+, \sigma^- \) and linearly polarized light \[105\]. In this basis we have \[106\] \[107\]

\[ e_i \cdot A_{ljm_j}^{l'j'm'_j} = (-1)^{j - m_j + s + j' + 1} \sqrt{(2j + 1)(2j' + 1)(2l + 1)(2l' + 1)} \]  

(2.18)

\[ \times \left\{ \begin{array}{cc} j & 1 \\ l' & s \\ 0 & 0 \end{array} \right\} \left( \begin{array}{cc} j & 1 \\ -m_j & \mu_i \end{array} \right) \left( \begin{array}{cccc} l & 1 \\ 1 & l' \end{array} \right), \]  

(2.19)

where \( s = 1/2 \) for the valence electron, and \( \mu_1 = 1, \mu_2 = -1, \mu_3 = 0 \). The quantity between curly braces represents a Wigner-6j symbol, and the quantity between brackets denotes a Wigner-3j symbol \[106\].

The Wigner symbols are often equal to zero, leading to the well-known selection rules for dipole transitions. That is, the dipole moment operator cannot make transitions between states \( |n, l, j, m_j\rangle \) and \( |n', l', j', m'_j\rangle \) unless all of the following conditions are satisfied:

- \( l' = l \pm 1 \),
- \( m_j - \mu_i = m'_j \)
- \( |j - j'| \leq 1 \)

2.2.3 Response to electric fields

As a first application of the dipole matrix elements we compute the response of Rydberg atoms to an external electric field, denoted by \( \mathbf{F} \). The field couples to the electron dipole moment as \(-\mathbf{F} \cdot \mathbf{\mu}\), such that the Hamiltonian for the atom becomes
\[\hat{H} = \hat{H}_0 + \epsilon \mathbf{F} \cdot \mathbf{r}.\]  

(2.20)

The electric field operator couples the states \(|n, l, j, m_j\rangle\), such that they are no longer eigenstates of the Hamiltonian. The new eigenstates will become superpositions of the old ones, and the atomic energy levels will shift. This shift is called a Stark-shift \cite{97}, and we can calculate the new eigenstates and corresponding Stark shifts explicitly by diagonalising the Hamiltonian (2.20).

Figure 2.2 shows the Stark shifts of the 30\(S_{1/2}\) states, and those states lying closest to it, as a function of electric field strength. Such a figure is commonly referred to as a Stark map. The electric field is oriented along the \(z\)-axis, such that it only couples states with the same value of \(m_j\) due to the selection rules. The other states, with \(|m_j| \neq 1/2\), are not shown. But within the manifold of \(m_j = 1/2\) all states are coupled together, such that all the crossings in the Stark map are in fact anti-crossings.

At zero field, the energy shifts due to the quantum defects are visible, showing how the degeneracy of the states with low orbital angular momentum \(l \leq 3\) is lifted. These states respond quadratically to weak fields in the perturbative regime. This is understood from the selection rule \(\Delta l = \pm 1\), which couples them only to states with a different energy, and also causes the perturbation to vanish in first order. Thus, the perturbative energy shift \(\delta E\) of the Rydberg atoms in the electric field is

\[\delta E = \frac{1}{2} \alpha_0 F^2,\]  

(2.21)

with the polarisability \(\alpha_0\) given by

\[\alpha_0 = 2 \sum_{n', l', j', m_j' \neq n, l, j} \frac{|\langle n, l, j, m_j | \hat{\mu} | n', l', j', m_j' \rangle|^2}{E_{n, l, j} - E_{n', l', j'}}.\]  

(2.22)

Using that the the dipole moment between \(nS\) and \((n + 1)P\) states scales as \((n^*)^2\), and the level spacing scales as \((n^*)^{-3}\), we see that the polarisabilities should scale as \((n^*)^7\), as confirmed by experiment in Ref. \cite{108}. The new eigenstates \(|\tilde{\psi}\rangle\) in the presence of the field are called Stark states, and to first order in the perturbation we have

\[|\tilde{\psi}\rangle = |nljm_j\rangle + F \sum_{n', l', j', m_j' \neq n, l, j} \frac{\langle nljm_j | \hat{\mu} | n'l'j'm_j' \rangle}{E_{nlj} - E_{n'l'j'}} |n'l'j'm_j'\rangle,\]  

(2.23)

where it should be noted that the only nonzero terms in the sum are those abiding the selection rules of the dipole operator, i.e. with \(l' = l \pm 1\). An important observation from Eq. (2.23) is that the perturbed eigenstates acquire a permanent dipole moment in the presence of the electric field, namely

\[\langle \tilde{\psi}|\hat{\mu}|\tilde{\psi}\rangle = -\alpha_0 F \hat{e}_z.\]  

(2.24)
Figure 2.2: Stark map showing the electric field dependence of the energies of Rydberg states, for the $30S$ state and nearby states, and with $|m_j| = \frac{1}{2}$. All states up to $l = 20$ are included.

Figure 2.3: Zoomed-in Stark map for the (a) $30S$, (b) $30P$, (c) $30D$, and (d) $30F$ states. All $S, P, D$, and $F$ states behave quadratically initially. The $D$ states have an avoided crossing with the $29F$ state, reversing the direction of the Stark shift.
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In contrast to the $S, P, D,$ and $F$ states, the high-$l$ states are all degenerate, and in this case there exists no such thing as a weak field and a perturbative regime: regardless of the magnitude of the perturbation, it is always much (infinitely) larger than the energy differences between coupled states. As a result, the energies of the $l > 3$ states are shifted linearly as a function of field strength, producing a manifold of states, all fanning out from a single point. This manifold of states, with $l > 3$, is commonly referred to as the hydrogenic manifold.

2.2.4 Laser excitation

In this section we discuss the optical excitation of a ground state atom to a Rydberg level using laser light. The most important contribution to the interaction $\hat{H}_{AL}$ of an atom with monochromatic laser light is the interaction of the atomic dipole moment $\hat{\mu}$ with the electric field amplitude:

$$\hat{H}_{AL} = -\hat{\mu} \cdot \mathbf{E} \cos(\omega t),$$  \hspace{1cm} (2.25)

with $\mathbf{E}$ the electric field amplitude of the laser light field with frequency $\omega$. In writing Eq. (2.25), we assume that the spatial variation of the electric field over the extent of the atom is negligible. This approximation is called the electric dipole approximation \footnote{109}. In addition, the interaction with the magnetic field is neglected, being several orders of magnitude weaker than the electric field component.

According to the selection rules discussed in the previous Sections, the operator $\hat{H}_{AL}$ couples only states with different angular momentum $l$. Consequently, this means that there are two lasers needed in order to excite a Rydberg $nS$ state: one laser excites the ground state ($5S$) to a $P$ state, and the second laser excites from the $P$ state to the $nS$ level. In practice, the most commonly used intermediate $P$ level is the $5P_{3/2}$ state \footnote{59, 62, 107}. The associated laser wavelength is then 780 nm for the $5S_{1/2} - 5P_{3/2}$ transition, which is near-infrared. The $5P_{3/2} - nS$ transition to the Rydberg state has an associated wavelength of 480 nm, which is a blue laser. Figure 2.4(a) schematically shows this excitation scheme.

As illustrated in Fig. 2.4(a), the excitation lasers can be detuned from their respective transitions, with $\delta_e$ the detuning of the lower (red) transition, and $\delta_r$ the total, two-photon detuning from the Rydberg level. Denoting $|g\rangle$ for the $5S_{1/2}$ ground-state, $|e\rangle$ for the $5P_{3/2}$ intermediate state, and $|r\rangle$ for the Rydberg state, the Hamiltonian of the atom in the three-level basis becomes, after making the well-known rotating wave approximation, and transforming to a rotating frame \footnote{110},

$$H_{AL} = \begin{pmatrix} 0 & \Omega_{ge}/2 & 0 \\ \Omega_{ge}/2 & -\delta & \Omega_{er}/2 \\ 0 & \Omega_{er}/2 & -\Delta \end{pmatrix},$$  \hspace{1cm} (2.26)
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Figure 2.4: (a) Rydberg excitation scheme, employing a near-infrared 780 nm laser to excite the \(|5S_{1/2}\rangle\) state to the \(|5P_{3/2}\rangle\) state, and a blue 480 nm laser to excite from the \(|5P_{3/2}\rangle\) state to the (Rydberg) \(|nS_{1/2}\rangle\) state. The detuning of the 780 nm laser is denoted by \(\delta_e\), and the total, two-photon detuning from the Rydberg level is denoted by \(\delta_r\). (b) For large \(\delta_e\), the original three-level system can be reduced to an effective two-level system, with effective coupling \(\Omega\) and detuning \(\Delta\), by adiabatically eliminating the intermediate level.

where we have defined the Rabi frequencies

\[
\Omega_{ge} = \langle g | \hat{\mu} \cdot \mathbf{E} | e \rangle, \quad \Omega_{er} = \langle e | \hat{\mu} \cdot \mathbf{E} | r \rangle.
\] (2.27)

Refs. \[107\] and \[59\] discuss these coupling strengths and their magnitudes in more detail.

The intermediate \(5P_{3/2}\) state has only a very short lifetime of 26ns \[59\], due to a high rate of spontaneous emission. Therefore, to avoid decoherence during excitation, the detuning \(\delta_e\) of the lower transition is typically chosen to be very large such that the \(5P_{3/2}\) level does not get significantly populated during the excitation process \[59\]. The \(5P_{3/2}\) state can then be removed from the physical description of the system by a process called \textit{adiabatic elimination}, leading to an effective two-level system with Hamiltonian

\[
\hat{H} = \begin{pmatrix} 0 & \Omega/2 \\ \Omega/2 & -\Delta \end{pmatrix}.
\] (2.28)

The effective coupling strengths between the ground state \(|g\rangle\) and the Rydberg state \(|r\rangle\) is given by \[111\]
\[ \Omega = \frac{\Omega_{ge} \Omega_{er}}{2\delta_e}, \quad (2.29) \]

and the effective detuning \( \Delta \) of the Rydberg level given by \[111\] \[112\]:

\[ \Delta = \delta_r + \frac{\Omega_{ge}^2}{4\delta_e} - \frac{\Omega_{er}^2}{4\delta_e}. \quad (2.30) \]

Figure 2.4(b) shows the effective two-level scheme.

In the remainder of this chapter we will also consider the case where a second Rydberg level is coupled. This can be done in several ways, as shown in Fig. 2.5. The first option, depicted in Fig. 2.5(a), is to couple directly from the ground state, employing a different intermediate state. For instance, the \( 5S - 6P - nS \) pathway would be a viable excitation scheme \[59\]. Alternatively, it is possible to use the same intermediate \( 5P \) state, and only add an additional blue 480nm laser to the experimental setup, as shown in Fig. 2.5(b). Finally, it is possible to directly couple the two Rydberg levels by using microwave radiation \[113\], as shown in Fig. 2.5(c).

We switch notation and denote the ground state by \( |0\rangle \), the first Rydberg level by \( |1\rangle \), and the second Rydberg level by \( |2\rangle \). The far-detuned intermediate \( P \) levels can again be adiabatically eliminated, after which we can write down the general Hamiltonian describing the above excitation schemes:

\[ \hat{H} = \begin{pmatrix} 0 & \Omega_{01}/2 & \Omega_{02}/2 \\ \Omega_{01}/2 & -\Delta_1 & \Omega_{12}/2 \\ \Omega_{02}/2 & \Omega_{12}/2 & -\Delta_2 \end{pmatrix}, \quad (2.31) \]

with \( \Omega_{ij} \) the coupling between levels \( i \) and \( j \), and likewise \( \Delta_i \) the detuning from level \( i \). This Hamiltonian describes an effective triangle scheme as shown in Fig. 2.5(d). The couplings and detunings can be straightforwardly evaluated using Eqs. (2.29) and (2.30) for each of the transitions (mutatis mutandis). In the case of two distinct intermediate states, \( \Omega_{12} \) is zero, leading to a \( V \)-scheme. For the microwave scheme, \( \Omega_{01} \) is zero, leading to a ladder scheme. For the scheme with two blue lasers, all couplings are nonzero and we retain the full triangle scheme.

### 2.3 Two atoms: interactions

Having reviewed some of the key single-particle properties of Rydberg atoms, we are now in a position to investigate the interactions between them. The interactions are typically of the van der Waals type, scaling as \( C_6/R^6 \), with \( R \) the interatomic distance. The coefficients \( C_6 \) have been studied extensively in literature, either through numerical diagonalisation or perturbatively, see e.g. Refs. \[114\] \[116\]. Here, we employ a slightly different calculation strategy, using a perturbative effective Hamiltonian \[105\]. This
2.3.1 Dipole-dipole interactions

The primary interaction mechanism between atoms in the Rydberg state is through dipole-dipole interactions. For large atomic separations, the dipole-dipole interaction energy operator of two Rydberg atoms is of the form

$$\hat{V}_{dd}(\mathbf{R}) = \hat{\mu}_1 \cdot \hat{\mu}_2 R^3 - \frac{3 (\hat{\mu}_1 \cdot \mathbf{R}) (\hat{\mu}_2 \cdot \mathbf{R})}{R^5},$$

(2.32)

where $\mathbf{R}$ is the separation vector between the two atomic cores, and $\hat{\mu}_1$ and $\hat{\mu}_2$ are the dipole moment operators of atom 1 and 2, respectively.

Typically, the dynamics of the internal degrees of freedom of ultracold Rydberg atoms happen on timescales much shorter than their external degrees of freedom, i.e. the positions of the Rydberg atoms. Therefore, for computing the interaction energy we can treat the atomic cores as if they are fixed at their positions, with relative separation $\mathbf{R}$. The only remaining degrees of freedom are then those of the two valence electrons. We will assume the atomic separation to be large enough such that the atomic core of atom 1 is not perturbed by the valence electron of atom 2, and vice versa, and also large enough such that electron exchange does not play a role. The resulting Hamiltonian governing the dynamics of the two valence electrons then becomes

$$\hat{H} = \hat{H}_a \otimes \mathbf{i} + \mathbf{i} \otimes \hat{H}_a + \hat{V}_{dd}(\mathbf{R}),$$

(2.33)
where $\hat{H}_a$ is the single atom Hamiltonian of Eq. (2.1). The state of the two valence electrons can be described by (superpositions of) pair states of the form $|n,l,j,m_j\rangle \otimes |n',l',j',m'_{j}\rangle$, which are eigenstates of the first two terms in Eq. (2.33), but not of the dipole-dipole interaction term $\hat{V}_{dd}(R)$. Finding the new eigen-energies of the Hamiltonian (2.33) is the topic of the next few Sections.

### 2.3.2 A toy model: the van der Waals interaction

Our job is now to find the new eigenvalues of the Hamiltonian (2.33), the shifts in energy compared to the unperturbed case are exactly the interaction energies between two Rydberg atoms. Due to the multitude of states involved, this job is typically one to be performed numerically. However, it is instructive to first consider a toy model, with only two possible pair states in existence.

The first pair state in our toy model is the state $|nS\rangle \otimes |nS\rangle$, in which both atoms are in the $l = 0$ state with principal quantum number $n$. The second state is $|n'P\rangle \otimes |n''P\rangle$, and it is coupled to the first pair state by the dipole operator. The difference in unperturbed energies between the two pair states is $\Delta E$. The Hamiltonian for this system is, in this basis,

$$
\hat{H}_{2\times2} = \begin{bmatrix}
0 & V_0/R^3 \\
V_0/R^3 & \Delta E
\end{bmatrix},
$$

(2.34)

where we have explicitly indicated the $R^3$ dependence of the matrix elements of the interaction operator, defining $V_0 = R^3 \langle nS|\hat{V}_{dd}(R)|n'P\rangle$. The exact eigenvalues of the Hamiltonian are

$$
E_{\pm} = \frac{1}{2} \left( \Delta E \pm \sqrt{\Delta E^2 + 4V_0^2/R^6} \right).
$$

(2.35)

The new energies of the system depend on $R$, and we can distinguish two important regimes.

Firstly, for small distances $R$, when the off-diagonals are much larger in magnitude than the differences between the diagonal elements, i.e. $V_0/R^3 \gg \Delta E$, the energies become

$$
E_{\pm} \simeq \pm \frac{V_0}{R^3}.
$$

(2.36)

The interaction energy is that of two permanent dipoles, with its characteristic $R^{-3}$ dependence. The interaction is so strong that we cannot treat it as a small perturbation, and the initial unperturbed basis states become completely mixed. This regime is named the resonant dipolar regime [107].

The second regime occurs for large distances $R$, when the off-diagonal terms are now much smaller than the energy differences on the diagonal. In this case $V_0/R^3 \ll \Delta E$, and the energies become
The energies assume a $R^{-6}$ dependence on the distance, and defining $C_6 = V_0^2 / \Delta E$, we see that this is the well known van der Waals interaction energy. The $C_6$ coefficients are found to scale as $(n^*)^{11}$, which can easily be checked by noting that the dipole matrix elements scale as $(n^*)^2$, and hence $V_0$ scales as $(n^*)^4$, while the energy differences $\Delta E$ as $(n^*)^{-3}$.

Of course, we could have obtained the result of Eq. (2.37) also by directly applying quantum mechanical perturbation theory. The first order energy corrections vanish (i.e. $\langle nSnS | \hat{V}_{dd}(R) | nSnS \rangle = 0$, by the selection rules), the second order energy correction would return exactly $C_6 / R^6$. In this perturbative regime the new eigenstates still strongly resemble the initial basis states.

In fact, there exists a third regime where the interaction scales as $1/R^7$. This scaling only occurs at very large distances $R$, and is due to retardation effects that occur when one takes into account the quantization of the electromagnetic field [117], and the travel times of photons that are exchanged between the particles [57, 105, 109]. The transition to this regime occurs at distances comparable to the wavelength that is associated with the energy differences between the states involved in the interaction.

In this work, the dominant transitions involved in the interactions are between $|n, S\rangle$ and $|n \pm \Delta n, P\rangle$ states, with $\Delta n = -1, 0, 1$. For $n = 30$, the associated wavelengths are $\sim 10^3 \mu m$, which is typically much larger than the system size of interest. Additionally, to ensure full convergence in the calculations, transitions between states with $|\Delta n| \leq 6$ are included, which have associated wavelengths of about $10^2 \mu m$, and larger. Transitions between $P$ and $D$ states, or even higher angular momenta, have typically larger wavelengths associated with them. The same holds for higher values of $n$: for $n = 40, 50, 60$, and $\Delta n = \pm 6$, the wavelengths are $\sim 300, 700, and 1300 \mu m$, respectively.

As we will not be considering systems sizes larger than $100 \mu m$, the $1/R^7$ scaling regime is not accessed, and we can always assume to be in the perturbative van der Waals regime for large $R$, without having to quantize the electric field. On the other hand, the transition between the $R^{-3}$ and $R^{-6}$ regimes depends on the ratios of the coupling strengths and energy differences of all possible transitions between all possible internal atomic states. This transition is therefore nontrivial to calculate, and needs to be computed separately for each particular system under investigation.

### 2.3.3 The effective Hamiltonian

As we have seen in the previous Section, the $C_6$ coefficients for the van der Waals interaction can be obtained using perturbation theory. Indeed, several such studies have been carried out in the literature [56, 114, 116]. However, here we will also require off-diagonal matrix elements of the van der Waals interaction in the atomic basis, which are not easy to obtain using ‘standard’ perturbation theory [114]. Therefore, we will employ a more
sophisticated version of perturbation theory, that of the effective Hamiltonian [105].

Suppose we have an unperturbed Hamiltonian $\hat{H}_0$ with known eigenstates $|i\rangle$ and eigenvalues $|E_i\rangle$. We are interested in a certain subset $S$ of eigenstates $|s\rangle$, with $s \in S$, whose energies $E_s$ are well separated from all other energies $E_t$, with $t \in T = S^C$. Figure 2.6 schematically illustrates this situation.

![Figure 2.6: Energy levels $E$ showing a distinct partitioning into energetically separated subsets $S$ and $T$, which are weakly coupled by the operator $\lambda \hat{V}$.](image)

Now suppose the system we are interested in has a Hamiltonian of the form

$$\hat{H} = \hat{H}_0 + \lambda \hat{V},$$

(2.38)

with $\hat{V}$ a perturbing (nondiagonal) operator that couples the eigenstates $|i\rangle$. The parameter $\lambda$ is a scalar, assumed small in the sense that

$$|\langle s|\lambda \hat{V}|t\rangle| \ll |E_s - E_t|, \quad s \in S, \quad t \in T.$$  

(2.39)

That is, the states in our subset of interest $S$ are only weakly coupled to the states outside the subset. Note that we pose no restriction on the magnitude of the coupling within the subset $S$ due to the operator $|V\rangle$.

If the system initially occupies one of the states of the subset $S$, it will mostly stay in this subset. States in $T$ constitute fast degrees of freedom of our system: they are only briefly visited, and we have only very small probabilities of not finding the system in a state belonging to $S$. Therefore, in good approximation we can describe the system state using only states in $S$, and ‘forget’ the states in $T$. However, the states $|s\rangle$ can still be coupled among themselves through states in $T$, affecting their energy levels and dynamics.

Now, the effective Hamiltonian is a Hamiltonian governing the dynamics of the states in $S$, eliminating the states from the subspace $T$, while still correctly taking into account the effect of having the fast degrees of freedom present. The matrix elements of this effective Hamiltonian are, for states $s, s' \in S$, and to second order in $\lambda \hat{V}$ [105].
\begin{align}
\langle s | H_{\text{eff}} | s' \rangle &= E_s \delta_{ss'} + \langle s | \lambda \hat{V} | s' \rangle \\
&+ \frac{1}{2} \sum_{t \in T} \langle s | \lambda \hat{V} | t \rangle \langle t | \lambda \hat{V} | s' \rangle \left[ \frac{1}{E_s - E_t} + \frac{1}{E_{s'} - E_t} \right].
\end{align}

The first term contains simply the unperturbed energy levels of the states in $S$, the second term contains the direct couplings between them by the operator $\hat{V}$. The third term contains the indirect couplings, where the system visits an intermediate state belonging to $T$. The first two terms are exact, only the third term is the result of a perturbative expansion. Note that we can recognize the 'standard' perturbation theory result in the above expression, when the subspace $S$ consists only of a single state $|s\rangle$.

We have already encountered an example of an effective Hamiltonian in Sec. 2.2.4, where the intermediate $5P_{3/2}$ state was eliminated, reducing the three-level atom to an effective two-level system. Indeed, Eqs. (2.29) and (2.30) for the coupling strengths and level shifts follow immediately from applying Eq. (2.40), after making the identification $S = \{ |g\rangle, |r\rangle \}$ and $T = |e\rangle$.

With the second order effective Hamiltonian of Eq. (2.40), we can now efficiently compute various $C_6$ interaction coefficients, by substituting the dipole-dipole interaction operator $\hat{V}_{dd}(R)$ defined in Eq. (2.32) for $\lambda \hat{V}$. For simplicity we assume the separation vector $R$ to be oriented along the $z$-axis. The unperturbed Hamiltonian $\hat{H}_0$ is taken to be the sum $\hat{H}_a \otimes \hat{I} + \hat{I} \otimes \hat{H}_a$ of the single particle Hamiltonians $\hat{H}_a$ [see Eqs. (2.33), (2.1)].

The $C_6$ interaction coefficients for pair states $|ab\rangle$, with energy $E_{ab}$, are then given as the matrix elements of the operator $\hat{H}_{\text{eff}} - \hat{H}_0$:

$$C_{6ab} / R^6 = \langle ab | (\hat{H}_{\text{eff}} - \hat{H}_0) | ab \rangle = \langle ab | (\hat{H}_{\text{eff}} - E_{ab}) | ab \rangle.$$

Likewise, effective couplings induced by the interaction between pair states $|ab\rangle$ and $|cd\rangle$ can be computed as

$$C_{cdab} / R^6 = \langle ab | (\hat{H}_{\text{eff}} - \hat{H}_0) | cd \rangle = \langle ab | \hat{H}_{\text{eff}} | cd \rangle.$$

In the next 3 Sections we will explore several cases of interacting Rydberg $S$ states of interest.

2.3.4 $nS - nS$ interactions

We start by computing the interactions energies and couplings between $|nS\rangle \otimes |nS\rangle$ states, where both atoms are in the same $nS$-state. An atom in the $|nS\rangle$ state necessarily has $j = 1/2$, and thus can be either in the $m_j = +1/2$ or the $m_j = -1/2$ state. Using the shorthand notation $|\pm\rangle$ for denoting the $|nS,m_j = \pm1/2\rangle$ state, we have four possible pair states: $|++\rangle, |+-\rangle, |-+\rangle$, and $|--\rangle$ which form the subset $S$ of states of interest.
All four states have the same energy, and are not degenerate with any other states, such that we can apply the effective Hamiltonian methodology.

In the basis $S$ we thus obtain the following matrix elements for $\hat{H}_{\text{eff}} - \hat{H}_0$:

$$
\hat{H}_{\text{eff}} - \hat{H}_0 = \frac{1}{R^6} \begin{pmatrix}
C_{++}^+ & 0 & 0 & 0 \\
0 & C_{+-}^+ & C_{-+}^+ & 0 \\
0 & C_{++}^- & C_{-+}^- & 0 \\
0 & 0 & 0 & C_{+-}^-
\end{pmatrix},
$$

(2.43)

where we have used the notation of Eqs. (2.41) and (2.42). We can distinguish four $C_6$ coefficients, but because of symmetry considerations we have $C_{++}^+ = C_{--}^-$ and $C_{+-}^- = C_{-+}^-.$

We also find off-diagonal elements $C_{+-}^- = C_{-+}^+$, which are typically much smaller than the diagonal elements. These off-diagonal elements would cause a system starting in (e.g.) the $| -+ \rangle$ state to (slowly) oscillate between the $| -+ \rangle$ and $| + - \rangle$ state due to the dipole-dipole interaction, and thus cause the atoms to exchange their $m_j$ quantum numbers. We see that the states $| -+ \rangle$ and $| + - \rangle$ are thus actually not eigenstates of the Hamiltonian, these are the symmetric and antisymmetric combinations $(| -+ \rangle \pm | + - \rangle)/\sqrt{2}$, with associated eigenvalues $C_{++}^+ \pm C_{+-}^-.$

In total we thus find three possible interaction energies for the two atoms in $nS$ states, but the splitting is only very small. Figure 2.7(a) shows the interaction energy as a function of the principal quantum number $n$, plotted on a log-log scale. Clearly, the interaction strength grows extremely fast as a function of $n$, following the $n^{11}$ scaling derived in Sec. 2.3.2, and the $C_6$ coefficient traverses many orders of magnitude when we increase $n$ from 25 to 80. Figure 2.7(b) shows the same $C_6$ coefficients, but now with the $n^{11}$ scaling divided out. Incidentally, this altered scale also reveals the presence of the three distinct interaction curves, with the top curve corresponding to the interaction energy of the $(| - + \rangle + | - - \rangle)/\sqrt{2}$ state, the middle curve to the $| + + \rangle$ and $| - - \rangle$ states, and the bottom curve to the $(| - + \rangle - | - - \rangle)/\sqrt{2}$ state.

From Fig. 2.7 we can also see that $nS - nS$ interaction energies are always positive, meaning that Rydberg atoms in $S$ states repel each other. Experimentally, this is a convenient feature, as attractive interactions cause the atoms to collide and consequently ionise.

A final point of interest is the role of the quantisation axis. Here, we have chosen the atoms to be aligned along the $z$-axis. In this case, the dipole-dipole operator cannot change the sum of the $m_j$ quantum numbers, leading to zero-valued matrix elements between $| + + \rangle$ and the other states, and likewise for $| - - \rangle$. Should we have chosen a quantisation axis not aligned with $R$, then the dipole-dipole coupling can change the sum of $m_j$, and we would have found a different sparsity pattern for the effective Hamiltonian [as opposed to that of Eq. (2.43)], and different eigenstates, with different interaction
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Figure 2.7: \( C_6 \) coefficients for \( nS - nS \) interactions, plotted on (a) log-log scale, illustrating the \( n^{11} \) scaling, and (b) plotted with the \( n^{11} \) scaling divided out, revealing the presence of 3 distinct curves corresponding to the interactions between the various sublevels \( m_j = \pm \frac{1}{2} \).

energies. The interaction strength thus has an angular dependence, even for \( S \) states\(^{107, 116}\). However, the difference in interaction energies as a function of the orientation is only very small, on the order of 1 – 3\%. Therefore, in practice we can ignore any angular dependencies and the distinction between internal states, and suffice with simply assigning a single \( C_6 \) coefficient for \( nS \) states. For higher orbital angular momenta, as in \( P, D, F \) states, there is a significant angular dependency which cannot be ignored.

In the remainder of this work, we shall only be concerned with \( nS \) states, and will only report interaction coefficients between atoms in the \( m_j = +1/2 \) state, with the atomic separation vector \( \mathbf{R} \) aligned along the quantisation axis, \( \hat{e}_z \).

2.3.5 \( n_1S - n_2S \) interactions

As a second case of interest, we compute the interactions between an atom in the \( n_1S \) state, and another atom in the \( n_2S \) state, with two different principal quantum numbers \( n_1 \neq n_2 \). Using the shorthand notation \(|1\rangle\) to denote the \(|n_1S\rangle\) state, and \(|2\rangle\) for the \(|n_2S\rangle\) state, the manifold of pair states consists of the states \(|12\rangle\) and \(|21\rangle\). Again, the two pair states have the same energy and we can safely apply the effective Hamiltonian formalism.

In matrix form, the effective Hamiltonian becomes

\[
\hat{H}_{eff} - \hat{H}_0 = \frac{1}{R^6} \begin{pmatrix}
C_{12}^6 & C_{11}^S \\
C_{21}^S & C_{21}^S \\
\end{pmatrix}, \tag{2.44}
\]
Figure 2.8: Schematic illustration of the two relevant interaction processes in $n_1S - n_2S$ interactions. (a) The interaction associated with the $C_{6}^{12}$ coefficient, where the dipole-dipole operator first transfers the two particles to $P$ states, and then back to their original states. (b) The $C_{6}^{S}$ process, where the particles have swapped excitations after the interaction.

with only two distinct values for the matrix elements, namely

$$C_{6}^{12} = C_{6}^{21} = R^6 \langle 12 | (\hat{H}_{\text{eff}} - \hat{H}_0) | 12 \rangle,$$  \hspace{1cm} (2.45)

and

$$C_{6}^{S} / R^6 = \langle 12 | (\hat{H}_{\text{eff}} - \hat{H}_0) | 21 \rangle.$$  \hspace{1cm} (2.46)

Figure 2.8 depicts schematically the associated interaction processes. Fig. 2.8(a) is the interaction associated with the $C_{6}^{12}$ coefficient. The dipole-dipole coupling first excites the $|n_1S\rangle \otimes |n_2S\rangle$ state to a pair of $P$ states, and then back again returning the particles to their original state. Fig. 2.8(b) depicts the process associated with the $C_{6}^{S}$ coefficient. Here, the dipole-dipole interaction again first excites both particles to a $P$ state. However, the second step returns the first particle to the initial state of the second particle, and vice versa, effectively swapping their excitations.

A two particle system where we have excited one atom to some $n_1S$ Rydberg state, and the other atom to the $n_2S$ Rydberg state will therefore not stay in that state. As time progresses, the atoms start to oscillate between the $n_1S$ and $n_2S$ states. Again, the eigenstates of the two-particle system would be the symmetric and anti-symmetric combinations $(|12\rangle \pm |21\rangle)/\sqrt{2}$. At a later stage we will explore the consequences of the swapping process in a many-particle system and show that this can lead to exotic quantum phases in the ground state of a Rydberg gas with mixed excitations.

First, it is important to investigate the magnitude of the swapping interaction. Figure 2.9 shows the $C_{6}^{12}$ and $C_{6}^{S}$ coefficients as a function of $n_2$, while keeping $n_1$ fixed at $n_1 = 30$ in Fig. 2.9(a), and $n_1 = 65$ in 2.9(b). The coefficients are scaled to the $n_1S - n_1S$ interaction coefficient $C_{6}^{11}$. The $C_{6}^{12}$ coefficient is of similar magnitude as $C_{6}^{11}$ for $n_1$ close to $n_2$. For $n_2 > n_1$, the interaction becomes attractive, and the $C_{6}^{12}$ coefficient grows in magnitude for increasing $n_2$, due to increasing polarisability of the
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Figure 2.9: $C_6^{12}$ and $C_6^S$ coefficients, in units of $C_6^{11}$, for the $n_1S - n_2S$ interactions, as a function of $n_2$ with (a) $n_1 = 30$ and (b) $n_1 = 65$.

The $n_2S$ state. The sometimes jagged appearance of the $C_6^{12}$ curve stems from accidental near-resonances of the intermediate $P$ pair states taking part in the interaction process.

The $C_6^S$ coefficient associated with the swapping process is seen to be significant compared to $C_6^{12}$ only when $|n_1 - n_2| = 1$, for larger differences between $n_1$ and $n_2$ it quickly becomes orders of magnitude smaller. The swap for each individual particle can only be completed via an intermediate $P$ state, which cannot be close in $n$ to both $n_1$ and $n_2$ at the same time. The terms in the effective Hamiltonian (2.40) that contribute to the swapping process will therefore always involve small matrix elements of the dipole operator, due to the decreasing overlap of the radial wavefunctions.

To see how $C_6^{12}$ and $C_6^S$ progress as a function of $n_1$, we plot both coefficients for $n_1 - n_2 = 1$ in Fig. 2.10. An immediately apparent feature is the occurrence of two resonances: close to $n_1 = 25$ and $n_1 = 39$ the $C_6^{12}$ and $C_6^S$ coefficients diverge. At these values of $n_1$, there happens to be a pair of $P$-states with nearly the same energy as the $|n_1S⟩ ⊗ |n_2S⟩$ pair. The corresponding terms in the summation in the effective Hamiltonian of Eq. (2.40) blow up as a result, and they dominate the value of the matrix element.

The resonant pair states in question are the $|n_2P_{1/2}⟩ ⊗ |n_2P_{3/2}⟩$ state for the resonance near $n_1 = 25$, and $|n_2P_{3/2}⟩ ⊗ |n_2P_{3/2}⟩$ state for the resonance near $n_1 = 39$. Figure 2.11 shows the difference in energy $\Delta E$ between the $|n_1S⟩ ⊗ |n_2S⟩$ state and the $|n_2P⟩ ⊗ |n_2P⟩$ states. The energy differences are seen to approach zero and switch sign. Also, for $n_1 > 40$, the energy difference with the $|n_2P_{3/2}⟩ ⊗ |n_2P_{3/2}⟩$ state stays very close to zero ($\sim 0.3$ GHz), giving rise to a very broad resonance. Ref. [116] also noted this resonance, in the context of $nP - nP$ interactions.

As stated earlier in Sec. 2.3.2, for the perturbation theory to be valid, the dipole-dipole matrix elements must be much smaller than the energy differences $\Delta E$ between the
Figure 2.10: $C_{6}^{12}$ and $C_{6}^{S}$ coefficients for $n_{1}S - n_{2}S$ interactions, as a function of $n_{1}$ and with $n_{2} = n_{1} - 1$. There are two clear resonances, around $n_{1} = 25$ and $n_{1} = 39$, indicated by vertical dashed lines.

Figure 2.11: Energy defect $\Delta E$ of the $|n_{1}S\rangle \otimes |n_{2}S\rangle$ pair state with $|n_{2}P\rangle \otimes |n_{2}P\rangle$ pair states. Two of the curves approach and pass through zero, giving rise to the two resonances observed in Fig. 2.10.
states they connect. Therefore, close to a resonance the critical radius \( R_c \) for which we are in the van der Waals regime will diverge. The critical radius \( R_c = R_c^{(12)} \) determined by the \( n_1 S - n_2 S \) interactions is given by

\[
R_c^{(12)} = \max_{n',n''} \left\{ \frac{\langle n_1 S n_2 S | \hat{V}_{dd} | n' P n'' P \rangle}{|\Delta E(n_1, n_2, n', n'')|} \right\}^{1/3},
\]

where \( \Delta E(n_1, n_2, n', n'') \) is the energy difference between the \( |n_1 S \rangle \otimes |n_2 S \rangle \) and \( |n' P \rangle \otimes |n'' P \rangle \) pair states. Analogously, we can define a critical radius \( R_c^{(11)} \) for \( n_1 S - n_1 S \) interactions. The van der Waals regime is entered when \( R \gg R_c \). Actually, we can state this criterion a bit less strict in the form: \( R^3 \gg R_c^3 \). If the distances \( R \) are twice the distance \( R_c \), the matrix elements and state admixing amplitudes are already reduced by a factor of 1/8 (rather than 1/2). Figure 2.12 shows the relevant critical radii \( R_c \) encountered in the calculation of the \( C_6 \) coefficients for \( n_2 = n_1 - 1 \), as a function of \( n_1 \). At \( n_1 = 25 \) and \( n_1 = 39 \) we clearly see the divergences due to the two resonances. Using the criterion \( R > 2R_c \) we find for \( n = 30 \) that the van der Waals regime starts at around \( R > 1 \mu m \).
2.3.6 Interactions in the presence of an electric field

In the previous section it was shown that the energy differences between the \( S \) and \( P \) pair states can have a large influence on the \( C_6 \) interaction coefficients. Since electric fields influence the energy levels in turn, it is possible to tune the interaction strength between Rydberg atoms using external electric fields. A particularly well-known effect is the so-called Förster resonance, where the electric field shifts another pair state into resonance with the pair state of interest (e.g. \(|nS\rangle \otimes |nS\rangle\)), inducing strong dipolar \( 1/R^3 \) interactions \[62, 79\]. In this work, we will only employ weak electric fields to change the van der Waals interaction coefficients, and aim to stay away from resonances and the associated strong state mixing. As before, the electric field is in the direction of the quantization axis, as is the atomic separation vector.

To compute how the van der Waals interaction strengths are altered, one should in principle first calculate the new single particle eigenstates in the presence of the electric field by diagonalising the Hamiltonian (2.20). The new single particle states can then be used in the effective Hamiltonian to compute the interaction strengths perturbatively. However, for weak electric fields it turns out to be sufficient to only use the Stark-shifted energies, and ignore the corrections to the single particle eigenstates. The error made in this approximation can be estimated as \( \Delta E_{\text{pair}}/\Delta E_{\text{sp}} \), where \( \Delta E_{\text{pair}} \) is the energy difference between the nearest pair states participating in the interaction, and \( \Delta E_{\text{sp}} \) is the energy difference between the associated single particle states. For the results presented in this section, the error is in the order of \( 1−2\% \), which is comparable to the error made in neglecting the angular dependence of the interaction between \( S \) states.

For completeness, the results have been verified with the full numerical calculation that includes the corrections to the eigenstates, confirming the \( 1−2\% \) error estimate.

Fig. 2.13 shows the various \( C_6 \) coefficients for \( n_1 S − n_1 S \) and \( n_1 S − n_2 S \) interactions, for \( n_1 = 30 \), and \( n_2 = n_1 − 1 \). As a function of field strength, the \( C_{6}^{11} \) and \( C_{6}^{22} \) coefficients for the \( nS − nS \) interactions show a gentle decrease. The \( C_{6}^{12} \) and \( C_{6}^{S} \) coefficients show a more rapid variation, because of the presence of the near-resonant \(|n_2P\rangle \otimes |n_2P\rangle\) pair states. At \( F \approx 22.5\, \text{V/cm} \), the \(|n_2P_{3/2}\rangle \otimes |n_2P_{3/2}\rangle\) state is shifted into resonance and the coefficients diverge.

However, Fig. 2.13 is not the full story. As noted before in Sec. 2.2.3 in the presence of an electric field the Rydberg states acquire a permanent dipole moment, aligned along the \( z \)-axis and of magnitude \( \alpha_0 F \) [see Eq. (2.24)], leading to dipolar interactions between them of the form \( C_3/R^3 \). The \( C_3 \) interaction coefficient is given by the first order term of the effective Hamiltonian (2.40), which has become nonzero. For the field and atoms aligned along \( z \) it evaluates to \( C_3 = -2\alpha_1 \alpha_2 F^2 \), \( (2.48) \) where \( \alpha_1 \) and \( \alpha_2 \) are the polarisability of atoms 1 and 2, respectively, as given by Eq. (2.22). Since \( S \) states always have positive polarisability, the dipolar interaction is found to be attractive. Would the atoms have been aligned at an angle \( \theta \) to the \( z \)-axis instead,
Figure 2.13: $C_6$ coefficients as a function of electric field strength, for $n_1 = 30$ and $n_2 = 29$. A resonance occurs at $F = 22.5\,\text{V/cm}$, as indicated by a vertical, dashed line.

Figure 2.14: (a) Comparison between van der Waals interactions and direct dipole-dipole interactions between induced dipoles in the presence of an electric field of 10V/cm. (b) Comparison of critical radii governing the regime in which the van der Waals interactions are dominant. The regime is limited from above by the critical radius $R_c^{(3)}$ beyond which field-induced dipole-dipole interactions dominate (circles), and limited from below by the critical radii $R_c^{(12)}$ and $R_c^{(11)}$ below which the 'ordinary' resonant dipole-dipole interactions dominate (i.e. those not induced by the external field).
we would have retrieved the well known dipolar interaction \( \alpha_1 \alpha_2 F^2 (1 - 3 \cos^2 \theta) / R^3 \), as expected for two dipoles \( \alpha_1 F \) and \( \alpha_2 F \) aligned along the \( z \)-axis. Of course, there also exists a \( C_3^S \) coefficient associated with swapping the states in an \( n_1 S - n_2 S \) interaction, but it is approximately two orders of magnitude smaller than the other \( C_3 \) coefficients, and it is not easily expressed in terms of the polarisabilities.

In any case, however weak the electric field, for large enough \( R \) the interaction will become dipolar. Figure 2.14(a) shows the situation for \( n_1 = 30 \) and \( F = 10 \text{V/cm} \), comparing the \( C_1^{11}/R^6 \) interaction energy against the \( C_1/R^3 \) dipolar interaction energy. The tipping point where the dipolar interaction becomes stronger is denoted by \( R_c^{(3)} \), and it occurs at \( R_c^{(3)} = (C_6/C_3)^{1/3} \), which for \( F = 5 \text{V/cm} \) is at \( R_c^{(3)} \approx 5 \mu\text{m} \). The field-induced dipolar interaction thus establishes an upper limit to the van der Waals regime. Figure 2.14(b) shows the critical radii \( R_c^{(3)} \) as a function of field strength \( F \). Also plotted are the lower bounds \( R_{11} \), \( R_{12} \) below which the perturbative treatment of the van der Waals interaction is no longer valid according to Eq. (2.47). The critical radius \( R_c^{(12)} \) clearly shows the resonance at \( F = 22.5 \text{V/cm} \).

### 2.4 Many atoms: spin Hamiltonians

Having discussed one Rydberg atom in Sec. 2.2 and two Rydberg atoms in Sec. 2.3, the natural next step from a physics point of view is of course *many* Rydberg atoms. In this Section we will discuss systems of \( N \) particles, which can be excited to one or two Rydberg \( S \) states. We will see how such many-body systems can be mapped onto elementary models of interacting spins known from condensed matter physics. In particular, Rydberg atoms arranged on a lattice are seen to provide a near perfect realisation of the transverse Ising and XXZ models, as well as a spin-1 system. The realisation of the latter two systems is novel in the context of Rydberg physics, and relies on the inclusion of the second Rydberg state, with the associated swapping process providing a new, and key, ingredient.

Models of interacting spins on a lattice are of fundamental interest in condensed matter physics, as they provide elementary models to study quantum matter and quantum magnetism [118, 119]. The Hamiltonians governing the physics of such spin systems contain spin-spin interaction terms, which compete with other, *non-commuting* terms in the Hamiltonian. Each term favours a different type of ordering, which corresponds to a many-body product state in which each of the particles is in a definite single-particle state. For example, an external magnetic field could favour ferromagnetic ordering, whereas a competing spin-spin interaction term prefers anti-ferromagnetic ordering. In case a single term dominates the Hamiltonian, we thus find ground states of the system which can be understood from classical statistical mechanics. When the parameters of the Hamiltonian are varied, the balance between competing terms shifts and the system can undergo a quantum phase transition [120].
Near a quantum critical point, two terms become equally important. Due to the non-commutativity of the competing terms, the preferred orderings cannot be satisfied simultaneously, causing the ground state to ‘quantum melt’ into a superposition of product states. The individual particles become entangled and strong long-range correlations appear in the system [121–123]. Such exotic quantum phases can give rise to novel conductance phenomena, and are, e.g., thought to underly high-$T_c$ superconductivity [124, 125]. Other famous collective phenomena such as ‘normal’ superconductivity and the Fractional Quantum Hall effect also rely on entangled ground states, i.e., the BCS [126] and Laughlin [127] ansatz. Additionally, in quantum information, entanglement is viewed as a resource for performing quantum computing tasks [128–131]. Therefore, researchers are always on the lookout for reliable ways of generating entanglement between the constituents of a many-particle system.

Here, by showing that such spin systems can be realised with Rydberg atoms, we demonstrate how many-body systems of Rydberg atoms can be used as a quantum simulator for elementary condensed matter systems, and how Rydberg systems can support nonclassical, entangled ground states.

2.4.1 Many-body Hamiltonian

We consider a system of $N$ atoms, in the frozen gas limit, at fixed positions $r_i$, $(i = 1, 2, . . . N)$. The atoms are coupled to two Rydberg states $n_1S$ and $n_2S$. Each atom can be described as an effective three level system, where the atoms can be in each of three possible states: $|0\rangle \equiv |g\rangle$, the single particle ground state, and $|1\rangle \equiv |n_1S\rangle$, and $|2\rangle \equiv |n_2S\rangle$, the two Rydberg states.

The many-particle states can be written in a basis of product states of the individual particle states $|0\rangle, |1\rangle$ and $|2\rangle$, of the form

$$|\alpha_1\alpha_2\alpha_3 \ldots \alpha_N\rangle, \quad \alpha_1, \alpha_2 \ldots \in \{0, 1, 2\}. \quad (2.49)$$

For example, the state $|0102\ldots\rangle$ denotes the state with particle one in the ground state, particle two in the first Rydberg state, particle three in the ground state, particle four in the second Rydberg state, etc. We define projection operators

$$\sigma_{\beta\gamma}^{(i)} = \sum_{\alpha_1, \alpha_2 = 0, 1, 2} |\alpha_1\alpha_2/\ldots/\beta_i/\ldots/\alpha_N\rangle \langle \alpha_1\alpha_2/\ldots/\gamma_i/\ldots/\alpha_N|, \quad \beta, \gamma = 0, 1, 2 \quad (2.50)$$

which act as $|\beta\rangle \langle \gamma|$ on atom $i$, and as the identity operator on all other atoms. Recalling Eq. (2.31), the single particle Hamiltonian $\hat{H}^{(i)}$ for atom $i$ in the laser field, with detunings $\Delta_1, \Delta_2$ and coupling strengths $\Omega_{ab}$, with $ab = \{01, 02, 12\}$, can be written as

$$\hat{H}^{(i)} = -\Delta_1 \sigma_{11}^{(i)} - \Delta_2 \sigma_{22}^{(i)} + \sum_{ab} \frac{\Omega_{ab}}{2} \left( \sigma_{ab}^{(i)} + \sigma_{ba}^{(i)} \right). \quad (2.51)$$
The many-particle Hamiltonian now becomes a sum of the single particle Hamiltonians, with an added term for the pairwise van der Waals interaction potentials:

\[
\hat{H} = \sum_{i=1}^{N} \hat{H}^{(i)} + \sum_{i \neq j} \frac{1}{|r_i - r_j|^6} \left\{ \frac{1}{2} C_6^{11} \sigma_{11}^{(i)} \sigma_{11}^{(j)} + \frac{1}{2} C_6^{22} \sigma_{22}^{(i)} \sigma_{22}^{(j)} + C_6^{12} \sigma_{22}^{(i)} \sigma_{11}^{(j)} + C_6^{S} \sigma_{21}^{(i)} \sigma_{12}^{(j)} \right\},
\]

(2.52)

where the \(C_6\) coefficients are those calculated in Sec. 2.3.5. Hidden inside the Hamiltonian (2.52) are several paradigmatic interacting spin models from condensed matter theory, and in the following we will highlight the important ones. For simplicity, we restrict ourselves to a system of Rydberg atoms aligned on a one-dimensional regularly spaced chain, with interparticle distance \(a\). Furthermore, we assume that there is no coupling directly between the Rydberg levels, i.e. \(\Omega_{12} = 0\). Before proceeding with further manipulations of the Hamiltonian (2.52), we first investigate the role of the detunings and the couplings, in the ground state of the system.

Positive values of the detunings \(\Delta_1, \Delta_2\) drive the system towards states with the particles excited to the \(|1\rangle, |2\rangle\) states. This can be seen from the first two terms of Eq. (2.51), where the operators \(\sigma_{11}^{(i)}\) and \(\sigma_{22}^{(i)}\) associate an energy \(-\Delta_1\), or \(-\Delta_2\) with particle \(i\) being in the states \(|1\rangle\), or \(|2\rangle\), respectively. The values of the detunings therefore determine the total number of particles in each of the states \(|0\rangle, |1\rangle\) and \(|2\rangle\). Figure 2.15 shows the expectation value of the number of particles \(\Sigma_\alpha\) that is in the single-particle state \(|\alpha\rangle\), for \(\alpha = \{0, 1, 2\}\), in the many-body ground state for a 1D system of \(N = 7\) particles.

In the limit of zero laser intensity (\(\Omega_{01} = \Omega_{02} \rightarrow 0\)), the eigenstates of the Hamiltonian have a fixed, integer number of excitations, as is visible in the left column of Fig. 2.15. Nonzero couplings (\(\Omega > 0\)) couple these states, such that the ground state becomes a superposition of states with varying excitation numbers. This is visible in the right column of Fig. 2.15, where the discrete integer steps of the \(\Omega = 0\) case have 'quantum melted' due to non-commutativity of the \(\Omega\) terms with the other terms in the Hamiltonian, and the expectation value \(\Sigma_i\) takes on non-integer values.

When one of the detunings, say, \(\Delta_2\), is very large in magnitude and negative, then the system will no longer occupy the corresponding \(|2\rangle\) state. The \(|2\rangle\) state can be removed from the system description. On the other hand, when at least one of the detunings \(\Delta_1, \Delta_2\) is large and positive, the single-particle ground state \(|0\rangle\) will no longer be occupied, and it can be removed from the system description. It is precisely in these two limits that our system can be mapped onto the famous transverse Ising and XXZ models for interacting spin-1/2 chains. When all three levels can be occupied, we find an interacting spin-1 chain. In the following Sections we discuss these three models in turn, with special attention to the XXZ model.
Interacting Rydberg Atoms as Quantum Spin Models

Figure 2.15: Expectation value of the number of particles $\Sigma_\alpha$ in state $|\alpha\rangle$, with $\alpha = \{0, 1, 2\}$, in the many-body ground state of the Rydberg system with $N = 7$ particles on a 1D lattice with spacing $a$. The expectation values are plotted as a function of the detunings $\Delta_1, \Delta_2$, which are in units of $2C_6^{(11)}/a^6$ and $2C_6^{(22)}/a^6$, respectively. (a), (c), (e): No coupling, $\Omega_{01} = \Omega_{02} = \Omega = 0$. (b), (d), (f): Nonzero coupling, $\Omega_{01} = \Omega_{02} = \Omega = 1$MHz.
2.4.2 Transverse Ising model

First we consider the case that $\Delta_2$ is very large and negative. The $|2\rangle$ state will then no longer be populated and can be removed from the system description, leaving a system of two-level atoms with single particles states $|0\rangle$ and $|1\rangle$. Making the identification $|0\rangle \equiv |\downarrow\rangle$, and $|1\rangle \equiv |\uparrow\rangle$, we can write the Hamiltonian (2.52) in terms of the spin-1/2 Pauli matrices $\sigma_z, \sigma_x$:

$$\hat{H} = \frac{1}{2} \sum_{i=1}^{N} \left\{ -\tilde{\Delta}\sigma_{z}^{(i)} + \Omega_{01}\sigma_{x}^{(i)} \right\} + \sum_{i=1}^{N-1} W\sigma_{z}^{(i)}\sigma_{z}^{(i+1)} + \Gamma,$$  \hspace{1cm} (2.53)

with

$$\tilde{\Delta} = \Delta_1 - 2W,$$  \hspace{1cm} (2.54)

and

$$W = C_6/4a^6.$$  \hspace{1cm} (2.55)

The last term, $\Gamma$, contains the boundary effects and all contributions from interactions beyond the nearest neighbour terms:

$$\Gamma = -W(\sigma_{z}^{(1)} + \sigma_{z}^{(N)}) + W \sum_{|i-j|>1} \left( \sigma_{z}^{(i)} + \sigma_{z}^{(i)}\sigma_{z}^{(j)} \right) |i-j|^{-6}.$$  \hspace{1cm} (2.56)

Apart from this term, Eq. (2.53) is the quantum Ising Hamiltonian \[120\]. The deviation is small: the next-nearest neighbour terms are at least a factor $2^6$ smaller than the nearest neighbour interactions, and for a large number of particles $N$ the boundary terms become relatively insignificant. Moreover, it is possible to study the system on a ring lattice, e.g., using the Spatial Light Modulator discussed in Chapter 4, such that there is no longer a boundary in the system.

The realisation of the Ising model with Rydberg atoms has also been thoroughly explored theoretically, e.g., in Refs. \[89, 92, 94, 95, 133\]. It is for this reason that we will only briefly highlight its main properties here, before moving on to the novel spin-1 and XXZ Hamiltonians. When $\Omega_{01} = 0$, the Hamiltonian (2.53) reduces to the classical Ising model used for modeling magnetism in solids. The spin-spin interaction term favours anti-ferromagnetic ordering of the ground state for $W > 0$, which is the case for our Rydberg system. The $\tilde{\Delta}$-term takes the role of an external magnetic field, driving the system towards states with all particles in the $|\uparrow\rangle$ (Rydberg) state when $\tilde{\Delta} > 6W$, or all particles in the $|\downarrow\rangle$ state when $\tilde{\Delta} < -2W$. For intermediate values, the many-body ground state will have a fraction of the atoms in the $|\uparrow\rangle$ state, with crystalline order between the excitations \[89, 91\]. For $N \rightarrow \infty$ the excitation fraction forms a complete
devil’s staircase. Since the ∆ and W terms commute, the crystalline ground state is a classical product state.

When Ω_{01} is nonzero, the ground state of the system will become non-classical, as the σ_x operator does not commute with σ_z. Consequently, the constraints posed by the Ω, ∆ and W-terms cannot be satisfied simultaneously anymore. The ground state will become a superposition of the classical product states defined in Eq. (2.49). As a result, the position of the excitations and their number are no longer well-defined, and the sharp steps in excitation number visible in Fig. (2.15) will ‘quantum melt’. This allows for the staircase to be climbed, by starting in the |000...⟩ state with a large and negative ∆, which is then adiabatically varied in time. In the next Chapter we will investigate this method of climbing the staircase in more detail, and explore how states with crystalline order in the Rydberg excitations can be prepared in disordered gases.

### 2.4.3 Spin-1 model

When all three levels are significantly populated, we can again construct a mapping onto a system of interacting spins. This time, having three levels present, the system is mapped onto a spin-1 system, according to

\[ |1⟩ = |1⟩ \rightarrow |1, +1⟩, |0⟩ = |1⟩ \rightarrow |1, 0⟩, |2⟩ = |1⟩ \rightarrow |1, -1⟩ \]

(using the |s, m_s⟩ spin notation). The corresponding spin-1 operators for atom i are, written as Pauli matrices in the basis \{ |1⟩, |0⟩, |2⟩ \},

\[
\hat{J}^x(i) = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \quad \hat{J}^y(i) = \frac{i}{\sqrt{2}} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & -1 \\ 0 & 1 & 0 \end{pmatrix}, \quad \hat{J}^z(i) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix},
\]

and

\[
\hat{J}_\pm(i) = \hat{J}^x(i) \pm \hat{J}^y(i). \quad \hat{J}_z(i) = \hat{J}_x(i) \pm \hat{J}_y(i).
\]

The atomic projection operators can subsequently be written as

\[
\sigma^{(i)}_{\alpha\alpha} = \frac{1}{2} \left[ \left( \hat{J}^z(i) \right)^2 \pm \hat{J}^z(i) \right], \quad \sigma^{(i)}_{\alpha\beta} = \frac{1}{\sqrt{2}} \left[ \hat{J}^x(i) \pm \left( \hat{J}^x(i) \hat{J}^z(i) + \hat{J}^z(i) \hat{J}^z(i) \right) \right],
\]

where in both cases the + sign is understood for \( \alpha = 1 \), and the minus sign for \( \alpha = 2 \). Finally, we make the identification

\[
\sigma^{(i)}_{12} = \frac{1}{2} \left( \hat{J}^z(i) \right)^2, \quad \sigma^{(i)}_{21} = \frac{1}{2} \left( \hat{J}^z(i) \right)^2,
\]
such that the many-body Hamiltonian can now be written

\[ \hat{H} = \sum_{i=1}^{N} \left\{ -\frac{\Delta_+}{2} \hat{J}_z^{(i)} - \frac{\Delta_-}{2} \left( \hat{J}_x^{(i)} \right)^2 + \frac{\Omega_+}{2\sqrt{2}} \hat{J}_x^{(i)} - \frac{\Omega_-}{2\sqrt{2}} \left( \hat{J}_x^{(i)} \hat{J}_z^{(i)} + \hat{J}_z^{(i)} \hat{J}_x^{(i)} \right) \right\} + \sum_{i=1}^{N-1} \left\{ W^- \hat{J}_z^{(i)} \hat{J}_z^{(i+1)} + D \left[ \left( \hat{J}_x^{(i)} \right)^2 \hat{J}_z^{(i)} \hat{J}_z^{(i+1)} + \hat{J}_z^{(i)} \left( \hat{J}_z^{(i+1)} \right)^2 \right] + W^+ \left( \hat{J}_z^{(i)} \right)^2 \left( \hat{J}_z^{(i+1)} \right)^2 + S \hat{J}_z^{(i)} \right\} + \Gamma', \]

where we have defined the detunings and couplings \( \Delta_{\pm} = \Delta_1 \pm \Delta_2 \), and \( \Omega_{\pm} = \Omega_{01} \pm \Omega_{02} \), and the interaction coefficients

\[
W^\pm = \left( C_{6}^{11} + C_{6}^{22} \pm 2C_{6}^{12} \right) / 4a^6,
\]

and

\[
D = \left( C_{6}^{11} - C_{6}^{22} \right) / 4a^6.
\]

Finally, the spin-swapping operator \( \hat{J}_s \) is given by

\[
\hat{J}_s^{(i)} = \hat{J}_z^{(i)} \hat{J}_x^{(i)} \hat{J}_x^{(i+1)} + \hat{J}_z^{(i)} \hat{J}_z^{(i+1)} \hat{J}_z^{(i+1)},
\]

with corresponding interaction coefficient

\[
S = C_{6}^{S} / 2a^6.
\]

The bilinear term \( W^- \) is the familiar Ising interaction. The bi-quadratic interaction \( W^+ \) corresponds to higher order processes in magnetic systems. It may lead to interesting non-magnetic phases such as dimerized and Haldane phases in one dimensional \( S = 1 \) Heisenberg model [138] and spin nematic phase in higher dimensions [139], and may even change a second-order transition into a first-order one [140]. The term \( \Gamma' \) contains again all boundary terms and beyond-nearest-neighbour interactions.

The parameters \( \Delta_{-} \) and \( \Omega_{+} \) play the role of longitudinal and transverse magnetic fields, respectively. The term \( \Omega_{-} \) represents a local-quadrupolar order [141]. Finally, the parameter \( \Delta_{+} \) causes an axial zero-field splitting between the spin components \( |0\rangle \) and \( |1\rangle, |2\rangle \). The latter is one of the foremost factors determining the properties of many magnetic complexes, and in particular a characteristic feature of Haldane chains [142, 143]. When \( \Delta_{+} \) is large, the single-particle ground state will become energetically unfavourable, and can be eliminated, in which case we arrive at an effective spin-1/2 XXZ model.
2.4.4 XXZ model

The final paradigmatic spin Hamiltonian to be discussed here, that can be realised with Rydberg atoms on a 1D lattice, is the XXZ model \([118]\). When at least one of the detunings is very large and positive, the single-particle ground state will no longer occupied and can be eliminated. In this case, the Hamiltonian (2.62) reduces to the following spin-1/2 Hamiltonian:

\[
\hat{H} = \frac{1}{2} \sum_{i=1}^{N} \left\{ \left[ -\Delta + 4D \right] \sigma_{z}^{(i)} + \Omega_{12} \sigma_{x}^{(i)} \right\} + \Gamma'' + \\
+ \sum_{i=1}^{N-1} \left\{ W^- \sigma_{z}^{(i)} \sigma_{z}^{(i+1)} + S \left[ \sigma_{x}^{(i)} \sigma_{x}^{(i+1)} + \sigma_{y}^{(i)} \sigma_{y}^{(i+1)} \right] \right\}, \tag{2.67}
\]

where we the interaction coefficients \(W^-\), \(D\) and \(S\) are defined in Eqs. (2.63), (2.64) and (2.66), respectively, and \(\Omega_{12}\) is the effective coupling remaining after adiabatic elimination of the single particle ground state. Again, we have defined a separate term for the contributions from the boundary and interactions beyond nearest-neighbour:

\[
\Gamma'' = -D \left( \sigma_{z}^{(1)} + \sigma_{z}^{(N)} \right) + \sum_{|i-j|>1} \frac{W^- \sigma_{z}^{(i)} \sigma_{z}^{(j)} + S \left[ \sigma_{x}^{(i)} \sigma_{x}^{(j)} + \sigma_{y}^{(i)} \sigma_{y}^{(j)} \right]}{|i-j|^6}. \tag{2.68}
\]

If we assume \(\Gamma''\) is negligible, and set \(\Delta = 2D\) and \(\Omega_{12} = 0\), then the first line of the Hamiltonian (2.67) is entirely zero. The remaining second line is exactly the well known Heisenberg XXZ Hamiltonian \([118]\). Its properties have been studied extensively in condensed matter physics, as a model for quantum magnetism \([118, 119, 144]\). Like the quantum Ising model, the XXZ model possesses non-classical, strongly entangled ground states. In the XXZ model, the entanglement arises from a competition between the (non-commuting) \(W^-\) and \(S\) interaction terms in the Hamiltonian.

The \(W^-\) interaction term favours ferromagnetic ordering when \(W^- < 0\), and antiferromagnetic ordering when \(W^- > 0\). The \(S\)-term is the exchange interaction, swapping two adjacent spins, and favours an entangled, non-classical ground state called a quantum liquid. The sign of the exchange interaction can be changed by applying a transformation which depends on the lattice position \(i\) \([144]\):

\[
\sigma_{x}^{(i)} \rightarrow (-1)^i \sigma_{x}^{(i)} \tag{2.69} \\
\sigma_{y}^{(i)} \rightarrow (-1)^i \sigma_{y}^{(i)} \tag{2.70} \\
\sigma_{z}^{(i)} \rightarrow \sigma_{x}^{(i)}. \tag{2.71}
\]

This transformation changes the sign of the \(x\) and \(y\) components of the spins at all odd lattice sites, and transforms \(S \rightarrow -S\), and \(W^- \rightarrow W^-\). It is therefore sufficient to
consider only the case $S > 0$ in the following. It should be noted however that this transformation is only valid in the nearest-neighbour approximation.

Dependent on the magnitude of $W^-/|S|$ we can then distinguish three distinct phases of the XXZ model \cite{118, 144}. For $W^-/|S| < -1$, the system is in the ferromagnetic Ising phase (I-F), for $W^-/|S| > 1$ the system is in the Néel ordered, or anti-ferromagnetic Ising phase (I-AF). In between, $-1 < W^-/|S| < 1$ the system is in the XY phase. To see which phases can be realised by the Rydberg system, we plot in Fig. 2.16 the parameters $W^-$, $S$ and the ratio $W^-/|S|$ as a function of the principal quantum number $n$. Apparently, the XY (grey shaded area) and I-AF phases can be realised in the Rydberg system. However, precisely close to the interesting transition points at $n \approx 25$ and $n \approx 39$ the resonances discussed in Sec. 2.3.5 will occur. Furthermore, $n$ is not an easily and smoothly tunable parameter in an experiment.

![Figure 2.16](image)

**Figure 2.16:** (a): Interaction parameters $W^-$ and $S$ for the XXZ Hamiltonian (2.67), realised with Rydberg atoms in the $n_1 S$ and $n_2 S$ states, with $n_2 = n_1 - 1$, and plotted as a function of $n_1$. (b): Ratio of the interaction parameters, showing that the XY and Ising antiferromagnetic (I-AF) phases are accessible. The XY phase (shaded area) supports a quantum liquid ground state.

Therefore, we consider tuning the interaction parameters using an electric field $F$, as discussed in Sec. 2.3.6. The field is oriented along the direction of the 1D lattice. The resulting interaction parameters for $n = 30$ are plotted in Fig. 2.17 where we see that again the XY and I-AF phases are accessible. This time however, it is possible to tune smoothly through the transition by varying the electric field. In the entire parameter
Figure 2.17: (a): Interaction parameters $W^-$ and $S$ for the XXZ Hamiltonian realised with Rydberg atoms in the 30$S$ and 29$S$ states, plotted as a function of external field strength $F$. (b): Ratio of interaction parameters, showing that the XY and I-AF phases are accessible. The field also allows for the transition to be probed, and for a Heisenberg antiferromagnetic and pure XY Hamiltonian to be realised at $F = 8.16\, V/cm$ and $F = 4.12\, V/cm$, respectively.

range there are no resonances, as can be gathered from Fig. 2.14(b). Moreover, with the continuous parameter $F$ at our disposal we can now realise two interesting special cases of the XXZ Hamiltonian. At $F = 4.12\, V/cm$ the $W^-$ interaction is exactly zero, and the Hamiltonian (2.67) reduces to the well-known XX and XY Hamiltonians, which possess a rich phase diagram when we allow the $\sigma_z$ and $\sigma_x$ terms in the first term of Eq. (2.67) to become nonzero [121, 122, 145]. At the point $F = 8.15\, V/cm$ we have $W^- = 0$ and the Hamiltonian takes the form of the famous antiferromagnetic Heisenberg Hamiltonian [119, 124]. Finally, at $F = 10.65\, V/cm$ the exchange interaction $S$ becomes zero, and we realise a pure antiferromagnetic phase with Néel order.

To visualise the transition from the XY phase to the I-AF phase, we plot in Fig. 2.18 the density of excitations to the $|1\rangle$ state, for $N = 9$ particles and $n = 30$, in the many-body ground state of the full Hamiltonian (2.67). That is, we set $\Omega_{12} = 0$ and $\Delta_- = 4D$, but the boundary and beyond-nearest-neighbour interactions ($\Gamma''$) are included. The colors in Fig. 2.18 indicate the probability of finding a particle in state $|1\rangle$, as a function of the field strength, and for each lattice position. The particles are
Figure 2.18: Probability density of $n_1S$ excitations in the many-body ground state, for $N = 9$ atoms on a 1D lattice with spacing $a$, as a function of particle position and electric field strength. For clarity, the atoms are endowed with some small spatial extent. Vertical dashed lines indicate the transition from the XY phase to the I-AF phase ($F = 8.16\, \text{V/cm}$) and the pure antiferromagnetic phase ($F = 10.65\, \text{V/cm}$). In the XY phase, some residual antiferromagnetic order is visible, which we ascribe to boundary effects. The density of excitations to the $|1\rangle$ state is exactly the complement of this graph.

endowed with some small spatial extent for clarity. Clearly, for $F < 8.15\, \text{V/cm}$, the probability of finding a $|1\rangle$ excitation is roughly 0.5 for each of the lattice sites. By computing the entropy of entanglement [146] of a single spin with respect to the others, we have verified that the ground state is indeed entangled. When $F$ is increased, the probabilities shift towards a situation with alternating high and low probability on each site, until a pure, unentangled, antiferromagnetic configuration is reached at the point $F = 10.65\, \text{V/cm}$. The total sum of the excitations to the $|1\rangle$ state is conserved as $F$ is varied, and equal to 5. That is, when we would perform a measurement on the system, we will always find a total of 5 particles in the $|1\rangle$ state, with the remaining 4 particles in the $|2\rangle$ state.

The above range of interaction parameters of the Hamiltonian is unique for $n = 30$, for other values of $n$ it is not possible to tune through the transition between the XY and I-AF phases. The only exception being $n = 26 - 29$, where the same is in principle possible, however much larger fields are needed in those cases. For $n = 31$ to $n = 39$ the system starts in the I-AF phase, and stays there for increasing field strength, until a resonance occurs. For $n \geq 40$, the system starts and stays always in the XY phase for increasing field strength, until a resonance occurs.
2.5 Conclusions and outlook

Rydberg atoms have highly exaggerated properties, due to the large spatial extent of the wavefunction of the valence electron. In particular, the range of the van der Waals interaction between Rydberg atoms can extend to several micrometers. The associated $C_6$ coefficients are highly tunable, by choice of $n$, and by imposing external fields. Moreover, the interactions are switchable: by exciting or de-exciting atoms the interactions can be switched on and off.

We have calculated the van der Waals interactions between Rydberg atoms in $S$ states, revealing a swapping mechanism in the case when the two atoms have different principal quantum numbers $n_1$ and $n_2$. In such a swapping interaction, the two Rydberg atoms switch state. The swapping is found to be only significant when $|n_1 - n_2| = 1$. The $C_6$ coefficients associated with the swapping, as well as those associated with the regular interaction between the $n_1S$ and $(n_1 - 1)S$ states, are found to have resonances close to $n_1 = 25$ and $n_1 = 39$.

A many-body system of atoms coupled to the two Rydberg $n_1S$ and $n_2S$ states gives rise to a near perfect implementation of several paradigmatic spin systems known from condensed matter physics. Besides a previously known quantum Ising model, we find novel Rydberg implementations of a spin-1 system and the famous XXZ model. The inclusion of the second Rydberg level is essential, it provides a key ingredient in the form of the swapping interaction.

The tunability of the Rydberg interactions allows for a large parameter regime to be probed. In particular, we focus on a system of 30$S$ and 29$S$ Rydberg atoms arranged on a 1D lattice as an implementation of the XXZ model. By varying an external electric field between 0 and 15V/cm, the many-body ground state can be tuned smoothly between an entangled, quantum liquid, XY phase, and an ordered antiferromagnetic phase whose properties can be understood by classical statistical mechanics. Moreover, it should be noted that all of the above spin models are available with the same setup, consisting of two laser couplings and a static electric field. Mere changes in laser detuning, intensity and electric field strength allow one to move in a continuous fashion between models.

Looking to the future, the first issue to address is the experimental realisability. An obvious approach is by adiabatic chirping of the laser intensity and detuning, a method already successful (in theory) for preparing crystalline ground states of the quantum Ising model (see e.g. Ref. [91], and also the next Chapter). For the $n = 30$ XXZ system, it remains to be seen if the ground state can actually be prepared within the lifetime of a Rydberg atom, which is limited at around 20 $\mu$s [147]. Other methods could be dissipative state preparation, as suggested in Ref. [148]. Should the lifetime turn out to be a limiting factor, then there is always the possibility to switch to higher values of $n$, considering that lifetimes scale as $n^3$. Although at the loss of some tunability, the quantum liquid phase would still be accessible for $n > 40$.

Once experimental realisability is addressed, there are many more possibilities and
extensions for realising spin models. Field induced dipole moments can give rise to controlled next-nearest neighbour interactions. Furthermore, instead of S states, one could envisage orientation-dependent interactions with D states. Microwaves could selectively shift states in and out of resonance, adding another degree of tunability. Going to higher dimensional lattices, a degree of frustration can be introduced, possibly giving rise to the fabled, and much sought-after spin liquid ground state [149, 150].

In conclusion, interacting Rydberg systems can act as highly flexible quantum simulators of some of the most fundamental condensed matter spin systems. The addition of a second coupled Rydberg S state greatly enriches the available physics. Finally, the entangled many-body states that can be generated in the Rydberg system will have great prospects in quantum information science.

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Bibliography


Interacting Rydberg Atoms as Quantum Spin Models


Interacting Rydberg Atoms as Quantum Spin Models


Adiabatic Formation of Rydberg Crystals with Chirped Laser Pulses

Abstract - Ultracold atomic gases have been used extensively in recent years to realize textbook examples of condensed matter phenomena. Recently, phase transitions to ordered structures have been predicted for gases of highly excited, “frozen” Rydberg atoms. Such Rydberg crystals are a model for dilute metallic solids with tunable lattice parameters, and provide access to a wide variety of fundamental phenomena. We investigate theoretically how such structures can be created in four distinct cold atomic systems, by using tailored laser-excitation in the presence of strong Rydberg-Rydberg interactions. We study in detail the experimental requirements and limitations for these systems, and characterize the basic properties of small crystalline Rydberg structures in one, two and three dimensions.
3.1 Introduction

Since the development of techniques to cool and trap atomic gases with laser radiation in the 1990’s, it has been possible to realize and study many-body phenomena in dilute, ultracold gases. Some of these phenomena are straight out of solid-state textbooks, such as the Mott-Hubbard transition \[151, 152\]: the transition from a “conducting” state to an “insulator” state. In optical lattices, which are artificial crystals of light created by standing-wave laser beams, bosonic atoms could be trapped with a filling factor of unity, and a superfluid to Mott-insulator transition could be observed.

One serious shortcoming of such cold atom experiments with regard to the comparison with condensed matter physics is that neutral ground-state atoms interact via short-range Van der Waals interactions, which are of course much weaker than the Coulomb interactions between electrons in solids. Ultracold gases can be made strongly-interacting by using Feshbach resonances \[153, 154\], however, it does not mean that the system is automatically strongly-coupled \[155\]. These gases are typically very dilute, such that the Van der Waals range \(r_0 = (mC_6/h^2)^{1/4}/2\) of the ground-state atoms, where \(m\) is the atomic mass and \(C_6\) the Van der Waals coefficient, is much smaller than the average distance between the particles \(\rho^{-1/3}\), with \(\rho\) the density. The coupling parameter \(\Gamma\) that indicates the transition between weakly coupled and strongly-coupled systems, is given by the ratio of interaction energy \(E_{\text{int}}\) and kinetic energy \(E_{\text{kin}}\). Even for a unitary Fermi gas \[156\], which is by definition strongly interacting, the coupling parameter is smaller than 1: \(\Gamma = |E_{\text{int}}/E_F| \approx 0.6\), with \(E_{\text{kin}} = E_F\) the Fermi energy.

A much closer realization of a strongly-coupled condensed matter system would arise if the atoms could be separated into ions arranged on a lattice, and surrounding electrons that provide the conductance phenomena. On the way of creating such a dilute metallic system, an intermediate step would be the excitation of an ultracold gas to a system with a few strongly-coupled Rydberg atoms \[157\]. Rydberg excitations into electronic \(s-\)orbital states interact, similar as the ground-state atoms, via Van der Waals interactions. However, the Van der Waals coefficient scales very rapidly with the principal quantum number as \(C_6 \sim n^{11}\) leading to a strong repulsive interaction, and therefore it is possible to reach the regime \(r_0 > \rho_e^{-1/3}\) where \(\rho_e\) indicates the Rydberg atom density. In this regime neighboring atoms feel and avoid each other, and under strong-coupling conditions of small or negligible kinetic energy it may lead to ordered or correlated structures. Moreover, further collective excitation of these structures brings both ions and electrons into the strongly-coupled ultracold plasma regime when the charge-density distributions of neighboring atoms overlap. Such a system may then be considered as a dilute model-system for metallic solids \[158\].

Robicheaux and Hernández predicted already in 2005 many-body correlations which can rise from the Rydberg blockade effect (also referred to as dipole blockade effect) in a disordered gas \[159\], hinting at the possibility of ordered structures. Later, Weimer et al. showed that a phase transition could occur from a strongly interacting Rydberg
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gas to a crystalline phase \([160]\), for positive detuning from the ground-state to Rydberg transition. Since then, several proposals have been made to create correlated systems that involve Rydberg atoms: for instance by means of dynamical crystallization of Rydberg atoms starting from ground-state atoms \([161, 163]\), or by weakly dressing ground-state atoms to form super-solid droplet crystals \([164, 166]\). In this Chapter, we investigate a very practical scheme which was first proposed by Pohl, Demler and Lukin \([161]\), which allows for a laser-assisted, adiabatic self-assembly of a Rydberg atom lattice. We investigate the robustness of a tailored excitation scheme to build up a correlated crystal state from a disordered atomic gas. Although this Chapter has a theoretical nature, our investigations are inspired by the present ultracold atom set-up in our group in Eindhoven. This experiment provides us with a tool to detect spatially correlated Rydberg crystal structures, by means of an accelerator infrastructure (see Fig. 3.1). The set-up contains a magneto-optical trap for rubidium atoms with appropriate 780 nm trapping lasers. We routinely trap \(\approx 10^8 \ ^{85}\text{Rb}\) atoms at densities up to \(10^{17}\) atoms/m\(^3\) and with temperatures below 1 mK. Both pulsed (10 ns YAG-pumped dye) and continuous-wave, single-frequency (300 mW solid-state) 480 nm lasers are present for Rydberg excitation \([167]\). The atom trap is built inside an accelerator, followed by a beam transport system that allows one to create magnified images of both ions and electrons onto a microchannel plate and phosphor screen assembly that is observed with a cooled CCD camera. The acceleration field can be switched on to 10kV/cm on sub nanosecond time scales. This set-up has been used to study electron and ion beams generated from the laser-cooled atoms by near-threshold photo-ionization and by field-ionization of Rydberg atoms. Both ions and electrons can be detected with a multichannel plate as a measure for the total number of Rydberg atoms produced. By directly ionizing the sample, and thus creating an ultracold plasma, followed by a rapid extraction of charged particles, we demonstrated that this technique can be used to produce cold bright beams of ions and electrons, with temperatures in the beam of the ions of \(T=1\text{mK}\) and for the electrons of \(T=10\text{K}\) \([167, 168]\).

This Chapter is organized as follows: In Section 3.2 we give a theoretical description of a cold atomic gas in the frozen gas limit, and the corresponding Hamiltonian that describes it. In Section 3.3 we argue how Rydberg crystals can be created by adiabatically chirping the laser frequency. In Section 3.4 we look in detail into four distinct cold atomic systems which we consider for creating Rydberg lattices. In Section 3.5 we present our results for crystal formation using tailored excitation pulses for one-dimensional systems. In Section 3.6 we investigate a few basic crystal properties of two- and three-dimensional systems, and finally we conclude in Section 3.7.
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Figure 3.1: Schematic overview of our experimental set-up, on which the model parameters are based. (a) Atoms are cooled and trapped in a Magneto-optical trap. (b) Then an ionization beam is utilized to create an ultracold plasma. Due to the accelerator, the electrons and ions can be used to create bright beams of electrons or ions. The ultracold particle accelerator (pictured in (c)) provides us with a tool to spatially resolve the Rydberg lattices.

3.2 System description

Throughout this Chapter we will be working in the frozen gas limit, where the excitation dynamics happen on a time scale so fast that we can neglect all motion of the atoms [161] [169], i.e., ignore their kinetic energy.

We will investigate four different systems: (i) Atoms in a deep optical lattice. (ii) Condensed atoms in a BEC, where the Thomas-Fermi approximation can be made. (iii) Thermal atoms in a dipole trap, where temperatures can be in the µK regime. And finally, (iv) Thermal atoms in Magneto-Optical Trap (MOT) conditions, with temperatures typically in the sub-mK regime.

Under all of the above conditions, the frozen gas approximation is satisfied. Even for the highest temperatures in the MOT, it takes tens of µs for atoms to move significantly on the crystal scale of 5 − 10µm [161] [169]. The Doppler limit, which gives rise to the temperature of the atoms in the MOT, can be expressed as a velocity that is about 0.14 m/s for rubidium. With this velocity, at the time scales of interest of 1 − 10µs, we can safely assume that the interaction energy between Rydberg atoms is constant. Nevertheless, a dephasing mechanism based on Doppler broadening due to the thermal motion of the atoms could potentially frustrate lattice formation. In the following sections, we show how crystal creation relies on adiabatic changing of system parameters. In 3.2 we verify, in a two-particle picture, that Doppler broadening does not affect such dynamics.

In general, the systems that we describe consist of $N$ atoms in a laser field. We assume an effective laser coupling between the ground state and a particular Rydberg state, such that we can describe each atom as a two level system. The $N$-body excitation state can then be represented by basis states of the form $|n\rangle$, where $n$ is a $N$-dimensional
vector with elements \( n_i = \{0, 1\} \), indicating whether the \( i \)-th atom is in the ground (0) or excited (1) state. First, we consider the special situation in which the \( N \) atoms are localized at fixed positions. We will shortly show how this picture can be applied to more general systems. The positions of the \( N \) atoms are denoted with the \( 3 \times N \) vector \( \mathbf{r} \), where \( \mathbf{r}_i \in \mathbb{R}^3 \) is the position of the \( i \)-th atom. In the rotating frame, the Hamiltonian governing the dynamics of the excitation state vectors \( |n⟩ \) of the atoms is given by

\[
\hat{H}_{\text{exc}}(\mathbf{r}) = -\sum_{i=1}^{N} \hbar \Delta \hat{\sigma}_{11}^{(i)} + \frac{1}{2} \sum_{j=1}^{N} \hbar \Omega \left( \hat{\sigma}_{10}^{(j)} + \hat{\sigma}_{01}^{(j)} \right) + \sum_{i=1, j>i}^{N} \frac{C_6}{|\mathbf{r}_i - \mathbf{r}_j|^6} \hat{\sigma}_{11}^{(j)} \hat{\sigma}_{11}^{(i)},
\]

(3.1)

where \( \hat{\sigma}_{11}^{(i)} = |1_i⟩⟨1_i| \) counts whether the \( i \)-th atom is excited, and \( \hat{\sigma}_{10}^{(i)} = |1_i⟩⟨0_i| \) and \( \hat{\sigma}_{01}^{(i)} = |0_i⟩⟨1_i| \) change the state of the \( i \)-th atom. The first two terms in Eq. (3.1) describe the atom-laser interaction, with \( \Delta \) the laser detuning and \( \Omega \) the effective Rabi frequency of the system. The third term equals the Van der Waals interaction between atoms in the Rydberg state.

In practice there are often many individual atoms spaced so closely together, that we can safely say that within a certain group of atoms there will never be more than one excitation due to the Rydberg blockade [170]. That is, the interaction energy associated with more than one excitation within a certain group of atoms is so large, that any multiply excited state is no longer resonant with the excitation laser. Moreover, we cannot (e.g. because of detection resolution), or need not, distinguish which particular atom carries an excitation. Such groups of atoms can be said to coherently share an excitation, and are commonly called superatoms [171]. We can partition the space into \( M \) volumes \( V_k \), \( k = 1..M \), each of which contains a group of \( M_k \) atoms which together represent the \( k \)-th superatom. The Hamiltonian (3.1) can be equivalently applied to describe superatoms instead of single atoms, where the positions \( \mathbf{r}_i \) are taken to be weighted averages of atomic positions within a superatom, and the Rabi frequency \( \Omega \) is replaced by a collective Rabi frequency \( \Omega_k \) for the \( k \)-th superatom [159, 170–173]:

\[
\Omega \rightarrow \Omega_k = \sqrt{M_k} \Omega.
\]

(3.2)

This collective Rabi frequency reflects the fact that there are multiple atoms in a superatom, and that the superatom as a whole is more susceptible to the laser light. This latter fact becomes important later on, once we consider dense, inhomogeneous systems. Moreover, in Sec. 3.5 we shall perform simulations of the \( N \)-body system and superatoms will prove to be a useful means for reducing the size of the Hilbert space under consideration.

Having introduced the Hamiltonian for a collection of particles localized at fixed positions, it is now time to turn our attention to more general systems such as Bose-Einstein condensates with fully coherent wavefunctions, or statistical mixtures found in
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thermal clouds. To this end, we expand the Hilbert space $H_e$ of excitation configurations $|n\rangle$, with the Hilbert space of $N$-body spatial wave functions $H_r$, to form the total product Hilbert space $H_{total} = H_r \otimes H_e$.

A convenient basis for this statespace is given by states

$$|r\rangle \otimes |n\rangle,$$  \hspace{1cm} (3.3)

where $|r\rangle$ is the state with all $N$ particles localized at positions specified by the $3N$-dimensional vector $r$. The Hamiltonian operator (3.1) is straightforwardly extended to the total Hilbert space:

$$\hat{H} = \int d^3r |r\rangle \hat{H}_{exc}(r) \langle r|.$$  \hspace{1cm} (3.4)

It is in this final step where the frozen gas approximation becomes apparent: in absence of kinetic energy, the Hamiltonian $\hat{H}$ is diagonal in the spatial components.

Using the basis states (3.3), and assuming that all particles start in the ground state $|n\rangle = |0\rangle$, we can write the general state of the system at $t = 0$ as

$$|\Psi(0)\rangle = \int d^3r c_0(r) |r\rangle \otimes |0\rangle.$$  \hspace{1cm} (3.5)

When $t > 0$, this state starts to evolve under the action of the Hamiltonian (3.4), written down formally with the time evolution operator $\hat{U}(t)$:

$$|\Psi(t)\rangle = \hat{U}(t)|\Psi(0)\rangle = \int d^3r c_0(r) \hat{U}(t) \left( |r\rangle \otimes |0\rangle \right).$$  \hspace{1cm} (3.6)

The frozen gas Hamiltonian (3.4) however cannot make transitions between states with different spatial parts $|r\rangle, |r'\rangle$, with $r \neq r'$. Therefore, the time evolution operator of Eq. (3.6) will only affect the excitation part $|n\rangle$ of the many-particle state $|r\rangle \otimes |n\rangle$, and not the spatial part $|r\rangle$, so that we can write

$$\hat{U}(t) \left( |r\rangle \otimes |0\rangle \right) = |r\rangle \otimes \hat{U}_{r}^{exc}(t)|0\rangle.$$  \hspace{1cm} (3.7)

Here, the time evolution operator $\hat{U}_{r}^{exc}(t)$ governs the excitation state of all the particles under the action of the Hamiltonian (3.1), given the fact that they are pinned at definite locations $r$. The general time-dependent state in Eq. (3.6) can then be written as

$$|\Psi(t)\rangle = \int d^3r c_0(r)|r\rangle \otimes |\chi_{r}(t)\rangle,$$  \hspace{1cm} (3.8)

where we have defined excitation state vectors.
that describe the excitation state of a given atomic configuration $|r\rangle$. The frozen gas Hamiltonian (3.4) only affects the excitation part $|\chi_r(t)\rangle$, which thus carries the time dependence whereas the spatial part $|r\rangle$ remains “frozen”. It should be noted that $c_0(r)$ and $c_{n,r}(t)$ thus satisfy separate normalization conditions given by

$$\int d^3r |c_0(r)|^2 = 1, \quad \sum_{n=0}^{2N-1} |c_{n,r}(t)|^2 = 1.$$ (3.10)

From the above discussion, we can conclude that the time evolution of a system in the state (3.5) can be broken down into the time evolution of the individual, position basis components $|r\rangle$ that make up the total state. Each such component evolves completely independent from the others as a result of the diagonality of the Hamiltonian (3.4).

Expectation values of observables of interest, $\hat{O}$, that are independent of momentum, such as the excitation density, are computed for each of the individual time evolved configurations. The results are combined in an incoherent sum, in which the coefficients $|c_0(r)|^2$ appear in terms of their modulus squared:

$$\langle \hat{O} \rangle = \int d^3r |c_0(r)|^2 \langle r \otimes \chi_r(t) | \hat{O} | r \otimes \chi_r(t) \rangle.$$ (3.11)

Similarly, expectation values of observables in statistical mixtures, such as thermal clouds in a trap, are also computed using incoherent sums over individual time-evolved configurations. Here, the coefficients $|c_0(r)|^2$ in Eq. (3.11) are replaced by the statistical probabilities of finding a particular atomic configuration $r$. In the remainder of this Chapter we shall therefore employ Eq. (3.11) for both coherent states as well as thermal clouds, with the understanding that the $|c_0(r)|^2$ coefficients are statistical probabilities in the latter case.

### 3.3 Adiabatically creating Rydberg crystals

In the previous Section we have argued how the time evolution of the many-particle state can be broken down into the time evolution of the individual, position-basis states $|r\rangle \otimes |\chi_r(t)\rangle$ that make up the total state $|\Psi\rangle$ of Eq. (3.8). To see how a crystal state can adiabatically evolve in the frozen gas limit, we now single out a particular component configuration $|r\rangle$ of the atoms, and analyse the time evolution of the corresponding excitation state $|\chi_r(t)\rangle$. Since we restrict ourselves to one particular configuration $|r\rangle$, we shall omit the spatial ket from the product state notation.

In the limit of zero laser intensity, $\Omega \to 0$, the uncoupled states $|n\rangle$ are eigenstates
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Figure 3.2: (Schematic) Energy of the basis states $|r⟩\otimes|n⟩$ (thin, gray lines), as a function of the detuning, in the limit of zero laser intensity ($\Omega \rightarrow 0$), and for a given, fixed, choice of the frozen atom positions $r$. All energies depend linearly on the detuning with a slope given by the number of excitations $m$, and the offset at $\Delta = 0$ is determined by the Van der Waals interaction energy between the Rydberg atoms. For each value of $m$ there exists a specific configuration with the lowest energy (thick, red line), in which the Rydberg excitations are typically regularly ordered (schematically pictured configuration on the left). Less ordered states have higher energies (example configuration on the right). Note that the atomic positions do not change, merely the excitation localization is different between the two configurations. For non-zero laser coupling $\Omega > 0$, the ground state (dashed line) becomes a superposition of basis states and separates in energy, which can be adiabatically followed in the direction of the arrow when slowly changing the detuning.

of the Hamiltonian (3.1). The energy $E_n$ of each state $|n⟩$ assumes a simple linear dependence on the detuning:

$$E_n = -m\Delta + E_{int},$$

(3.12)

where $m = \sum_i n_i$ is the total number of excited atoms in the state $|n⟩$, and $E_{int}$ is the total interaction energy between the excited atoms in this particular configuration. Clearly, the energies of all states with the same number of excited atoms have the same slope as function of the detuning $\Delta$. However, depending on the specific location of the $m$ excitations, each state has a different offset, $E_{int}$, at $\Delta = 0$ due to the interaction energies.

As such, all states with a specific excitation number $m$ form a manifold in the $E - \Delta$ plane, as illustrated in Fig. 3.2 by sets (a “manifold”) of gray lines (individual states) with the same slope. Now, there are two crucial points to be noted.
Firstly, the lowest lying state within each manifold is typically an ordered, crystalline state, indicated with a solid, red line in Fig. 3.2. That is, the excitations will tend to be found at regular spacings (in Sec. 3.6 we discuss the crystal structure in more detail). The states with the same excitation number, but a less ordered distribution of excitations will typically have a much higher energy. This is indicated schematically with two example configurations in Fig. 3.2. For each value of the detuning, there will be a ground state with one specific excitation number, and one specific configuration of excitations, which has the lowest energy of all possible states. For $\Omega \to 0$, the ground state will always be an ordered, crystalline state.

The second important point concerns what happens when $\Omega > 0$, and regards the possibility to adiabatically create a crystal state. As the laser intensity $\Omega$ is increased from zero, it couples the different states $|n\rangle$ through the second term in the Hamiltonian (3.1). Crossings between states from adjacent manifolds in Fig. 3.2 now turn into anticrossings. In particular, the ground state now becomes a superposition of states $|n\rangle$, and separates in energy from the others, as indicated by a black, dashed line in Fig. 3.2 lying below all others.

A scheme to generate a crystal state would then be as follows. We start the system with a large negative detuning, $\Omega = 0$, and all atoms in their atomic ground state, such that we are also in the ground state of the many particle system. When the laser coupling is then turned on, and subsequently the detuning is raised slowly enough, the many particle state will adiabatically follow the ground state [161], as indicated by the blue arrow in Fig. 3.2. This process can continue until some final detuning is reached, at which point the laser coupling $\Omega$ is switched off again. If this last step is also performed adiabatically, the system will end up in the $\Omega = 0$ ground state, which is one of the previously discussed ordered crystal states, with a given number of Rydberg excitations each of which is localized on exactly one of the frozen atoms, and regularly spaced, forming a crystal.

The preceding discussion has focused on one particular configuration of atoms $|\mathbf{r}\rangle$. However, the system state is typically built up from many different configurations, either as a superposition of the form (3.5) in the case of a BEC, or a statistical mixture in the case of a thermal ensemble. As explained in the previous section, expectation values of observables for such systems are formed by incoherent sums of the results computed for such individual configurations. Typically, crystal structures are retained in this summation procedure.

In the next section we will outline experimental conditions under which adiabatic crystal preparation is theoretically possible, followed by Sec. 3.5 where we will perform simulations of such experiments.
3.4 Physical systems

In this section we discuss in more detail the different experimental systems where Rydberg crystal creation is possible. We strict ourselves to a (quasi) 1D geometry for simplicity as well as for computational feasibility, however, it should be noted that the principle of crystal creation is straightforwardly extended to higher dimensions. We consider $^{85}$Rb atoms initially in the ground state $5s^2 S_{1/2}$, which we can excite to the $n = 65$ Rydberg state $ns^2 S_{1/2}$ by using a two-step laser excitation scheme. The first step involves an off-resonant $5s-5p$ 780 nm transition with Rabi frequency $\Omega_1$ and detuning $\hbar \delta$, and the second step involves a $5p-ns$ 480 nm transition with Rabi frequency $\Omega_2$. The two-photon Rabi frequency is then given by $\Omega = \Omega_1 \Omega_2 / 2\delta$. For the $n = 65$ Rydberg state, the Van der Waals coefficient is given by $C_6 = 2.4 \times 10^{-58}$ Jm$^6$ \[^{[174]}\]. We also consider a fixed 1D excitation volume, of length $a = 45 \mu m$, which will be defined by the excitation laser profiles and / or the spatial dimensions of the atomic system.

The four main types of systems in ultracold atom experiments are (i) optical lattices, (ii) Bose-Einstein condensates, (iii) thermal clouds in an optical dipole trap (high density and low temperature), and (iv) thermal clouds in a MOT (low density and relatively high temperature). For each of these systems we will show under which conditions Rydberg crystal creation should be possible, and we provide some typical numbers as well as investigate possible drawbacks and advantages.

(i) Optical lattices: The conceptually simplest system is formed by ultracold atoms trapped in a deep optical lattice. Here we consider a slightly idealised system as an example from which we can understand the more complicated systems. We assume that within the excitation volume there is a fixed and uniform number of atoms $N_A$ per site, and that the excitation volume is sharply defined. The lattice is assumed so deep that the atoms are in the Mott insulator phase and there is no hopping, such that the atoms are trivially in the frozen gas limit. The system is then represented by a regularly spaced chain of superatoms, each representing $N_A$ individual atoms. Typically, $N_A$ is simply equal to 1, and lattice spacings of $1 \mu m$ are commonly realised \[^{[175]}\], such that there in the order of 40 – 50 atoms within the excitation volume, all experiencing the same coupling to the laser field.

The crystal states in turn are formed by regularly spaced excitations of the atoms. The critical detunings $\Delta_{m+1}^m$ for which the lowest energy excitation configuration changes from $m$ to $m + 1$ excitations, can be estimated as \[^{[161]}\]

$$\hbar \Delta_{m+1}^m = \frac{C_6}{a^6} \left( m^7 - (m-1)^7 \right). \quad (3.13)$$

Here we have assumed that the excitations are regularly spaced, and ignored next-nearest neighbour interaction energies. Thus, to create a crystal of $m$ excitations, the final value of the detuning at the end of a chirp should lie between $\Delta_{m-1}^m$ and $\Delta_{m+1}^m$.

Finally, we like to mention that crystal creation in optical lattices in a 1D geometry
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has already been extensively investigated by Schachenmayer et al. in reference [162], finding interesting effects due to discrepancies between lattice and excitation spacing. Similarly, quasi 1D systems in a ring geometry have been thoroughly studied by Olmos et al. [172, 173, 176, 177] who, for instance, make use of symmetries to reduce computational costs.

(ii) Bose Einstein Condensates: The next type of system we consider is a Bose-Einstein condensate (BEC) in the Thomas-Fermi limit [178]. The main difference with the optical lattice is a much higher number of particles in the excitation volume, as well as a non-uniform continuous density distribution. As we saw in the superatom picture described in Sec. 3.2, this affects the way the system responds to laser light. In the Thomas-Fermi (TF) limit, the density profile $\rho(r)$ assumes a parabolic form as a function of position $z$ along the tube: $\rho(z) = \rho_0 [1 - (z/R_z)^2]$, where $R_z$ is the TF-radius of the condensate and $\rho_0$ is the central density. The excitation volume can be created by a wide 780 nm beam illuminating the entire condensate, and a narrowly focused 480 nm laser passing through the cloud, requiring $R_z = 22.5 \mu m$ to obtain a $45 \mu m$ long tube. The diameter of this tube will be set to $4 \mu m$, which is reasonable regarding the diffraction limit for this wavelength.

Typically, $\rho_0 \approx 10^{14} \text{ cm}^{-3}$ [170, 179], leading to $O(10^4)$ particles in the excitation volume and a condensate well within the Thomas-Fermi limit.

Since the density of the system is not uniform, the many-particle position configurations $|r\rangle$ of Eq. (3.8) also have a non-uniform probability distribution $c_0(r)$ associated with them. In particular, the probability distribution $P$ of the position $r_L$ of the leftmost particle is equal to

$$P(r_L \leq r) = F(r_L)^N, \quad (3.14)$$

where $F(r)$ is the cumulative distribution function of the single particle probability distribution. The probability density for the rightmost particle is found by mirroring Eq. (3.14) around $r = 0$. These probability distributions are important, as the outermost particles of a particular configuration $|r\rangle$ are certain to carry an excitation in the corresponding ground state. As a result, the crystal size $a$ used in calculating the critical detunings $\Delta^{m+1}_m$ in Eq. (3.13) will vary between different configurations $|r\rangle$, and the critical detunings will likewise assume a probability distribution for their values. Fig. 3.3(a) shows the probability distribution of critical detunings $\Delta^5_4$, $\Delta^6_5$, and $\Delta^7_6$, relevant for creating a 5 or 6 excitation crystal, calculated with $3.6 \cdot 10^4$ particles in the excitation volume. As opposed to the optical lattice case, the distributions now have a certain width, for instance the $\Delta^5_6$ value has a spread of about 0.5 MHz about its mean value. To create a 6 excitation crystal with high fidelity one therefore has to have a final detuning at least 0.5 MHz away from the mean value of $\Delta^7_6$. The peaks are well enough separated that it is at least always possible to find a suitable detuning at which we are certain to have a definite excitation number in the ground state. As we will shortly see in the case of thermal clouds, this need not always be the case. The inset of Fig. 3.3(a) shows the
probability distribution for the locations of the excitations in the 5-excitation ground state. Due to the probability distribution of the outermost particles, the peaks are not infinitely sharp but have a finite width.

(iii - iv) Thermal clouds in optical dipole traps and MOTs: Thermal clouds with Gaussian density distributions form the starting point for many experiments in ultracold atoms. There is a great variety in sizes and density, so we will treat two specific cases, clouds confined in an optical dipole trap (ODT) and clouds contained in a magneto-optical traps (MOT).

Clouds confined in an ODT are much smaller in size than those in a MOT, and they reach much higher densities. Typical peak densities $\rho_0$ in ODTs range between $10^{12} - 10^{15}\text{cm}^{-3}$ \cite{170, 180}.

We consider a cylindrically symmetric density of the form

$$\rho(r, z) = \frac{N}{(2\pi)\kappa^2\sigma_z^3} \exp(-r^2/2\kappa^2\sigma_z^2 - z^2/2\sigma_z^2), \quad (3.15)$$

Figure 3.3: Target detuning probability densities for (a) a BEC in the Thomas-Fermi limit, (b) a thermal cloud in an optical dipole trap, and (c) a thermal cloud in a magneto-optical trap. The critical values for the optical lattice, as given by Eq. (3.13), are superimposed as vertical, dashed lines. Insets: probability distribution for the locations of excitations in the 5-excitation ground state.
where \( \kappa \) is the aspect ratio of the cloud, \( N \) the total number of atoms in the cloud, and \( \sigma_z \) the width of the distribution in the \( z \)-direction. To create an excitation volume of \( 45\mu m \) long in the ODT, we can proceed in the same fashion as was the case of a BEC, by using a defocused 780 nm beam illuminating the entire sample, and a narrowly focused 480 nm beam aimed through the center of the cloud along the \( z \)-axis. For typical numbers \( N = 4 \cdot 10^5 \) atoms, an aspect ratio of \( \kappa = 10 \) and a width of \( \sigma_z = 7\mu m \), we get 2000 particles in a tube of \( 4\mu m \) in diameter. These values are precisely such that the expectation values of the positions of the outermost particles in the tube are spaced \( 45\mu m \) apart.

As was the case for the BEC, the nonuniform probability distribution for the particle density causes the critical detunings of Eq. (3.13) to assume a probability distribution as well. Fig. 3.3(b) shows the probability density for the critical detunings \( \Delta_5, \Delta_6, \) and \( \Delta_7 \) relevant for creating a 5 or 6 excitation crystal in our excitation volume in the optical dipole trap. Apparently, in this case the probability densities overlap, which means that there exist no values of \( \Delta \) in which the ground state is certain to have one particular number of excitations, there is always a finite probability to have one more, or one less excitation than expected. The inset of Fig. 3.3(b) shows the probability distribution for the locations of the excitations in the 5-excitation ground state.

Clouds in a magneto-optical trap (MOT) are typically much larger in size, with diameters of \( O(1 \text{ mm}) \). The \( 45\mu m \) excitation volume therefore needs to be created by shaping the intensity profiles of the excitation lasers. By passing the 780 nm laser through an aperture, it is possible to create a beam of width \( 45\mu m \) and approximately uniform intensity locally within the atom cloud. Intersecting this beam with a small, focused beam with diameter \( 4\mu m \) of the 480 nm laser near the center of the atom cloud results in the desired 1D excitation volume. Inside this volume the atomic density is practically uniform, and with typical densities of a MOT of \( \rho = O(10^{10} - 10^{11}) \text{ cm}^{-3} \) there are approximately 10 to 60 atoms at uniformly distributed positions contained within the excitation volume. Figure 3.3(c) shows the critical detunings for such a system for the particular case of 30 atoms. The probability distributions form peaks with distinctly asymmetric shapes, which are a result of the sharp boundaries of the excitation volume. The inset of Fig. 3.3(c) shows the probability distribution for the locations of the excitations in the 5-excitation ground state, which also exhibit the characteristic asymmetry due to the sharp boundary of the excitation volume.

As a final note to this section, we would like to mention that it is in principle possible to change system parameters such as the dimensions of the excitation volume. However, due to the 6-th power scaling of the Van der Waals interaction energies this has a large effect on the other system parameters. For instance, changing all length scales by a factor \( \lambda \) is equivalent to changing all time scales by a factor of \( \lambda^6 \), and consequently all frequencies such as \( \Omega \) and \( \Delta \) need to be scaled by a factor \( \lambda^{-6} \). Even a simple change of the excitation volume length from \( 45\mu m \) to \( 90\mu m \) would therefore require all timescales to be multiplied by a factor of 64 in order to obtain the same results. The chirps that
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we will construct for our particular excitation volume and particular experimental setup
can therefore be said to be truly tailor made.

3.5 Performing the chirp

3.5.1 Chirp properties

Having outlined the properties of the physical systems of interest, we proceed by com-
posing a realistic chirp of the laser detuning $\Delta$ and intensity $\Omega$. The basic ingredients
for such a chirp are as follows.

1. Start by switching on the laser and increase the coupling strength $\Omega$ from zero up
to some final value. This is done at a large, negative detuning with all particles
starting in their atomic ground state, coinciding with the many body ground state.

2. Once the coupling $\Omega$ is sufficiently high, the detuning can be increased from its
initial value towards its final target value $\Delta_f$ between $\Delta_{m-1}$ and $\Delta_{m+1}$.

3. Switch off the laser, by lowering the coupling strength back to $\Omega = 0$ again.

The above three steps should be carried out as adiabatically as possible, yielding a crystal
state at the end of the chirp, such as depicted in the insets of Fig. 3.3. In Figure 3.4,
panels (a), we show various typical chirps, modified by a computer algorithm to maximise
adiabaticity given a constrained time frame of $\sim 5\mu s$ in which it is to be carried out.
Each chirp is tailor made for a specific system. The remaining panels (b) and (c) of Fig.
3.4 will be discussed in Sec. 3.5.3.

Starting with step (i) of the chirp, the first feature to be noted is the shape of $\Omega(t)$ in
the first section, which is roughly exponential. As the coupling is still small, the energy
levels are closely spaced together, and only small changes to the Hamiltonian can be
made without making transitions to excited states. When the coupling is stronger, the
ground state gets pushed to lower energies and separates further from the other energies,
and as a consequence we can make larger changes to the Hamiltonian and increase the
rate of change of the coupling. As the final value is reached, the ramp abruptly stops and
$\Omega$ remains constant. Since the Schrödinger equation is a first order differential equation
in time, such sharp changes in slope do not affect adiabaticity and there is no need to
smoothen the chirp.

When in the second step the detuning is increased, we found that the maximum slope
varies non-monotonically throughout the chirp. Regions where the detuning can only be
changed relatively slowly are not necessarily correlated to the critical detunings $\Delta_{m+1}$
described in Sec. 3.4. This is a sign of a complicated underlying energy landscape, whose
details are beyond the scope of this Chapter.

The most critical step of the chirp is the final step, switching off the laser and low-
ering $\Omega$ back to zero again. When the detuning is large and positive, the energy scales
in the system become larger, such that the coupling strength becomes relatively smaller. Moreover, in the case of a non-uniform density, such as the ODT or BEC, the $\sqrt{M_k}$ enhancement of the coupling in Eq. (3.2) necessitates a very fine control over the final stages of switching off the coupling, as even small laser intensities can still couple strongly to the central, high density region of the excitation volume. Because of these two phenomena, step (iii) typically takes up the greatest amount of time of the total crystal creation procedure. In the case of the ODT and BEC the time scale for switching off the laser is an order of magnitude larger than the combined time for switching it on and chirping the detuning. Finally, it should be noted that the rate of change in $\Omega$ varies, as another sign of a complicated underlying energy landscape. We also found that it is favourable to decrease the detuning slightly while ramping down the intensity.

3.5.2 Simulating excitation dynamics

We now have all the necessary ingredients to perform a realistic chirp and simulate an attempt to create a Rydberg crystal in each of the four systems described in Sec. 3.4: an optical lattice, a BEC and thermal clouds in an ODT and a MOT. We aim to create 5 or 6 excitations in total, due to limitations imposed by the computational complexity.

Following the ideas outlined in Sec. 3.2, the simulation can be broken down into two parts. The first part consists of simulating the time evolution of the excitation dynamics of a given set of atoms at fixed positions. Then, in the second part a Monte Carlo integration over particle distributions is performed, summing many simulations for different atomic positions weighted by the probability that a certain configuration occurs.

For the simulation of the excitation dynamics for a fixed configuration, we perform a simulation of the full quantum many particle state, governed by the Schrödinger equation with Hamiltonian (3.1). We use a fifth order adaptive stepsize Runge Kutta time integration [181], with the Cash-Karp parameter scheme [182]. For the optical lattice we assume 45 particles in the excitation volume, whereas in the case of the BEC and the ODT we assume $N = O(10^3 - 10^4)$ atoms in the excitation volume, and 30 in that of the MOT. Since the parameter space for $N$ two level systems scales as $2^N$, we need to make some approximations to make the simulations tractable. The first approximation consists of reducing the $O(10^3 - 10^4)$ atoms in the ODT and BEC to 40 superatoms, by employing the algorithm suggested in Ref. [159] where we recursively replace the closest spaced pair of (super)atoms by a weighted average position. In the end, we replace the couplings to the superatoms by the $\sqrt{M_k}\Omega$ coupling of Eq. (3.2), where $M_k$ denotes the number of atoms represented by the $k$-th superatom. But even the resulting $2^{40}$ states space is too large to handle, and we reduce the number of states further following the suggestions put forth in Refs. [160] [161] [183], where we omit all states with more than a certain maximum number of excitations ($> 8$ in our case), and also omit all states where the excitations are spaced too close together. This latter approximation is made under the assumption that these states will never be significantly populated due to the
Rydberg blockade. These three approximations reduced the state space to $O(10^5)$ states, a number low enough to allow for a few hundred simulations per day on an 8 core system.

3.5.3 Results

For each of the systems described in Sec. 3.4 we have performed simulations of a tailor made chirp of laser detuning and intensity. The laser chirp is pre-determined per system by an algorithm that attempts to maximize adiabaticity, while keeping constraints on the total duration of the chirp. In this way, we have constructed chirps that create 6 excitations in the optical lattice and BEC, and 5 excitations in the thermal clouds in an ODT or MOT. All chirps have a duration of about 5 $\mu$s.

For the optical lattice, the simulation only needed to be performed for a single configuration of atoms, since we assumed that the ground state atoms are at very well localized positions. For the other three systems we have performed many Monte Carlo simulations to account for the probabilistic distribution of the ground state particles. At the beginning of each simulation, we draw new particle positions from the appropriate particle distribution, and then simulate the time dynamics when this particular atom configuration is subjected to the laser chirp. In total, we have performed 600 Monte Carlo samplings for the BEC, 1650 samplings for the ODT and 3300 samplings for the MOT.

After all the simulations are completed, the results can be aggregated in order to compute expectation values of various observables. In particular, for each system we compute expectation values for the number of excitations, the excitation density, and the pair correlation function.

These expectation values can be constructed using the excitation number operator $\hat{\rho}_e(r)$, which records the number of excitations measured at the (1D) distance $r$ from the origin (not to be confused with the many particle position vector $r$). We define this operator through its matrix elements in the basis (3.3):

$$\langle r' \otimes n' | \hat{\rho}_e(r) | r \otimes n \rangle = \delta(r - r') \delta_{nn'} 1_{r}(r, n),$$

(3.16)

where $1_{r}(r, n)$ is an indicator function, which takes on the value of 1 if the state $|r \otimes n\rangle$ has an excited particle at position $r$, and 0 otherwise. The corresponding expectation value is found from Eq. (3.11):

$$\langle \hat{\rho}_e(r) \rangle = \int d^3r \sum_{n=0}^{2^N-1} |c_0(r)|^2 |c_{n,r}(t)|^2 1_{r}(r, n).$$

(3.17)

Note that the coefficients $c_0(r)$ follow from the Monte Carlo sampling, whereas the coefficients $c_{n,r}(t)$ are calculated in the simulation of the time dynamics.

Using the operator $\hat{\rho}_e(r)$ we can additionally compute the second-order density cor-
relation function \[ G_{(2)}(r, r') = \langle \hat{\rho}_e(r)\hat{\rho}_e(r') \rangle - \delta(r - r')\langle \hat{\rho}_e(r) \rangle, \tag{3.18} \]

which measures the conditional probability of finding an excitation at point \( r' \), given that there already is an excitation found at \( r \). It is more convenient to compute the integrated second-order density correlation function

\[ G^{(2)}(r) = \int G^{(2)}(r, r + R) \, dR, \tag{3.19} \]

which measures just the probability of finding an excitation a distance \( r \) away from any other excitation. Calculated on a discrete grid, the correlation function \( G^{(2)} \) can be thought of as a histogram of pair separations.

The final observable we calculate is the probability \( p(m) \) of finding \( m \) excitations, which can be calculated as

\[ p(m) = \int d^3 r |c_0(r)|^2 \sum_{n=0}^{2^N-1} |c_{n,r}(t)|^2 \, 1_m(n), \tag{3.20} \]

where we have defined an additional indicator function \( 1_m(n) \), which takes on the value 1 if the state \( n \) has \( m \) excitations, and 0 otherwise.

We are now ready to discuss the results of the simulations. Figure 3.4 shows the calculated expectation values for each of the four systems we are interested in: (i) optical lattice, (ii) Bose-Einstein condensate, (iii) thermal cloud in an ODT, (iv) thermal cloud in a MOT. For each system, the top panel, (a), shows the chirp of the laser intensity \( \Omega \) and detuning \( \Delta \), the second panel, labeled (b), shows the expectation values \( p(m) \) of Eq. (3.20) of the number of excitations, the third panel, (c), shows the excitation density (3.17), where all expectation values are plotted as a function of time.

There are a number of similarities and differences to be noted between the four systems. Starting with the top panel (a), we see that the shape of each chirp follows roughly the three-step scheme described in Sec. 3.5.1, although the length of each segment varies between systems. Typically, the third step of switching the laser off takes the longest time.
Figure 3.4: Simulation results for Rydberg crystal creation in each of the four systems discussed in the text: (i) optical lattice, (ii) Bose-Einstein condensate, (iii) thermal cloud in an ODT, and (iv) thermal cloud in a MOT. All results are shown as a function of time. In each subfigure, the top panel, labeled (a) shows the laser intensity $\Omega$ and detuning $\Delta$, the center panel, labeled (b) shows the probability $p(m)$ of finding $m$ excitations. The bottom panel, (c), shows the spatial excitation density $\langle \hat{\rho}_e(r) \rangle$ during the chirp, indicated with colors ranging from white (lightest) to blue to red (darkest), where darker colors indicate a higher probability of finding an excitation at that position and time.
Panels (b) and (c) of Fig. 3.4 show clearly what happens during the three steps of the chirp. When the laser is switched on in the first step, the initial state $|0\rangle$ with all atoms in the ground state becomes a superposition of states with 0 or 1 excitations. Then, as the detuning is increased while the intensity is at its maximum, states with higher numbers of excitations start to mix into the system state. No particular number of excitations has a high probability of occurring (panel (b)), and the location of the excitations is also spread out across the excitation volume (panel (c)). Only when in the final step the laser is switched off do the excitations localize at regularly spaced positions, and the Rydberg crystal takes shape. First, the outermost excitations localize, and then excitations crystallize progressively inwards towards the center of the excitation volume. In panel (i.c) for the excitation density of the optical lattice, one can distinguish the individual atoms at each lattice site, and see how the excitations are initially shared among all atoms and crystallize only at the end of the chirp.

The middle section of the chirp takes the shortest time in the systems with high particle numbers, the BEC (ii) and the ODT (iii), as a result of the enhanced $\sqrt{M_k}$ coupling in the superatom picture [see Eq. (3.2)]. The effect of this enhanced coupling is also clearly visible in panels (a) of Fig. 3.4 for systems (ii) and (iii), where a disproportionally long time is needed to ramp down the last few percents of laser intensity. The center panels (b) show that in this long flat tail of the intensity chirp there are still large changes in the excitation number probabilities.

Fig. 3.5 shows the excitation density (3.17) at the end of the chirp (column (a)), and correlation function (3.19) at the end of the chirp (column (b)), for each of the four systems (i) - (iv) of interest.

Starting with the excitation density in the optical lattice in the top left panel (a.i), we see that the excitations are more or less perfectly localized on individual lattice sites. Upon close inspection there are small probabilities discernable, for finding the excitation at neighbouring lattice sites, indicating that the time evolution has not been perfectly adiabatic. The excitation density of the BEC in panel (a.ii) also exhibits distinct and well-separated peaks. However, the excitation density of the thermal cloud in an ODT in panel (a.iii) shows peaks that, although clearly visible, are not entirely separated spatially. This is due to the boundaries of the excitation volume not being as sharply defined as those of the BEC. In the ODT, the spread in the position of the outermost particles is largest of all four systems. Even the MOT excitation density in panel (a.iv) shows better separated peaks. Due to the sharp boundary of the excitation volume in this system (see Sec. 3.4) the peaks show a roughly similar shape to those of the BEC. However, they exhibit a larger spread around their mean positions, reflecting the much lower number (30) of particles present in the excitation volume. In effect, the low particle number offers fewer positions for the excitations to localize on, and the optimal positions leading to the lowest energies are often not occupied in a particular Monte Carlo simulation.

Finally, moving to column (b) of Fig. 3.5 showing the correlation function for each system, we can extract the following information. Interpreting the correlation functions...
Figure 3.5: (a): Excitation density, and (b): correlation function $G^{(2)}(r)$ at the end of the chirp for each of the four systems under consideration: (i) optical lattice, (ii) Bose-Einstein condensate, (iii) thermal cloud in an ODT and (iv) thermal cloud in a MOT. Note that the optical lattice and BEC have 6 excitations, whereas the ODT and MOT have 5 excitations.

$G^{(2)}$ as histograms for the distances between pairs of excitations, we clearly see that there are never two excitations within one peak of the excitation densities in column (a). Should this have been the case, then we would find nonzero probabilities at distances smaller than the inter-peak distance of $\sim 10\mu m$.

The first peak in $G^{(2)}$ appearing around $10\mu m$ is the distribution of pair separations of excitations in neighbouring peaks in the excitation density. Likewise the second peak in $G^{(2)}$ is the distribution of pairs in peaks separated by a single other peak, followed by the pair distributions of excitations in peaks separated by two peaks, etc. The height of the peaks in the correlation function $G^{(2)}$ directly reflects the number of pairs of peaks separated by 0, 1, 2,.. other peaks, leading to a linear decay in height. The width of the peaks signifies how regularly the excitations are spaced. Again, in case (iii) of the ODT the peaks are the least sharply defined, as a direct result of the excitation volume not being bounded sharply. In case (i), the optical lattice, there are small sub-peaks visible again, which are correlations between the large peaks and sub-peaks of the excitation density in panel (a.i).
3.6 Higher dimensional systems

In the previous section, we focused in detail on the adiabatic creation of one-dimensional Rydberg crystals. However, the dimensionality of the crystal structures can be controlled by the shape of the volume set up by the excitation lasers. As 1D structures are produced by a needle-shaped light field, so can 2D structures be created by a sheet of light intersecting the atomic cloud. 3D structures can arise in a light volume where excitations are not prohibited by dimensional restrictions. In this section we take a closer look at what type of 2D and 3D structures may arise when applying the same type of laser excitation techniques in 2D and 3D light volumes, and investigate the energy of the ground states as function of the number of Rydberg atoms. This latter quantity is of importance when determining the critical detunings $\Delta_{m+1}^m$ described in Sec 3.4 at which the system transitions from an $m$-excitation ground state to an $m+1$-excitation ground state.

While in the 1D case the crystal structures are basically similar to regularly spaced chains of Rydberg excitations, in higher dimensions the structures can be quite complicated. However, for simplicity, we assume that the crystal structures with $m$ excitations are confined by a circular plane (2D) or spherical volume (3D), which have hard wall boundaries and where the interior can be considered as homogeneous with no external potential.

We calculate the optimal crystal structures by using a Conjugate Gradients minimization of the energy of $m$ point particles interacting via a repulsive two-body Van der Waals $C_6/r^6$ potential:

$$E_{int}(r_1, r_2, \ldots, r_m) = \sum_{i=1}^{m} \sum_{j>i}^{m} \frac{C_6}{|r_i - r_j|^6}. \quad (3.21)$$

The minimization is performed with respect to the particle positions $r_i$. Since this energy represents a very complex $2m$ (2D) or $3m$ (3D) dimensional energy landscape, with many local minima, we have performed hundreds of minimizations for each Rydberg particle number $m$ with random initial conditions, and recorded the lowest energy configuration thus found as a good approximation for the global minimum. There exists a large body of literature in the field of Coulomb crystals where similar procedures are found to find crystal structures of trapped ions, see e.g. Refs. [185][189], and references therein.

In Figs. 3.6 and 3.7 examples can be seen of crystal structures of Rydberg excitations, for a given number of $m$. Typically, we observe in 2D that at low particle numbers $m$ all particles are located on the boundary of the volume, maximizing the distances between the nearest neighbours. As $m$ is increased the boundary fills up, until at a certain point it becomes energetically favourable for the new particle to be placed in the center of the volume. As the particle number is increased further, more particles will be located near the center, forming a ring shaped structure. This process continues until, in turn, a
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Figure 3.6: Examples of 2D crystal structures as function of Rydberg atom number $m$. Initially, when there are few Rydberg atoms, all excitations reside on the boundary of the excitation volume in order to maximise the interparticle distance. As the number of Rydberg atoms confined in the circular volume increases, the inner region of the excitation volume starts to fill as well. At $m = 7$ the first particle appears in the center, followed at $m = 10$ and $m = 12$ by a second and third one respectively. At $m = 18$, an additional shell starts to form at the center, progressively filling up ($m = 24, 26, ...$) until there are so many particles in the interior that boundary effects become negligible and a transition to bulk behaviour is observed, where a hexagonal lattice is formed in the central region ($m = 61$ and $m = 129$). The corresponding energies of these configurations are shown in Fig. 3.9(a).

Generally speaking, for larger numbers of excited atoms a competition occurs between lattice structures in the interior, and shell structures near the boundaries of the system, an effect familiar from Coulomb crystal studies [185, 188]. Positions on the boundary of the system are energetically favourable due to the (partial) absence of neighboring atoms, leading to boundary effects (shell structures), propagating inward until the boundary is sufficiently far away such that bulk behavior (lattice structures) start to dominate. For a sufficiently large number of Rydberg atoms, the structure can then be regarded as a
(quasi) periodic, homogeneous lattice. However, for small atom numbers we enter the regime of mesoscopic physics, where we create symmetric few-body clusters.

In 3D systems we see the same chain of events, with consecutive shells forming when the particle number is increased. Here, we expect lattice formation in the interior only at much higher Rydberg atom numbers since surfaces of spheres can accommodate much more particles than the circular boundary in the 2D case. We have verified that up to \( m = 1000 \) we still see distinct shell structures in 3D.

As a function of the number of Rydberg atoms in the lattice, we show in Fig. 3.8 where the atoms will be located as a function of the radial distance to the center of the excitation volume. Both in 2D and 3D the formation of shells is visible for increasing Rydberg atom number \( m \).

In Fig. 3.9 the minimum energy \( E(m) \) is shown as a function of the particle number \( m \), for the 2D and 3D case. The energy solutions show three distinct regimes: the few-particle regime where boundary effects dominate, the bulk regime with many particles, and inbetween the mesoscopic regime which connects the two limits. Each regime has a different scaling of energy versus particle number. In the bulk regime the energy scales as \( m^4 \) in 2D and as \( m^3 \) in 3D. This scaling behavior assumes a homogeneous distribution of the particles with a mean interparticle separation, and where each particle has a fixed number of neighbours. Furthermore, we assume only nearest neighbour interactions. In the few-particle regime, edge effects dominate, which results in a relatively larger interparticle separation as in the bulk regime. However, despite a lower than bulk energy, the energy increases more rapidly with \( m \). In the mesoscopic regime, the system is inbetween the edge-dominated and bulk dominated regimes, which results in energy scaling laws as \( \sqrt{m}m^4 \) for 2D and \( \sqrt{m}m^3 \) for 3D.

**Figure 3.7:** Example of a 3D crystal structure. (a) displays the structure as a whole, while in (b) the inner shells can be seen, with an indication of the average distance of the shell to the center of the structure.
Figure 3.8: Shell formation for a 2D (left) and 3D (right) Rydberg lattice. The Rydberg atom number density is indicated as a function of the radial distance to the center of the system (horizontal axis) and for a varying total number of Rydberg atoms (vertical axis). Each horizontal slice of the figure corresponds to particular configuration, as indicated for instance in Figs. 3.6 and 3.7. When the number of atoms is increased, additional shells appear (blue / red fuzzy lines) that move toward greater radial distance to make room for yet more inner shells. A large part of the atoms resides on the boundary of the system, corresponding to the bright line at radial distance $r = 1$. In the 2D system, the onset of lattice formation and breakdown of shell structure can be observed near $r = 0$ for high atom numbers where the shell structure becomes more diffuse.

Figure 3.9: Minimum energy $E$ as function of the number of Rydberg atoms $m$, for the distribution of $m$ atoms in (a) a two-dimensional circular plane, (b) a three-dimensional spherical volume. The insets display the behavior for smaller atom numbers. The continuous lines represent fits according to the mesoscopic (blue, solid line) and bulk (red, dashed line) scaling laws.
3.7 Conclusions and outlook

While crystalline phases have been predicted for strongly interacting Rydberg gases, it is not straightforward to achieve such an ordered structure starting from a disordered gas. A practical scheme to enter this regime has been proposed by Pohl et al. [161], using chirped laser pulses. In this Chapter, we investigate the feasibility to create one-dimensional Rydberg crystals in four distinct cold atomic systems, which vary in density and temperature, and demonstrate the importance of an excitation scheme which is exactly tailored to the specific system parameters. Even for a system that is relatively low in density and high in temperature, such as a magneto-optical trap, it is possible to design an excitation scheme that creates spatially ordered structures. A successful experimental realization of one-dimensional Rydberg crystals allows for a direct comparison to predicted analytical properties in a lattice [190]. For crystalline Rydberg structures in two or three dimensions, we calculate some basic properties such as the spatial structure, the corresponding ground state energy, and shell structure formation as function of the Rydberg atom number.

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Appendix 3.A A closer look at the frozen gas approximation

Throughout this Chapter, we assumed that the system is sufficiently cold such that we can make the frozen gas approximation, and hence neglect the kinetic energies of the atoms during the course of a typical experiment lasting a few microseconds. In a typical experiment, the laser detuning is swept through a few MHz within a few µs, and in this Appendix we investigate whether thermal motion under MOT conditions can significantly affect the final state after a sweep.

We consider a two particle two-level system with a time-dependent Rabi frequency and detuning, described by the coupled set of optical Bloch equations for the coupled two-particle amplitudes $|c_{gg}|^2$, $|c_{ge}|^2$, $|c_{eg}|^2$, and $|c_{ee}|^2$, where subscripts $g$ and $e$ denote ground or excited states. The time-dependent equations are given by
Figure 3.10: Velocity-averaged two-particle squared amplitudes as function of time, during the course of an adiabatic chirped transition from the ground state to the excited Rydberg state. Shown are $|c_{gg}|^2$ (red), $|c_{ge}|^2 + |c_{eg}|^2$ (blue), and $|c_{ee}|^2$ (gray). From $t = -4 \mu s$ to $t = 4 \mu s$, $\delta$ is chirped from -15 MHz to 15 MHz in a linear fashion. $\Omega$ is held constant at 1.4 MHz. The interaction energy $V_{int}$ of the $c_{ee}$ state is (a) 0.2 MHz and (b) 7.8 MHz, corresponding to a distance of about 11 and 6 $\mu m$ respectively. The absolute squares of the amplitudes are averaged over a Maxwell-Boltzmann distribution with a temperature corresponding to a Doppler limit of 0.5 MHz. Also shown are the instantaneous eigenstates of the ideal system with $v_1 = v_2 = 0$ (black line). The decaying Rabi oscillations can be seen, which are found to damp out in a Gaussian fashion with a time constant $\tau \approx 0.45 \mu s$, in agreement with Eq. 3.26. Note that the oscillations in (a) experience an adiabatic revival at $t = 4 \mu s$ because of the symmetry of the sweep.

\[
\begin{align*}
    i \frac{d c_{gg}(t)}{dt} &= \frac{\Omega(t)}{2} (c_{ge}(t) + c_{eg}(t)), \\
    i \frac{d c_{ge}(t)}{dt} &= \frac{\Omega(t)}{2} (c_{gg}(t) + c_{ee}(t)) + (-\Delta(t) + kv_2) c_{ge}(t), \\
    i \frac{d c_{eg}(t)}{dt} &= \frac{\Omega(t)}{2} (c_{gg}(t) + c_{ee}(t)) + (-\Delta(t) + kv_1) c_{eg}(t), \\
    i \frac{d c_{ee}(t)}{dt} &= \frac{\Omega(t)}{2} (c_{ge}(t) + c_{eg}(t)) + (-2\Delta(t) + kv_1 + kv_2 + V_{int}) c_{ee}(t). \tag{3.25}
\end{align*}
\]

The Doppler shifts $kv_i$ are added to the detuning, which allow for different velocities for particle $i = 1, 2$. A conservative estimate for these shifts is of order $3 \cdot 10^6$ rad/sec or 0.5 MHz, resulting from the effective transition wavelength of 300 nm, that gives rise to the laser wavenumber $k$. The dephasing that arises is a result of the distribution of velocities $v_i$ for an ensemble of atoms, which we assume to obey Maxwell-Boltzmann statistics, and this will damp Rabi oscillations. The time scale associated with this damping is

\[
\tau = \sqrt{\frac{2m(\Omega^2 + \Delta^2)}{\Delta^2 k^2 k_B T}}, \tag{3.26}
\]
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with \( m \) the atomic mass and \( k_B \) the Boltzmann constant. However, damping will not be a limiting factor for the experimental conditions which we consider in this Chapter, provided that the chirp is wide enough, as can be seen in Fig. 3.10. Here an adiabatic transition is shown for two ground state atoms to their excited Rydberg states, by presenting the velocity averaged two-particle amplitudes. The calculation is done for two different interparticle separations, and the squared amplitudes \( |c_{gg}|^2, |c_{ge}|^2 \) etc. are averaged over Maxwell-Boltzmann distributions, corresponding to the Doppler temperature, for velocities \( v_i \). The figure shows damped Rabi oscillations. Nevertheless, the amplitudes are able to follow adiabatically the instantaneous eigenstates if the chirp is slow enough, and wide enough, meaning that \( |\Delta(t_{\text{initial}}) - kv| \gg \Omega(t_{\text{initial}}) \) and \( |\Delta(t_{\text{final}}) - kv| \gg \Omega(t_{\text{final}}) \).

In conclusion, since the Rydberg crystal creation relies on adiabatic processes, dephasing of Rabi oscillations does not inhibit the crystallization process.

Bibliography


Chapter 3.


The Spatial Light Modulator

Abstract - Ultracold atom experiments often require precisely controlled optical potentials. This is particularly true for the Rydberg experiments discussed in this thesis. As a first step towards performing these Rydberg experiments, the creation of arbitrary optical potentials is investigated, both theoretically and experimentally, with the use of a spatial light modulator (SLM). An SLM is a commercially available device, able to imprint a spatially varying phase onto a laser light field and thus controlling the light intensity in the focal plane of a lens. Existing techniques and algorithms are explored for finding appropriate phase patterns, and improved upon with a novel feedback algorithm. Furthermore, experimental issues such as aberrations in the optical path are addressed and corrected. Finally, the SLM is implemented in an existing ultracold experiment, demonstrating the capability to shape excitation and ionisation lasers and imprint spatial structures inside an atomic cloud.
4.1 Introduction

In the field of ultracold atoms, the most valuable item in the toolchest of an experimenter is undoubtedly the laser. With the help of coherent laser light, atoms are trapped and manipulated with a level of control unthinkable a couple of decades ago [191]. After initial successes in spectroscopy, the creation of atomic clocks, ultracold chemistry, and optical lattices, attention is shifting towards even more advanced applications, such as the elusive quantum computer [192] and its close cousin, the quantum simulator [193, 194].

As the experiments become more and more complex, so too do the requirements to the laser fields that provide the trapping potentials, or that act as detection mechanisms, or perform other tasks, such as exciting atoms to high-lying Rydberg states. Needless to say, crafting and shaping arbitrary optical landscapes has become an important part of the experimental setup in atom optics.

There exist many methods of generating arbitrary light fields in cold atom experiments, such as time-averaged potentials for creating dipole traps [195, 196], intensity masking [197, 198], or diffractive optical elements [199–203]. In this Chapter we focus on diffractive optical elements (DOEs), that are transmissive or reflective optical elements that modulate the phase of a light field, without affecting the amplitude [204]. This way, it is possible to control the intensity of the light field in the far field, or equivalently, in the focal plane of a lens [205]. Since the DOE only modulates the phase of the light, but not its amplitude, no energy is absorbed. Consequently, the DOE is able to obtain high diffraction efficiencies, whereby a large fraction of the light can be made to contribute to the final desired intensity pattern [206].

Earlier DOEs typically came in the form of a slab of material with a variable thickness [204]. Light passing through, or reflecting from such an element thus traverses a variable optical path length, giving rise to a phase modulation. Such elements have to be manufactured and tailor made for each specific application, and are therefore very unflexible. Changing demands to the light pattern would require replacing the DOE in its entirety.

Recent years saw the emergence of liquid crystal based devices [207, 208], called Spatial Light Modulators (SLMs), allowing a reconfigurable phase pattern to be imprinted on a light field. Typically, an SLM is a device which can work either in transmission or reflection, with light passing through an array of pixels. Each of these pixels consists of a bi-refringent liquid crystal with an externally controllable polarisation axis. Linearly polarised light incident on a pixel emerges with a phase delay dependent on the orientation of the liquid crystals, thus providing spatial control of the phase of the light field.

The current state of the art Spatial Light modulators consist of millions of pixels, with up to hundreds of possible phase levels per pixel. These specifications allow for a great level of control of the light field in the spatial domain. This outstanding performance is however to be contrasted with that in the temporal domain. With refresh rates of around 60Hz, dynamic modulation on atomic timescales still seems far away.
However, for applications requiring only static light potentials, SLMs have already been very successful, for instance, in creating smooth ring traps for investigating superfluidity [209], arrays of dipole traps [210] [211], atom guiding [212] [214], arbitrarily shaped dipole traps [215], or shaping ultracold electron bunches [216] [217]. Among notable applications of SLMs outside ultracold atoms are holographic optical tweezers [218].

The crucial point in successfully using a spatial light modulator is the design of the phase pattern to be imprinted on the light field. Given a desired intensity pattern, there is no straightforward direct method of computing the required phase pattern, and often intricate numerical algorithms are required for designing the phase pattern. Luckily, a large amount of research has been done on this topic in the context of DOEs [204] [206] [210] [223], and more recently also with SLMs in mind [215] [224].

In this Chapter we will review the most successful methods for designing phase patterns, and show how they can be applied to SLMs in an actual experiment. The detailed knowledge of these algorithms helps us to understand and tackle issues that arise in practice.

In Section 4.2 we will discuss the basic experimental setup and operating principles of the SLM, i.e., how modulation of the phase can be made to control the light intensity in a certain region of space. In Secs. 4.3 and 4.4 we review two key methods for constructing phase patterns, and show how these can be used to create particular intensity patterns in Sec. 4.5. In Sec. 4.6 we discuss some typical issues arising in the use of a SLM, and show how our detailed knowledge of the phase design problem can help resolve those. We demonstrate a powerful feedback method capable of creating higher quality intensity patterns than previously presented in literature.

Throughout the Secs. 4.3 to 4.5 we present various example phase and intensity patterns. Besides theoretical results we shall also present experimental measurements, obtained with a 1920 × 1080 pixel HoloEye Pluto NIR II spatial light modulator with 256 discrete phase levels [225], and using 780nm near-infrared laser light. The active area of the HoloEye Pluto is 15mm × 8mm, and an \( f = 90 \) mm achromatic lens is used for focusing the diffraction images. Finally, in Sec. 4.7 this SLM is implemented in an existing experiment, where it is used to shape ultracold ion bunches, as a first step towards spatially controlled Rydberg excitation.

### 4.2 Phase modulation

In this section we will review the basic working principles behind operating a spatial light modulator. In particular we will review, through basic diffractive optics, how the modulation of the phase leads to intensity modulation.

The basic setup for a phase-modulating SLM is shown in Figure 4.1(a). Here, the SLM is located in the plane \( z = -L \), followed by a lens with focal length \( f \), placed at the origin \( z = 0 \), and an observation plane situated at the focal point of the lens at \( z = f \).
In scalar diffraction theory, the laser light field in free space is described by a complex valued function $U(x, y, z)$, possessing an amplitude and a phase. The time dependent phase $e^{i\omega t}$ can be omitted from the description. In the setup of Fig. 4.1 the light field is incident on the SLM at $z = -L$, and of the form $U(x, y, -L) = |U_0(x, y)|$, i.e. we assume it to be real-valued with constant phase. We shall carefully keep track of the light field amplitude and phase as it propagates through the optical system. The light field is transmitted through the SLM plane, which acts as a rectangular aperture, of size $L_x \times L_y$. Inside this aperture, the light field is subject to a modification of the phase, such that at an infinitesimal distance $h$ directly behind the SLM we have that

$$U(x, y, -L + h) = U_0(x, y)e^{i\phi(x, y)}\text{rect}\left(\frac{x}{2L_x}, \frac{y}{2L_y}\right)$$

(4.1)

where $\phi(x, y)$ is the phase delay imposed by the SLM, and the rectangular aperture function is defined as:

$$\text{rect}(\xi, \eta) = \begin{cases} 1 & (|\xi| < 1, |\eta| < 1) \\ 0 & \text{otherwise.} \end{cases}$$

(4.2)

To avoid having to write the rect function each time, we define the (real-valued) input amplitude $U_i$ as

$$U_i(x, y) = U_0(x, y)\text{rect}\left(\frac{x}{2L_x}, \frac{y}{2L_y}\right).$$

(4.3)

Assuming that the Fresnel approximation holds for the propagation between the SLM plane and the focal plane, and assuming that the SLM is positioned at precisely one focal length away from the lens, i.e. $L = f$, we then have that the light fields in the SLM and focal plane are related by a Fourier transform. That is, the light field $U_f = U(x', y', f)$, at the point $x', y'$ in the focal plane $z = f$, is given by

$$U_f(x', y') = \mathcal{F}\left[U_i(x, y)e^{i\phi(x, y)}\right]\left(\frac{x'}{\lambda f}, \frac{y'}{\lambda f}\right),$$

(4.4)

where $\lambda$ is the wavelength of the light, and $\mathcal{F}[\cdot](u, v)$ denotes the Fourier transform, evaluated at frequencies $u, v$. Displacing the SLM from the front focal plane of the lens ($L \neq f$) adds an unimportant quadratic phase to the light field, for which we found that it can be ignored to good approximation in practice.

As is apparent from Eq. (4.4), the intensity $|U_f|^2$ in the focal plane can be influenced by modulating the phase $\phi$ of the light field in the SLM plane. However, since we have no (dynamical) control over the amplitude, $U_i$, of the light field in the SLM plane, the calculation of the phase pattern is not just a straightforward inversion of the Fourier transform. Moreover, for a given desired intensity $|U_s|^2$ in the focal plane, there is
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Figure 4.1: [Symbols explained in main text] (a) Schematic overview of the basic SLM setup, consisting of the SLM and a focusing lens. Arbitrary intensity patterns are created in the focal plane. (b) Basic dimensions of the SLM, which is subdivided into $N_x \times N_y$ pixels, giving rise to a similarly pixelated drawing area. The finite diameter of the lens limits the drawing area to the indicated circular region in the focal plane, due to vignetting.

(trivial cases excepted) no exact solution $\phi$ such that $|U_f|^2 = |U_s|^2$. Nevertheless, good approximations can be found in practice for which $|U_f|^2 \simeq |U_s|^2$. In the next sections we will explore the various ways in which a good phase pattern can be generated.

Before continuing, we will first investigate the basic dimensions of the intensity pattern in the focal plane, which are dictated by the dimensions of the SLM, which is a $L_x \times L_y$ rectangle, consisting of $N_x \times N_y$ pixels. The smallest possible spot we can make in the focal plane is obtained when the SLM is illuminated with a uniform plane wave, and all pixels impose a constant phase on the light field. Assuming an ideal lens, the intensity pattern in the focal plane then takes on the form
\[ |U(x', y', f)|^2 = \text{sinc}^2 \frac{x'}{\Delta x} \text{sinc}^2 \frac{y'}{\Delta y}, \]  

(4.5)

with \( \text{sinc} a = \frac{\sin \pi a}{\pi a} \). The size \( \Delta x \times \Delta y \) of the spot is governed by the diffraction limit \[ |a| \].

\[ \Delta x \times \Delta y = \frac{\lambda f}{L_x} \times \frac{\lambda f}{L_y}. \]  

(4.6)

Figure 4.2 shows such a spot in the \( x \) dimension. The maximum intensity of the spot at the optical axis is followed by a minimum at \( x' = \Delta x \), after which decreasing oscillations continue with consecutive minima spaced \( \Delta x \) apart. The full width at half maximum (FWHM) is also approximately equal to \( \Delta x \).

The dimensions \( \Delta x \times \Delta y \) of Eq. (4.6) form an essential length scale in the focal plane, which we will call a focal unit. As is common in domains related by a Fourier transform, the focal unit is inversely proportional to the size \( L_x \times L_y \) of the SLM.

Corresponding to the \( N_x \times N_y \) pixels in which the SLM is subdivided, we can now also subdivide the focal plane into a grid of \( N_x \times N_y \) focal units, populating an area centered around the optical axis, as illustrated in Fig. 4.1(b). Essentially, this is our ‘drawing area’ in which we can control the intensity with the SLM. Outside this area, the light field continues periodically. Increasing the number of pixels of the SLM would result in more unit cells and thus a larger drawing area, whereas the total physical size of the SLM controls the sizes of the smallest features of the light field inside the drawing area.

To see why this should be so, we first approximate the light field in the SLM plane by a series of delta peaks,

\[ U(x, y, -L + h) \simeq \sum_{k,l} \delta(x - x_k)\delta(y - y_l)U_i(x_k, y_l)e^{i\phi_{kl}}, \]  

(4.7)

with \( x_k, y_l \) the physical center coordinates of pixel \((k, l)\), and \( \phi_{kl} \) the phase delay of that pixel. This approximation is justified as long as the pixel dimensions are much smaller than the total area of the SLM, i.e., when \( N_x, N_y \gg 1 \). Ref. [220] explores the effect of properly taking into account the physical dimensions of the pixels on the intensity pattern in the focal plane.

Continuing under the approximation (4.7), and applying the Fourier transform relation in Eq. (4.4), we now evaluate the light field in the focal plane, on our grid of focal units \((x'_m, y'_n) = (m\Delta x, n\Delta y)\):

\[ U(x'_m, y'_n, f) = \sum_{k=0}^{N_x-1} \sum_{l=0}^{N_y-1} U_i(x_k, y_l)e^{i\phi_{kl}}e^{-i2\pi(km/N_x+ln/N_y)}. \]  

(4.8)

Apparently, the light fields in the SLM and focal plane are related by a discrete Fourier transform (DFT). That is, if we define the two dimensional array \( U_i \), whose elements
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**Figure 4.2:** Diffraction limited 'sinc'-spot in the focal plane, indicating the typical smallest feature size $\Delta x = \lambda f/L_x$, which serves as our unit cell in the focal plane.

\[ U_{kl}^i = U_i(x_k, y_k)e^{i\phi_{kl}} \] (4.9)

hold the values of the light field at pixel $(k, l)$ of the SLM, and similarly define the two dimensional array $U_f$, whose elements

\[ U_{nm}^f = U_f(m\Delta x, n\Delta y) \] (4.10)

hold the value of the light field evaluated at point $(n, m)$ on the grid of focal units, then, the two arrays satisfy the relation

\[ U_{nm}^f = [\text{DFT}[U_i]]_{nm} := \sum_{k=0}^{N_x-1} \sum_{l=0}^{N_y-1} U_{kl}^i e^{-i2\pi(km/N_x+ln/N_y)}. \] (4.11)

The above discussion is crucial in understanding the discreteness of the light fields due to the pixelized nature of the SLM, and uncovering the essential focal unit. In the remainder of this chapter we shall see how we can use the $N_x \times N_y$ pixels of the SLM to control the light field in the focal plane at the grid of focal units. Although much of the theory will be stated in terms of continuous light fields and continuous Fourier transforms, it should be kept in mind that whenever a calculation or simulation is performed digitally on a computer, we are always working with the arrays defined in Eqs. (4.9) and (4.10) and perform discrete Fourier transforms.

As can be seen from Eq. (4.8), the drawing area is in principle repeated periodically in the focal plane. In practice, the drawing area is ultimately limited by vignetting due to the finite lens aperture. Points in the focal plane at a distance $v$ from the optical axis receive light from rays which pass through the lens at the angle $\theta \approx \frac{v}{f}$, as illustrated in Fig. 4.1(a). Beyond a certain angle $\theta > \theta_m$, not all light rays originating from the SLM
are collected by the lens aperture. Thus, as illustrated in Fig. 4.1(b), a circular lens of diameter \( D \) imposes an approximately circular area in the focal plane of radius

\[
\begin{align*}
 r_v & \simeq \frac{f}{L} \left( \frac{1}{2} D - \frac{1}{2} \sqrt{L_x^2 + L_y^2} \right),
\end{align*}
\]

which is free from vignetting and able to receive light from the entire SLM. Outside this area, moving away from the optical axis, part of the light is blocked and the resulting intensity pattern will become degraded, until it vanishes completely.

So far, we have only discussed the dimensions of the focal plane in the \( xy \) plane, and ignored the third dimension, the \( z \) direction. The depth of focus of the intensity pattern is commonly defined by the criterion [227]

\[
\Delta z = \frac{\min(\Delta x, \Delta y)^2}{\lambda},
\]

and the intensity pattern is assumed to be sharp for all points \( z \) satisfying \(|z - f| < \Delta z\).

### 4.3 Phase synthesis by gradients

Having discussed the basic SLM setup and the corresponding geometries, we turn our attention to the calculation of phase patterns. The simplest method of generating phase patterns follows from an elementary property of the Fourier transform, holding for arbitrary functions \( f(x, y) \):

\[
\mathcal{F} \left[ e^{2\pi i(ax+by)} f(x, y) \right] (u, v) = \mathcal{F} \left[ f(x, y) \right] (u - a, v - b).
\]

In words, multiplication by an oscillating exponential, which is equivalent to adding a linear function to the phase of the light field, will spatially shift the resulting intensity pattern in the focal plane with a shift proportional to the slope of the added phase. When then phase field induced by the SLM is purely linear, the resulting diffraction pattern is thus a displaced sinc-spot [see Eq. (4.5)], illustrated schematically by Fig. 4.3(a).

It is also possible to divide the SLM into segments, and set a different linear phase in each segment. Such piecewise linear phases produce a series of sinc-spots, with dimensions dictated by the size (and shape) of the corresponding segment in the SLM plane. This way, a primitive beam splitter can be realised, as shown schematically in Fig. 4.3(b) in 1D. Fig. 4.3(c) shows the 2D phase pattern (left) and intensity in the focal plane (right). This beam splitting method works as long as the segments are large compared to the pixel size of the SLM and the wavelength of the light.

In the same spirit it is possible to realise a beam shaping phase by means of a smoothly varying phase [228], where local gradients in the phase (in the SLM plane) displace the light in the focal plane, and thus redistribute the intensity of the incident light field. This
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Figure 4.3: (a) A linear phase on the SLM causes the focus of the lens to be displaced by a distance $a$. (b) Dividing the SLM into segments with different linear phases projects multiple spots at different positions in the focal plane, realising a simple beam splitter. (c) left: 2D version of the beam splitting phase through linear segments, right: the displaced spots as observed in the focal plane, where the displacements were randomly chosen.

Technique is referred to as geometrical beam shaping.

To formalise this notion, we follow Ref. [204] and consider a function $g: \mathbb{R}^2 \rightarrow \mathbb{R}^2$ that distributes the intensity in the SLM plane, $i^{(i)} = |U_i(x, y)|^2$, onto the intensity pattern $i^{(s)} = |U_s(x', y')|^2$ in the focal plane. The function $g$ is a coordinate transform $(x, y) \rightarrow g(x, y)$, satisfying

$$
\int_B i^{(i)}(x, y)dx\,dy = \int_{g(B)} i^{(s)}(x', y')dx'\,dy',
$$

for all regions $B$, i.e., no power is lost. Figure 4.4 illustrates schematically how the coordinate transform $g$ re-distributes the intensity of an incident Gaussian onto a rectangular intensity pattern. Should we obtain such a function $g$, then a smooth phase pattern satisfying [204, 228]...
\[ \nabla \phi(x, y) = \frac{2\pi}{\lambda_f} g(x, y) \]  
(4.16)

will in principle result in a good approximation of the desired intensity \( |U(x', y', f)|^2 \simeq |U_s(x', y')|^2 \) in the focal plane. However, in practice it is nontrivial or even impossible to find transforms \( g \) that yield a soluble Eq. (4.16) for most \( U_s \).

There exists a class of problems for which we can find tractable solutions of the beam shaping problem. Namely, for separable signals of the form,

\[ I_s(x, y) = |U_s(x, y)|^2 = i_1^{(s)}(x)i_2^{(s)}(y), \]  
(4.17)

and similarly separable input intensities of the form

\[ I_i(x, y) = |U_i(x, y)|^2 = i_1^{(i)}(x)i_2^{(i)}(y), \]  
(4.18)

it is possible to find coordinate transforms \( g \) that yield a smooth solution \( \phi \) of Eq. (4.16). This separation can in fact take place in any orthogonal coordinate system, for instance, polar coordinates \((r, \theta)\) are often applicable for rotationally symmetric signals \( U_s(r, \theta) = U_s(r) \). We consider here only separable signals in the Cartesian coordinates, as specified in Eq. (4.17). We start by defining the following integrals of the input intensities:

\[ I_1^{(i)}(x) = \int_{-\infty}^{x} i_1^{(i)}(x')dx', \]  
(4.19)

\[ I_2^{(i)}(y) = \int_{-\infty}^{y} i_2^{(i)}(y')dy', \]  
(4.20)

and, similarly, two integrals of the output intensities:

\[ I_1^{(s)}(x') = \int_{-\infty}^{x'} i_1^{(s)}(x)dx, \]  
(4.21)

\[ I_2^{(s)}(y') = \int_{-\infty}^{y'} i_2^{(s)}(y)dy. \]  
(4.22)

The coordinate transform (4.15) is now also separable, \( g(x, y) = (g_1(x), g_2(y)) \), and its components can be found from

\[ g_1(x) = [I_1^{(s)}(x)]^{-1}(I_1^{(i)}(x)) \]  
(4.23)

\[ g_2(y) = [I_2^{(s)}(y)]^{-1}(I_2^{(i)}(y)). \]  
(4.24)

We can subsequently integrate Eq. (4.16) and find the phase pattern.
Figure 4.4: Schematic illustration of the geometrical beam shaping method, where an incoming Gaussian intensity $i^{(i)}$ is reshaped into a different intensity pattern $i^{(s)}$ in the focal plane, in this case a square. The coordinate transform $g$ transforms a region $B$ in the SLM plane to a region $g(B)$ in the focal plane. Figure adapted from [229].

Figure 4.5: Measurements of (a) Gaussian input intensity in the SLM plane, which is shaped into (b) a square intensity pattern. (c) The beam shaping phase imprinted on the light field by the SLM. (d) Simulation showing fringes in the square intensity pattern, due to sharp edges in the input intensity.
\[
\frac{\lambda f}{2\pi} \phi(x, y) = \int_{-\infty}^{x} g_1(x'')dx'' + \int_{-\infty}^{y} g_2(y'')dy''.
\] (4.25)

The above equations are easy to implement numerically and thus reshape the incoming (separable) light field into arbitrary (separable) output intensities. Fig. 4.5 shows an example where an incoming Gaussian [Fig. 4.5(a)] intensity is reshaped into a square [Fig. 4.5(b)]. Panel (c) of Fig. 4.5 shows the phase pattern used to shape the beam.

A disadvantage of the geometrical beam shaping method is that one needs to know the input intensity accurately. Errors in the description of the input intensity are propagated into the output intensity with similar magnitude. The deviations from a perfectly flat surface of the square intensity of Fig. 4.5(b) are in large part due to inadequate descriptions of the input intensity, i.e. the deviations of the input intensity from an ideal Gaussian, visible in Fig. 4.5(a), are directly propagating through to the focal plane intensity pattern.

Additional problems arise when the input intensity is not smooth and contains high spatial frequencies. For instance, when the input intensity is not (close to) zero at the boundaries of the SLM and thus contains non-smooth edges in the intensity, fringes will appear in the intensity pattern in the focal plane. Fig. 4.5(d) shows a (simulated) intensity pattern when an attempt is made to shape a square intensity pattern from a Gaussian beam with a very large width, with nonzero intensity at the border of the SLM. Similar problems arise when the input intensity has a partly separated support, i.e., when it consists of multiple, spatially separated, input intensities [204].

The separability of the light fields is not a strict requirement for geometric beam shaping to be possible. At the cost of increased numerical complexity, Ref. [230] details the reshaping of the incoming light field into more general (inseparable) intensity patterns by means of finite element computations. For certain cases it is also possible to compute the phase pattern analytically in closed form, e.g., see Ref. [229] and references therein.

It is important to note that phase patterns generated by geometrical beam shaping create a point-to-point relation between the intensity distribution in the focal plane and the intensity distribution in the plane of the SLM. That is, the intensity pattern in a small area in the focal plane receives light only from a corresponding small area in the SLM plane, and adjacent points in the focal plane receive light from adjacent points in the SLM plane, as illustrated in Fig. 4.4. This is to be contrasted with the method to be discussed in the next section, which generates intensity patterns for which one single point in the focal plane receives equal contributions of light from all points in the SLM plane.

### 4.4 Phase synthesis by IFTA

Geometrical beam shaping is in practice only applicable to a small set of problems. To generate an SLM phase pattern that produces an arbitrary intensity in the focal plane,
the workhorse of choice is typically the Iterative Fourier Transform Algorithm (IFTA) [204, 221]. Fig. 4.6 shows a schematic representation of this algorithm.

The desired amplitude $U_s$ in the focal plane, and the input amplitude $U_i$ of the light field in the SLM plane constitute the inputs to the algorithm, which consists of a series of iterations that (virtually) propagate the light field back and forth between the two planes. Constraints to the light field are applied in both planes during the iteration process.

To understand the working of the algorithm and the nature of the constraints, we follow Luepken et al. [222], and define the two sets

\[ H_1 = \{ G : G \in \mathbb{C}^2 \land G = |U_i|e^{i\varphi} \} \]  
\[ H_2 = \{ G' : G' \in \mathbb{C}^2 \land \mathcal{F}[G'] = |U_s|e^{i\psi} \}, \]

whose elements $G, G'$ represent light fields in the SLM plane, and $\varphi, \psi \in \mathbb{R}^2$ are arbitrary phases. The first set is defined by constraints in the SLM plane, whereas the second set is defined by constraints to the Fourier transform of $G'$, i.e., constraints in the focal plane. Obviously, finding a function $G \in H_1 \cap H_2$ would solve the phase calculation problem. Unfortunately, in general such a function does not exist. However, we can strive to find a point that minimizes the distance between the two sets.

The IFTA algorithm does precisely that, by starting with an initial point $G' = \mathcal{F}^{-1}[\exp i\psi_0] \in H_2$, and then alternatingly projecting onto the two sets. As illustrated in Fig. 4.6, the projection onto $H_1$ is effected by simply replacing the amplitude $|G'|$ by $U_i$. The projection onto $H_2$ is performed by the composite operation of first Fourier transforming, thereby switching to the focal plane, then replacing the amplitude by $U_s$, and finally an inverse Fourier transform to arrive at a point in $H_2$. Levi and Stark [231] point out that these operations indeed constitute a projection, and that the IFTA algorithm indeed minimizes the distance between the two sets. However, the sets $H_1, H_2$ are typically not convex, such that there are many local minima.

IFTA algorithms, as described above, have been known for a long time in the field of phase retrieval [232, 233], where they are often referred to as the Gerchberg-Saxton algorithm. In phase retrieval problems, one is concerned with finding the required phase, given a certain intensity pattern throughout the focal plane. However, in the field of digital holography there exist additional freedoms which greatly enhance the quality of the intensity pattern and the speed of convergence of the algorithm.

For instance, one is typically only concerned with the light intensity in a small area in the focal plane, called the window, $W$. The window is often much smaller than the entire available drawing area, and can have an arbitrary shape. Treating the amplitude of the light field outside the window as a free parameter, drastically enlarges the set of acceptable phase patterns. As first pointed out by Akahori [234], this results in better quality intensity patterns, faster convergence, and the IFTA algorithm becomes less prone to stagnation.
The Iterative Fourier Transform Algorithm (IFTA) consists of a series of (virtual) back and forth propagations between the SLM plane and the focal plane, while applying constraints to the light field in each plane.

To take advantage of the phase and amplitude freedoms in the focal plane, the IFTA is usually divided in two stages [204, 206]. The first stage exploits the phase freedom (PF) of the signal. The constraint applied in the focal plane is that of the original Gerchberg-Saxton algorithm,

\[
|g(x',y')| = \begin{cases} 
|U_s(x',y')| & (x', y' \in W) \\
0 & \text{otherwise}
\end{cases}
\]  

The amplitude freedom is not used and the amplitude outside the window \(W\) is forced to be zero. As a result, almost all the energy incident on the SLM plane is thus diffracted into the window area.

The PF stage can be repeated with several different initial phases, from which the best result with the highest final diffraction efficiency is chosen. Diffraction efficiencies in the order of 90 – 95% are not uncommon, for instance for spot array patterns that will be described in the next section.

However, the quality of the light field resulting from the PF stage is usually not satisfactory, and the PF stage is therefore followed by the Amplitude Freedom (AF) stage. During this stage, the amplitude freedom of the light field outside the signal
The Spatial Light Modulator window is also employed, allowing energy to be directed outside the window area. This allows for a better match with the desired intensity by (often dramatically) enlarging the set $H_2$ [see Eq. (4.27)], but it comes at the cost of lowering the diffraction efficiency. The constraints applied in the focal plane during the AF stage are

$$|g(x', y')| = \begin{cases} m_1|U_s(x', y')| & (x', y' \in W) \\ m_2|g'(x', y')| & \text{otherwise} \end{cases}$$

(4.29)

where $m_1, m_2$ are scalars that govern the flow of intensity between the signal window area $W$ and the remainder of the focal plane. We follow Ref. [206], and use

$$m_1 = \frac{\int_W |U_s||g'|dA}{\int_W |U_s|^2dA},$$

(4.30)

and set $m_2 = 1$. There are many possible phase patterns to which the IFTA can converge, but by first performing the PF stage separately, the algorithm is steered towards solutions with higher diffraction efficiencies.

Eq. (4.29) could of course have equally well been used to described the PF stage, setting $m_1 = 1$ and $m_2 = 0$. Modern algorithms such as the MRAF algorithm of Ref. [224] use a fixed $m_1, m_2$, with $0 < m_1 < 1$ and $m_2 = 1 - m_1$, and forego the PF stage. In practice we did not find significant differences in performance between the choice for $m_1, m_2$ of Eq. (4.30) and the MRAF algorithm. Moreover, in Sec. 4.6.4 we explore a powerful feedback method using the measured intensity patterns, which makes the precise details and performance of the underlying algorithms largely obsolete.

In practice we did not find significant differences in performance between the choice for $m_1, m_2$ of Eq. (4.30) and the MRAF algorithm. Moreover, in Sec. 4.6.4 we explore a powerful feedback method using the measured intensity patterns, which makes the precise details and performance of the underlying algorithms largely obsolete.

In conclusion, the IFTA algorithm in its form of a generalized projection algorithm is a very flexible tool, and we shall see in next sections that by changing the constraints to the sets $H_1, H_2$ it can be adapted to perform many different tasks.
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**Figure 4.7:** Example arrays $U^i, U^f$, defined in Eqs. (4.9), (4.10), used for implementing the IFTA digitally on a computer. White squares represent array elements with the value 0, shaded squares represent nonzero array elements. (a): The discretised version of the Gaussian input amplitude, (b): the discretised version representing an array of spots. The red, dashed rectangle indicates the window area $W$. 

![Diagram showing example arrays U^i and U^f](image-url)
4.5 Particular patterns

In this section we will treat the generation of phases for two important types of intensity pattern using the IFTA, namely arrays of diffraction limited spots, and patterns where large areas of smoothly varying intensity are desired.

4.5.1 Arrays of spots

The IFTA as presented in the previous Section is particularly suited for beam splitting purposes, where one wishes to split the incoming input intensity into an array of small spots at arbitrary locations. The spots implied here are small in the sense that they constitute exactly one focal unit [see Eq. (4.6)], the smallest possible spot we can make, and with all neighboring focal pixels having zero intensity. Figure 4.8 shows several examples of pairs of such spots, in one dimension. Below each intensity pattern the corresponding array $U_f$, defined in Eq. (4.10), is shown. These arrays are the one dimensional equivalents of the example shown in Fig. 4.7(a), and are the inputs to the IFTA algorithm, defining the constraints to be applied in the focal plane. Aside from the spots, it is interesting to note how the intensity pattern reaches the desired 0 value precisely at each gridpoint inside the window area, while in between the gridpoints there is still some nonzero residual.

When the spot pattern is of separable form, i.e. its intensity satisfies Eq. (4.17), then the optimal phase pattern is also separable [235], and of the form $\phi(x, y) = \phi_1(x) + \phi_2(y)$. Here, the 1D phase patterns $\phi_1(x)$ and $\phi_2(y)$ are found by applying the IFTA algorithm individually for the intensities $i_1^{(s)}(x)$ and $i_2^{(s)}(y)$, respectively, which additionally results in a significant speedup of the calculation.

In most applications the array of spots is surrounded by a dark area of zero intensity, by making the window area $W$ larger than the spot array, as illustrated in Fig. 4.7(a). This way, the uncontrolled light outside the window is directed away towards areas where it is of no importance. However, enlargement of the window area typically comes at a price in terms of diffraction efficiency, i.e. the ratio of total intensity diffracted into the window area and the total intensity in the SLM plane. This is due to the larger number of constraints the IFTA algorithm has to satisfy. The larger the distance between the outermost spots and the window boundary, the lower the diffraction efficiency becomes.

The geometric beam shaping methods of Section 4.3 have the property that the intensity incident onto a small area in the focal plane, also originates from a small area in the SLM plane. In contrast, the phase patterns generated by the IFTA algorithm (as presented in the previous section) typically have the property that all points in the SLM plane contribute approximately equally to the intensity at a given point in the focal plane. As a result, the intensity pattern in the focal plane hardly depends on the intensity in the SLM plane, and errors in the description of the incident intensity have little consequence for the focal intensity. Typically, for the amplitude constraints in the
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Figure 4.8: Theoretically expected intensity for a $2 \times 1$ array of spots, spaced (a) 0, (b) 1, (c) 2, (d) 3, and (e) 4 focal units $\Delta x$ apart. Immediately below each intensity plot is the 1D array $U^f$ [See Eq. (4.10)] used to digitally represent the signal amplitude $|U_s|$, to be compared with its 2D equivalent depicted in Fig. 4.7(b). Dashed lines indicate the boundaries of the window area $W$.

SLM plane it suffices to simply set a uniform intensity, i.e. $|U_i(x, y)| = \text{constant}$. Of course, this will not be appropriate for extremely large deviations, for instance when the incident beam diameter is much smaller than the size of the SLM. In this case, the system’s diffraction limit is altered and the sizes of the spots change accordingly.

Figure 4.9(a) shows measurements of a $10 \times 10$ spot array created using the IFTA algorithm. The input intensity consisted of a very wide Gaussian beam, covering the entire SLM. The spot sizes are therefore at the diffraction limit of $4.7\mu m \times 8.8\mu m$. The FWHM of the spots becomes of the order of the camera pixel size of $4.65\mu m \times 4.65\mu m$. Figure 4.9(b) shows an array of spots created with the same phase pattern [displayed in Fig. 4.9(c)], but with a Gaussian input intensity with a much smaller waist. The spots appear at the same positions, but are enlarged, corresponding to the enlarged diffraction
Figure 4.9: (a) Measured spot intensity for a $10 \times 10$ array of spots, spaced $12 \times 6$ focal units apart, corresponding to $\sim 61\mu m$. The input intensity illuminates the entire SLM, leading to spot sizes of one focal unit large ($4.7\mu m \times 8.8\mu m$). (b): The same spot pattern, after changing the illumination beam for a much smaller Gaussian. The spots appear on the same location, but become much larger due to the altered, effective diffraction limit of the input intensity, no longer coinciding with the focal unit. (c) Phase pattern used to generate the intensity patterns in panels (a) and (b). (d): Cross section of the top row of spots in panel (b).

limit associated with the smaller input beam.

Finally, panel (d) of Fig. 4.9 shows a histogram of the top row of spots in panel (b). The standard deviation in the total intensity contained within each spot is 6%, whereas the theoretically expected standard deviation from simulations is 0.001%. For carefully aligned and aberration corrected systems [See Sec. 4.6.2], the standard deviation in spot intensity has been observed to be as low as $1 - 2\%$.

Although the examples of spot arrays given in this section consist of regularly spaced, rectangular, arrays, there is no principal objection to using the IFTA algorithm for arrays of arbitrarily positioned spots, as long as the spots are located on positions aligned with the grid of focal units. Finally, there also exist techniques for creating spots at truly arbitrary positions, as described in Ref. [236].
Figure 4.10: (a) Two adjacent points $x_1$ and $x_2$ in the focal plane with the same amplitude and phase. The physical light field interpolates between the two points and maintains approximately the same amplitude. (b) Two adjacent points with opposite phase, where the amplitude of the interpolated light field crosses zero. (c) A phase singularity in the light field is a point where the phase assumes all values between 0 and $2\pi$ in an arbitrary small area around the singularity. (d) Measured intensity at the location of a phase singularity, showing a dip in the intensity, called a speckle. The speckle size is on the order of the diffraction limit.

4.5.2 Speckle-free intensity patterns

Creating large areas of continuous, smoothly varying intensity, requires additional modifications to the IFTA algorithm. In particular, we need to concern ourselves with constraints to the phase of the light field in the focal plane, which we have treated as an unrestricted, free parameter thus far. However, a spatially rapidly varying phase can lead to unwanted fluctuations in intensity [204, 221]. Due to the discrete pixelated nature of the SLM surface, we can only control the complex amplitude at a finite sampling grid in the focal plane, the grid points being the center points of the focal units discussed in Sec. 4.2. In between the grid points, the complex amplitude follows from interpolation. As illustrated in Figs. 4.10(a) and (b), this can cause the intensity to drop to zero when the phase difference between two adjacent grid points is close to $\pi$. Another important
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problem occurs when the phase of the light field contains a singularity, as illustrated in Fig. 4.10(c), where the phase of the light field assumes all possible values between 0 and \(2\pi\) in an arbitrarily small area. Due to the singularity the amplitude of the light field must be zero, leading to dark spots in the intensity pattern, called speckles [See Fig. 4.10(d)].

Summarising, the phase of the light field can significantly influence the intensity, and in order to create a large area of continuous smoothly varying intensity, we must strive to create a light field with a smoothly varying phase and free of phase singularities. Patterns created with the geometric beam shaping method of Sec. 4.3 typically possess these properties, but unfortunately the method is limited to a small subset of all possible patterns. In the following, we will show how the IFTA method can be modified to allow for smooth intensity patterns, free of speckles.

In order to avoid creating phase singularities in the phase of the light field in the focal plane, the first step is to initialise the IFTA algorithm with a phase \(\psi_0\) (see Fig. 4.6) which is free of phase singularities. Often, the phase pattern of a simple defocused Gaussian spot of approximately the right size is used [215, 224]. Wyrowski et al. [204] propose a more ingenious initial phase. For an arbitrary signal intensity \(I_s = |U_s|^2\), they apply the operator SepOp, defined as

\[
\tilde{I}_s(x, y) = \text{SepOp}[I_s](x, y) := \int I_s(x, y) dy' \int I_s(x', y) dx'.
\] (4.31)

The resulting signal \(\tilde{I}_s\) is separable [see Eq. 4.17], and approximates the original signal \(U_s\). Figure 4.11 provides an example of the SepOp operation. When \(U_s\) was already separable, it remains unaltered.

Using the methods of geometric beam shaping, as described in Sec. 4.3, the inverse problem is now solved of finding the phase in the focal plane that distributes the energy in \(\tilde{I}_s\) back onto the SLM plane, matching the input intensity as closely as possible. Figure 4.12 illustrates this process of inverse beam shaping. The resulting phase pattern is a very good initial guess \(\psi_0\) for starting the iterations of the IFTA algorithm [See Fig. 4.6].

With a good initial phase pattern in the focal plane, free of singularities, the possi-
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Figure 4.12: (a) The inverse beam shaping problem is solved for the phase $\psi_0$, which shapes the separable approximation of the target signal onto the input intensity when propagated backwards, from the focal plane to the SLM plane. (b) The phase $\psi_0$ is then combined with the amplitude $|U_s|$ of the desired signal, to construct a good initial guess for the IFTA, providing a rough match with the input intensity when propagated back to the SLM plane.

bility remains that the IFTA will introduce new singularities during the iterations of the algorithm. To prevent this from happening, Aagedal et. al. [237] adjusted the algorithm such that in each iteration the phase $\phi$ in the SLM plane is not completely replaced with a new estimate $\phi_{\text{new}}$, but instead it is only slightly modified:

$$\phi_{n+1} = \beta \phi_{\text{new}} + (1 - \beta) \phi_n,$$

(4.32)

where $\beta$ is a number between 0 and 1. Initially, $\beta$ is chosen very small, and is gradually increased towards 1 as the algorithm progresses and comes close to convergence. Should speckles still persist in the final intensity pattern, then it is possible to detect and annihilate pairs of nearby speckles [237].

Figures 4.13 and 4.14 show a simulation and measurement, respectively, of a complicated intensity pattern created using the above described methodology.

Finally, it should be noted that phase patterns generated using the above methodology will to some extent inherit the point-to-point relation between light in the SLM plane and the focal plane, originating from the geometric beam shaping underlying the construction of the initial phase. Similarly to the geometric beam shaping results [See e.g. Fig. 4.5], we found that using a small beam diameter for the input intensity gives much better quality intensity patterns than when the entire SLM area is being illuminated.
Figure 4.13: Simulated intensity pattern in the focal plane, for a complicated intensity pattern.

Figure 4.14: Experimentally measured intensity pattern, created using the same phase pattern as in Fig. 4.13.
4.6 Further applications and issues

When working with a SLM, a few issues arise where practice deviates from idealised theory. In this section we briefly discuss some of these issues and ways to resolve them.

4.6.1 Pixelated surface

A first point of attention is the fact that the SLM is a pixelated structure. Being a periodic structure, multiple diffraction orders reflect off its surface, leading to a loss of efficiency. In the case of the HoloEye Pluto NIR 2, the main, 0th order beam, contains approximately 60% of the total incident energy [238].

Additionally, the pixels do not cover the entire surface area of the SLM. There are areas on the SLM which do reflect (or transmit) light, but do not modulate the phase of the light field. Similarly, light might reflect from the front surface coating, without passing through the optically active layer. All this unmodulated light is focused and collected by the lens in a single spot in the focal plane and on the optical axis. This spot is commonly referred to as the zeroth order spot, and it forms a cross-like diffraction pattern. We have measured the total intensity in this spot to be 16% of all the light collected by the lens.

As a result, the region close to the optical axis is unusable for creating the desired light patterns, and parts of the focal plane away from the axis have to be used instead. Sometimes the SLM itself is used to impose a lens phase onto the light field, such that the focal length of the physical lens is altered, or even no physical lens is needed at all. The plane of interest where the intensity pattern is created then no longer coincides with the focal plane of the physical lens. In this case, the zeroth order spot will still occur in the focal plane of the physical lens (if present), and hence in the plane of interest the zeroth order spot can become defocused and enlarged, and consequently interfere with the desired intensity pattern. By placing a mask at the focal point of the physical lens the unwanted light can be removed [212].

4.6.2 Aberrations

A second issue to be discussed here is that of aberrations in the optical system, inevitable for any real-world experimental setup. Each of the myriad of components in a typical beamline introduces small or large errors to the phase of the wavefront as it propagates along. With the help of the SLM however, these aberrations can be corrected, by superimposing the exact opposite of the aberration phase pattern onto the SLM. Of course, the critical point here is to accurately determine the phase error.

A common way to characterise phase aberrations is using Zernike polynomials [239, 240]. These polynomials form a complete, orthogonal set on the unit disk, and many of them are associated with well-known aberrations such as defocus, astigmatism, coma,
etcetera. However, for describing aberrations of the SLM it is more convenient to use a set of polynomials that is orthogonal a rectangular domain. Mahajan et al. [241] use the Zernike circle polynomials as a basis, which is re-orthogonalised using the Gram-Schmidt procedure. The resulting polynomials retain the character of the corresponding Zernike circle polynomials, and we shall refer to them as the Zernike rectangle polynomials.

![Figure 4.15](image)

**Figure 4.15:** (a) Array of spots imaged by a severely aberrated lens. (b) Spot array after determining and subtracting the phase aberration. (c) Coefficients of the Zernike rectangle polynomials describing the phase aberration. (d) Total phase aberration over the surface of the SLM.

As a proof of principle of phase aberration correction, we have determined and corrected the phase aberration of a simple optical system as in Fig. 4.1 with a deliberately poorly placed and low quality lens. To determine the phase aberration we modified the IFTA algorithm as follows. Firstly, the phase pattern $\phi_0$ for an array of spots is computed and imprinted on the SLM. The resulting intensity pattern in the focal plane is measured, and used as the ‘desired’ signal $U_s$. That is, we can use the IFTA to search for the aberration phase pattern $\phi_a$ for which the phase pattern $\phi_0 + \phi_a$ produces an intensity matching the measured $U_s$. To this end, the constraints in the SLM plane as defined in Eq. (4.26) are extended such that the phase $\varphi$ is restricted to the set
\[ \varphi = \phi_0 + \sum_{j=1}^{N} c_j \varphi_j, \quad c_j \in \mathbb{R}, \]  

(4.33)

where the phases \( \varphi_j \in \mathbb{R}^2 \) are the rectangular Zernike polynomials. Fig. 4.15(a) shows the original, aberrated intensity pattern, whereas Fig. 4.15(b) shows the intensity pattern after the aberrations have been corrected using the above procedure with \( N = 11 \). Panel (c) of Fig. 4.15 shows the resulting Zernike coefficients \( c_j \), and panel (d) shows the final phase error correction as imprinted on the SLM.

A subtle point of attention is the fact that we project the phase in the SLM plane onto the Zernike polynomials at each step, rather than running the IFTA algorithm unaltered and projecting afterwards. The reason behind this is that we wish to avoid introducing phase singularities in the phase \( \phi_a \), which cannot be described correctly using the Zernike polynomials. It is found that the best way of keeping the phase \( \phi_a \) smooth is by projecting onto the subset (4.33) every step of the IFTA.

There are in fact many different ways to determine the aberrations of an optical system [240]. The above method was chosen for two reasons, namely to illustrate the versatility of the IFTA algorithm and the fact that the aberration determination can be performed in place, without having to add or modify parts of the setup. Another important alternative method for aberration correction, that shares this last feature, is the Shack-Hartmann method [240, 242]. Here, a segmented linear phase pattern is imprinted on the SLM which should result in an array of spots, as was illustrated in Fig. 4.3. Large scale phase aberrations change the positions of the spots. By measuring the displacement of each spot, one can determine the mean change in the linear slope of each segment, from which the phase aberration as a function of position on the SLM can be reconstructed. The IFTA method of Sec. 4.5.1 provides a convenient way to construct a reference array from which the displacements can be determined, since large scale phase aberrations only affect the shape of the spots created this way, but not their positions.

4.6.3 Amplitude modulation

Another point of attention to be discussed in this section is the possibility that the SLM modulates not only the phase of the light field, but also its amplitude. The Holoeye Pluto NIR II appears to suffer from this problem. Fig. 4.16 shows the measured intensity of a Gaussian light beam, after having reflected off the SLM, as a function of a uniform, constant phase imprinted on the SLM. Clearly, the amplitude of the light field changes in a sinusoidal fashion as the phase is varied.

We consider uncovering the source of the amplitude modulation to be beyond the scope of this Thesis. However, the consequences of the amplitude modulation, and possible remedies, do warrant our attention here. Due to nature of their creation, the spot arrays as discussed in Sec. 4.5.1 do not suffer any significant degradation in quality, since the spot patterns are largely independent of the input intensity pattern. However, the
Figure 4.16: Measured intensity of a Gaussian probe beam, as function of a uniform phase φ₀ imprinted on the SLM.

good geometric beam shaping of Sec. 4.3 and the patterns based on beam shaping as discussed in Sec. 4.5.2 do suffer from degradation due to amplitude modulation.

Fig. 4.17(a) shows a square intensity pattern, which contains ripple-like aberrations in its lower right quadrant. These ripples are strongly influenced by adding a uniform, constant phase to the SLM phase, and hence we attribute them to amplitude modulation. Their origin can be explained as follows. Approximating the amplitude modulation as a sinusoidal function of the phase with amplitude α [see Fig. 4.16], and denoting the phase pattern responsible for creating the square by φ, we can write for the light field $U_f$ in the focal plane:

$$U_f = \mathcal{F}[(1 + \alpha \sin \phi)e^{i\phi}]$$

$$= \mathcal{F}\left[\left(1 + \frac{\alpha}{2i}[e^{i\phi} - e^{-i\phi}]\right)e^{i\phi}\right]$$

$$= \mathcal{F}[e^{i\phi}] + \frac{\alpha}{2i} \mathcal{F}[1] + \frac{\alpha}{2i} \mathcal{F}[e^{2i\phi}].$$

The first term of Eq. (4.36) simply produces the desired square, the second term produces a bright spot at the origin, which merges with the zeroth order spot. The third term however, produces a copy of the square, twice the size of the original and with amplitude α. This fact can be understood by inspecting Eq. (4.16), showing that a phase with twice the gradient produces twice the displacement of the light field. The resulting ghost copy of the desired intensity pattern can cause significant interference effects in areas where
the two overlap. Indeed, increasing the contrast in Fig. 4.17(a) reveals the ghost copy, as shown in Fig. 4.17(b).

A straightforward solution to the problem of amplitude modulation is a simple displacement of the intensity pattern using an additional linear phase, as described in Sec. 4.3. The ghost copy will be displaced twice the distance, and for sufficiently large displacement the two will no longer overlap, removing the interference fringes.

A somewhat more advanced method employs the versatility of the IFTA, by including the amplitude modulation in the propagation step, by replacing the Fourier transform [see Fig. 4.6] as follows:

\[ \mathcal{F} \left[ U_i e^{i\phi} \right] \to \mathcal{F} \left[ U_i (1 + \alpha \sin \phi) e^{i\phi} \right]. \] (4.37)

We found that in our simulations this in principle removes the amplitude modulation effects; however, in practice the results are less satisfactory. This suggests that the model of the amplitude modulation is perhaps still incomplete, and should perhaps be amended with an additional phase modulation term. In any case, the method of displacing the target intensity pattern, combined with the feedback method outlined in the next section proved sufficient to suppress the effects of the amplitude modulation, and therefore we chose not to investigate it any further.

### 4.6.4 Feedback

The final method of improving the quality of intensity patterns is perhaps intuitively the clearest of all, namely using direct camera feedback. The intensity pattern in the focal plane is observed with a camera, and after comparing it to the desired intensity \( |U_s|^2 \),
we can make adjustments to the signal leading to an improved quality.

To see how the signal should be adjusted, and more importantly, to reveal the limitations of this method, we first introduce some new notation. For a given desired, real-valued and positive signal amplitude $U_s$, we denote the final resulting physical light field in the focal plane as a transformation of $U_s$:

$$U^m[U_s] = U_s e^{iS} + E,$$

where $U_s e^{iS}$ is the theoretically expected value of the light field after the phase pattern is calculated. We ignore *algorithmic errors*, i.e. errors due to imperfections in the calculation of the SLM phase. The term $E$ is a complex-valued error term, describing the deviation from the theoretically expected light field, due to, e.g., phase aberrations, errors in the input intensity, or interference with stray, unmodulated light.

To correct for errors, we add a term $\delta$ to the signal and compute the corresponding phase pattern. The resulting physical light field is then of the form

$$U^m[U_s + \delta] = (U_s + \delta)e^{i(S + S'(\delta))} + E + E'(\delta),$$

where we have written explicitly the change in phase $S'(\delta)$ due to introducing the correction term $\delta$, as well as the change in the error term, denoted $E'(\delta)$.

We assume that the errors we are correcting are initially small, with the correction term of similar magnitude, and that the error due to the correction term is proportionally smaller, i.e.,

$$U_s \gg |E|, \delta \gg |E'(\delta)|.$$

The intensity *measured* in the focal plane is then, to first order in $\delta, |E|$

$$|U^m[U_s + \delta]|^2 = U_s^2 + 2U_s\delta + 2Re(U_se^{i(S + S'(\delta))}E^*).$$

If the phase $S'(\delta)$ introduced by the correction term satisfies the restriction $S'(\delta) \ll 1$, then we see that we can correct the error term, to first order, by choosing

$$\delta = -\text{Re}(e^{iS}E^*) = -\frac{|U^m[U_s]|^2 - U_s^2}{2U_s},$$

where $|U^m[U_s]|^2$ can be obtained from a measurement with $\delta = 0$. This correction method is the equivalent of the intuitive notion of darkening the signal in places where it is too bright, and vice versa.

However, in practice the condition $S'(\delta) \ll 1$ is often not satisfied, and we must find a correction term $\delta$ that satisfies

$$\delta = -\text{Re}(e^{i(S + S'(\delta))}E^*),$$
Figure 4.18: (a): Measurement of a square intensity pattern suffering from various aberrations. The RMSE is \( \sim 20\% \). (b) and (c): Cross sections in the \( x \) and \( y \) direction through the center of the square.

Figure 4.19: (a): Measurement of the square intensity pattern, after 50 iterations of the feedback algorithm, showing a marked improvement in quality. The RMSE for the entire intensity pattern is 6\%, whereas the RMSE for the surface only is 1.7\% (including only points with a measured intensity value above 190). Panels (b) and (c): Cross sections in the \( x \) and \( y \) direction through the center of the square.
where it should be noted that the right hand side now depends on \( \delta \). Since we do not know the phase of \( E \), and since additionally \( S'(\delta) \) depends on \( \delta \) in a nontrivial way, we resort to an iterative procedure to solve Eq. (4.43).

Starting with an initial guess \( \delta_0 = 0 \), we compute consecutive estimates for the correction term from measurements:

\[
\delta_{n+1} = \delta_n - \beta \frac{|U_m[U_s + \delta_n]|^2 - U_s^2}{2U_s},
\]

where the scalar \( 0 < \beta \leq 1 \) is a constant to stabilise the iterative procedure. Clearly, in case the sequence \( \delta_n \) converges, we must have that the measured intensity equals the desired signal \( U_s^2 \). This method actually not only corrects the first order errors, but in fact takes care of all orders at once.

Although convergence of the sequence \( \delta_n \) is hard to prove mathematically, the method yields good results in practice. Figure 4.18 shows a square intensity pattern suffering from various errors. Figure 4.19 shows the measured pattern, after correction with the iterative scheme, using \( \beta = 0.1 \), and 50 iterations. The Root-Mean-Square Error (RMSE) of the measured intensity is 6\% for the square. However, the majority of the RMSE contributions come from the edge of the square, due to the small radius of our Gaussian input intensity. The steep edge where the square intensity pattern should start cannot be matched by the diffraction limit of our system, leading to an inevitable mismatch. If we ignore the edge and restrict our attention only to the surface of the square, by only counting pixels with an intensity above 190 camera units (the square surface lies at \( \sim 210 \)), then we obtain a RMSE as low as 1.7\%. In any case, both RMSE values are below the currently accepted state-of-the-art results of 7\% RMSE for a square intensity pattern, as reported in Ref. [215]. These authors use a more advanced IFTA algorithm and also use a feedback method. Our measured surface smoothness of the square surpasses those results by a large amount, which is already apparent from a simple visual inspection. Fig. 4.20 shows some further examples of intensity patterns obtained using the feedback method.

The limitations of the feedback method become immediately apparent from Eqs. (4.42), (4.44), where we see that the correction method only works when \( U_s \gg |E| \). If not, the correction \( \delta \) is no longer small and it would introduce new errors of the same magnitude as \( |E| \). In particular this means that the feedback method does not work in the case that there is a nonzero measured amplitude in an area where \( U_s = 0 \), e.g. as occurs in the case of misfocusing. At such a point we cannot make \( U_s + \delta < 0 \), and in those cases we must resort to the methods described in Sec. 4.6.2.
Figure 4.20: Measured intensity patterns, generated using the feedback method. (a): CQT group logo, RMSE 10%. The intensity pattern contains three speckles in the upper right corner. By tuning the parameters of the IFTA they can be removed in principle. (b): Atomtronic OR-gate, RMSE 6%. (c): Portrait of Johannes Rydberg, Swedish physicist, RMSE 4%. (d): 3D crystal of Rydberg atoms, Fig. 3.7 of this thesis, RMSE 29%. The RMSE in (d) is high due to the thin lines which are below the diffraction limit imposed by the radius of our Gaussian input intensity. In general this is a difficult pattern due to the high contrasts on small scales. To match the constraints, a lot of light is diffracted outside the window area, the latter being visible as a dark rectangle surrounding the intensity pattern.
4.7 Implementation in an ultracold atom experiment

Currently, an experimental setup exists in our CQT group that is designed to create ultracold focused ion beams \[243-245\] and ultracold, coherent electron bunches for use in electron diffraction imaging \[246\]. For these purposes, a cloud of ultracold \(^{85}\)Rb atoms is created in a magneto-optical trap (MOT), located \textit{inside} an accelerator \[247\], which itself is contained inside a vacuum chamber. The atoms in the MOT can be ionised, and the ions (or electrons) can be extracted and accelerated towards a detector, which is able to spatially resolve single ions. Typically, around \(3 \cdot 10^8\) atoms are trapped, at densities of \(10^{16} \text{atoms/m}^3\), and temperatures of 0.2mK.

The conditions of density and temperature are already sufficient to explore many-body Rydberg physics, such as the creation of crystalline structures outlined in Ch. 3, while the detection mechanism of the accelerator and charged-particle detector allow for a direct observation of such structures. In order to facilitate creation of crystalline structures of Rydberg excitations, we are planning to use the spatial light modulator in the CQT setup. Not only would precise control of the excitation laser intensity be useful to define a sharp excitation volume, but it could also be used to pre-imprint crystalline structures in the intensity pattern of the excitation laser. This will greatly reduce the state space in which the dynamics take place (see Ch. 3 Sec. 3.3), facilitating adiabatic transfer.

Additionally, the SLM would also introduce a possibility to control the lattice structure, as opposed to the structures determined by the ground state of a uniform excitation laser intensity, discussed in Ch. 3 Sec. 3.6.

Finally, light structures created by the SLM could be used to probe the Rydberg blockade, by varying the spacing in an array of spots. In the future, such experiments could be expanded to entanglement experiments and the creation of quantum gates using Rydberg atoms.

4.7.1 Experimental setup

In this section we will discuss the current setup in more detail, and outline the design constraints under which the SLM has been incorporated into the experiment.

Figure [4.21] illustrates the basic geometry of the experiment, showing the vacuum chamber with the accelerator inside, and indicating the position of the MOT. We define the positive \(z\)-axis to be the direction towards which the particles (ions or electrons) are accelerated, and the \(y\)-axis to be the physical ‘up’ direction. The detector plane is oriented perpendicular to the \(z\)-axis, located to the right of the figure, at a distance of approximately 1.5m from the MOT.

The rubidium atoms in the MOT are ionised using a two-step ionisation scheme very similar to that for creating Rydberg atoms, as discussed in Ch. 2. A red 780 nm excitation laser, which excites the rubidium atoms to the \(^5\)P\(_{3/2}\) level, enters the vacuum chamber.
Figure 4.21: Schematic overview of the experimental setup, consisting of a Magneto Optical Trap (MOT) inside an accelerator. Employing a two step excitation scheme with a red 780 nm laser, and a blue 480 nm ionisation laser, the rubidium atoms in the MOT can be ionised, after which the ions are accelerated towards a detector (to the right of the image). Also indicated is the maximum focusing angle in the \(yz\) plane, \(\theta_{yz}\), which determines the diffraction limit of the 780 nm laser in the MOT.
and accelerator from below, and impinges on the atoms from behind, pointing primarily along the \(+z\) direction. To avoid illumination of the detector, the excitation laser points a little upwards, at an angle of \(\sim 5^\circ\). The blue 480 nm laser, responsible for the Rydberg excitation and ionisation, enters from above, pointing along the \(−y\) direction.

**Figure 4.22:** Optical beamline used for shaping the excitation laser with the SLM. Lens L1 creates the desired intensity pattern outside the vacuum chamber at point X, which is then imaged by lenses L2 and L3 in the MOT.

Because of the detector geometry being oriented in the \(xy\) plane, we wish to shape the red 780 nm excitation laser with the SLM. This way, the 2D intensity patterns shaping the ion, electron, or Rydberg densities can be imaged directly onto the detector. The red 780 nm excitation laser beam passes consecutively through the vacuum chamber window and two apertures of the accelerator structure. These apertures limit the maximum focusing angle \(\theta\) of the light at the position of the MOT, and hence put a lower limit on the spot sizes we can attain. Additionally, the MOT coils limit the focusing angle further.

As a design restriction for implementing the SLM, no elements are to be added inside the vacuum chamber. The closest possible place for the final imaging lens is then directly below the vacuum window, at a propagation distance of 30 cm from the MOT. The resulting maximum focusing angle \(\theta_{yz}\) is illustrated in Fig. 4.21 where we have added a subscript to indicate that this is the maximum focusing angle in the \(yz\) plane. In the \(xz\) plane, the focusing angle is different, since in that plane the vacuum window edge and MOT coils do not play a role. Consequently, the maximum focusing angle \(\theta_{xz}\) in the \(xz\) plane is larger than that in the \(yz\) plane. Plugging in the numbers of the accelerator and vacuum window, we find that the maximum attainable focusing angle in the \(yz\) plane is \(\theta_{yz} = 6.8^\circ\), and the maximum focusing angle in the \(xz\) plane is \(\theta_{xz} = 11.6^\circ\). Since the SLM beam is rectangular, it is therefore the logical choice to orient the long axis of the beam at the vacuum window in the \(x\) direction, and the short axis in the \(z\) direction.
The final design, incorporating all of the above restrictions is presented in Fig. 4.22, showing the basic beamline following the SLM (which is drawn in transmission). The various focal lengths of the lenses in Fig. 4.22 are listed in Table 4.1. First, the excitation laser beam is expanded with a telescope (not shown) such that it covers the entire SLM. Upon reflection from the SLM, the beam is parallel and rectangular with the dimensions of the SLM. The beam propagates towards lens L1, which creates the desired intensity pattern at the point marked ‘X’. A beam splitter cube is placed directly behind the lens, allowing a Point Grey Flea 2 camera to image the intensity pattern.

From point X, the beam expands again and passes through a pair of closely spaced achromatic lenses L2 and L3. The first lens of this pair is placed such that its focal point coincides with the point X. The combination L1, L2 acts as a beam expander, enlarging the beam dimensions in order to obtain a focusing angle in the vacuum chamber close to the maximally allowed values $\theta_{xz}, \theta_{yz}$. The second achromat L3 is placed directly before the vacuum window, and focuses the beam at the position of the MOT. The two achromats together perform a 1 : 1, diffraction-limited projection of the point X inside the MOT. The theoretically attained diffraction limit inside the MOT is then $\Delta x \times \Delta y = 4.7 \mu m \times 8.8 \mu m$. The reader is referred back to Fig. 4.8 for the dimensions of spot arrays associated with this diffraction limit. The depth of focus [see Eq. (4.13)] for these parameters is $\Delta z = 28 \mu m$. The system parameters are summarised in Table 4.1.

One of the MOT trapping beams propagates partly along the same path as the excitation laser, and is mixed in by a beam splitter cube directly after the point X. After propagating through the two achromatic lenses L2 and L3, the trapping beam would no longer be parallel. To counteract this, a fourth lens L4 is added, forming a 1 : 1 beam expander together with the $L2, L3$ achromats, ensuring the trapping beam is parallel and unaltered again once it enters the vacuum system.

Finally, a custom-made software program was written to control the SLM. For compatibility with existing software, the program was written in C, in the CVI / LabWindows

### Table 4.1: System parameters for the optical beamline following the SLM.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>units</th>
</tr>
</thead>
<tbody>
<tr>
<td>focal length lens L1</td>
<td>90.0</td>
<td>mm</td>
</tr>
<tr>
<td>focal length lens L2 and L3</td>
<td>290.6</td>
<td>mm</td>
</tr>
<tr>
<td>focal length lens L4</td>
<td>15.0</td>
<td>mm</td>
</tr>
<tr>
<td>beam width at L3</td>
<td>48.4</td>
<td>mm</td>
</tr>
<tr>
<td>beam height at L3</td>
<td>25.8</td>
<td>mm</td>
</tr>
<tr>
<td>diffraction limit $\Delta x$</td>
<td>4.7</td>
<td>$\mu m$</td>
</tr>
<tr>
<td>diffraction limit $\Delta y$</td>
<td>8.8</td>
<td>$\mu m$</td>
</tr>
<tr>
<td>depth of focus $\Delta z$</td>
<td>28</td>
<td>$\mu m$</td>
</tr>
</tbody>
</table>
environment, totaling approximately 13,000 lines of source code. The software is capable of generating all the phase patterns discussed in Secs. 4.3 to 4.5 as well as perform basic tasks such as adding a focusing phase to the beam, which moves the point X in the z-direction, or adding linear phases which provide control of the point X in the x and y directions.

The software interfaces directly with a Point Grey Flea 2 camera, placed at the location shown in Fig. 4.22 imaging the intensity pattern as it should occur in the MOT. Using this camera image, the input intensity incident on the SLM can be characterised, and the software can perform aberration correction as described in Sec. 4.6.2 and use the feedback method of Sec. 4.6.4 to improve the quality of the intensity pattern in the focal plane of lens L1.

4.7.2 Spatially controlled ionisation

After implementation of the SLM as described in the previous Section, some first proof-of-principle measurements have been performed. Using the SLM to shape the red 780 nm excitation laser, the blue 480 nm laser was set to directly ionise the excited atoms, with the resulting ions extracted and detected. The blue laser is expanded to a 2D sheet of light, with a Gaussian intensity in the z direction with a waist diameter of 150 µm.

The first measurements with the SLM in place showed severe aberrations in the ion signal, which are believed to be due to phase aberrations in the optical beamline, traced back to the vacuum window and the lenses L2, L3. The vacuum window in particular has a visibly varying thickness near its edge. Additionally, it is possible that the beam is clipped at the accelerator apertures. Of course, the SLM can in principle be used to correct such phase aberrations. The light field immediately behind the vacuum window is assumed to have a phase of the form

\[
\phi(x, y) = \phi_0(x, y) + \phi_L(x, y) + \phi_{abb}(x, y),
\]

(4.45)

where \((x, y)\) are coordinates in a plane directly behind the vacuum window, and \(\phi_0\) is the phase imprinted on the light field by the SLM. The phase \(\phi_L\) is the lens phase of an ideal lens, focusing at the MOT, and \(\phi_{abb}\) are the aberrations, i.e., any deviation of the actual phase from the theoretically expected phase. In writing expression (4.45), we have assumed the thin lens approximation [205], where we assume that a ray of light incident on lens \(L_2\) at point \((x, y)\) exits the vacuum window also approximately at \((x, y)\). We assume this approximation since the lenses and the vacuum window are closely spaced, and the rays of light we will be considering are not too narrow with respect to the actual displacements.

To characterise the phase aberrations \(\phi_{abb}\), we take a series of measurements, somewhat similar to those of the Shack-Hartmann scheme [240, 242]. The phase pattern imprinted on the SLM is divided into two parts, as shown schematically in Fig. 4.23.
A small circular area of the SLM is selected and called region $I$, and imprinted with a linear phase. This linear phase is chosen such that the light incident on region $I$ hits the MOT, and produces an ion spot near the center of the detector. The remaining area of the SLM outside the circular region is called region $II$, and it is imprinted with a linear phase pattern which causes all the light incident on it to be deflected at a large angle, such that it no longer hits the MOT. Therefore, the excitation laser impinging on the MOT is reduced to a single narrow beam, originating from the circular area $I$ on the SLM.

As this narrow beam passes through the lenses $L_2, L_3$ and exits the vacuum window at point $(x_0, y_0)$, it samples only a small part of the total phase aberration, in a small area around the point $(x_0, y_0)$. To a first approximation the phase aberration in this area, and experienced by our sample ray of light, is linear:

$$\phi_{abb}(x, y) \simeq \phi_{abb}(x_0, y_0) + (\nabla \phi_{abb}(x_0, y_0)) \cdot (x - x_0, y - y_0).$$  \hfill (4.46)

Referring back to Eq. (4.14) and Fig. 4.3, the linear phase causes the spot in the focal plane of the lens to be displaced by a distance $\delta(x_0, y_0)$ [See Fig. 4.24]. The movement of the spot in the focal plane can be detected by measuring the displacement of the ion spot on the detector. This way, we can determine the gradient of the phase aberration at the point $(x_0, y_0)$. Now, by changing the position of the circular area, but not its phase pattern, we can vary the coordinates $(x_0, y_0)$, and scan the entire surface of the vacuum window. Additionally, measurements in which the ion signal has vanished allow us to identify which parts of the excitation beam are clipped by the accelerator structure.

Figs. 4.24(a) and (b) show the measured ion spot displacements in the $x$ and $y$ directions, denoted by $\delta_x, \delta_y$, respectively, as a function of the sampling coordinates $(x_0, y_0)$, expressed in coordinates on the surface of the SLM. That is, $(x_0, y_0)$ is the location on the SLM surface from which the sampling beam originated. The sampling beam diameter was set at $1\text{mm}$, and a total of $20 \times 20 = 400$ measurements are taken. There is a clear dependence of the measured spot displacement on the sampling coordinates, indicating the presence of significant aberrations. Near the edges of the sampling area no ion signal was visible, and hence no data points are shown. Light emanating from these areas of the SLM is obstructed before it reaches the MOT. The total usable area of the SLM is determined to (approximately) range between $-4\text{mm} < x_0 < +5\text{mm}$, and $-3\text{mm} < y_0 < +3\text{mm}$.

The measured displacements are then converted to the corresponding phase gradient of the aberration, which is consequently fitted with a phase aberration of the form

$$\phi_{abb}(x, y) = \sum_{j=4}^{21} c_j \phi_j(x, y),$$  \hfill (4.47)

where the phases $\phi_j$ are the rectangular Zernike polynomials discussed in Sec. 4.6.2.
The first 3 terms are omitted, as these correspond to a constant phase \((j = 1)\), or an (arbitrary) overall displacement \((j = 2, 3)\).

The phase \(\phi_{abb}\) is then subtracted from the light field by the SLM, after which the scan is repeated. The results are displayed in Figs. 4.24(c) and (d), showing that the spot displacement is significantly reduced, and hence the aberrations have been removed to a large extent. The Zernike coefficients \(c_j\) describing the aberration are shown in Fig. 4.24(e), with the total phase aberration subtracted from the light field shown in Fig. 4.24(f). The most prominent aberrations are defocus \((j = 4)\), primary astigmatism \((j = 6)\), and primary spherical aberration \((j = 11)\) \[240\].

Fig. 4.25(a) shows the ion detector image with a \(10 \times 10\) spot array, with a spot spacing of \(40\mu\text{m} \times 70\mu\text{m}\), measured before the aberration correction was applied. Fig. 4.25(b) shows the same spot pattern, after applying the aberration correction, showing a significant improvement in pattern quality. There is still some significant signal between the spots; it has not yet been investigated whether this is due to residual errors in the light pattern, or inherent problems with the ion detection method.

Finally, in Fig. 4.25(c) and (d) we show some proof-of-principle measurements, further demonstrating the capability of the SLM to shape the excitation laser at the position of the MOT. Fig. 4.25(c) shows a \(300\mu\text{m} \times 200\mu\text{m}\) CQT group logo, and Fig. 4.25(d) shows a spot array of \(20 \times 20\) spots, spaced \(20\mu\text{m} \times 40\mu\text{m}\) apart. The inset shows a magnification of a \(60\mu\text{m} \times 60\mu\text{m}\) area, showing spots which have a full diameter of approximately \(10\mu\text{m}\), spaced \(20\mu\text{m}\) in the horizontal direction. These numbers are approaching the regime where the Rydberg blockade could be observed in the future.
Figure 4.24: (a) and (b): Measured spot displacements $\delta_x, \delta_y$ in the $x$ and $y$ direction, respectively, due to a phase aberration in the optical beamline. The displacements are measured for a 1 mm wide probe beam originating at various sampling locations $(x_0, y_0)$ on the SLM surface. (c) and (d): Measured spot displacements, after the aberration has been corrected. The probe beam is blocked in the noisy edges in panels (a)-(d), leaving an unobstructed area $-4\text{mm} < x_0 < +5\text{mm}$, and $-3\text{mm} < y_0 < +3\text{mm}$. (e): Coefficients of rectangular Zernike polynomials characterising the phase aberration. (f): Total phase aberration $\phi_{\text{abb}}$ as subtracted from the light field by the SLM.
Figure 4.25: Ion detector signal. The coordinates $x$ and $y$ are expressed as the coordinates in the MOT, from which each detected ion originated. (a): $10 \times 10$ array of spots, no aberration correction. (b): Same settings as (a), after aberration correction. (c): CQT group logo. (d): $20 \times 20$ array of spots. Inset shows a zoom of a $60\mu m \times 60\mu m$ square area.
4.8 Conclusions and outlook

Spatial light modulators (SLMs) are a promising new technology, ready to cater to the ever increasing demands on the optical potentials in ultracold atom experiments. In this Chapter, we have investigated the creation of arbitrary intensity patterns in the focal plane of a lens, through modulation of the phase of the light field with a SLM.

We have reviewed existing theory for the calculation of suitable phase patterns, and performed accompanying experiments with a 1920 × 1080 pixel SLM. Two important phase-calculation methods have been discussed: geometric beam shaping and the Iterative Fourier Transform Algorithm (IFTA). Although geometric beam shaping is only applicable to a small subset of intensity patterns, it helps in building a thorough understanding of the phase design problem. More advanced intensity patterns can be calculated using variations of the IFTA algorithm, which proved to be a flexible tool, readily adapted to different demands.

In practice, several experimental issues arise, such as amplitude modulation of the SLM, and aberrations to the optical beamline. These problems were tackled using the knowledge gained in theoretically reviewing the phase calculation algorithms. Finally, a powerful feedback method was developed, capable of generating intensity patterns with higher quality than the best known results reported in literature. Such feedback methods hinge on one of the key features of SLMs, i.e., their reconfigurability.

Having mastered the creation of arbitrary intensity patterns of laser light, the SLM was incorporated in an existing experimental setup. In this setup, ultracold atoms in a magneto-optical trap are ionised and the ions can be extracted and accelerated onto a detector able to spatially resolve single ions. The SLM is demonstrated to be capable of shaping the excitation laser, and imprint spatial structures visible in the ion signal. Significant improvement of the quality of the light field is achieved, by measuring and correcting aberrations in the optical system, while making extensive use of the capabilities of the SLM.

In the future, the SLM is expected to be a valuable addition to our setup. The SLM will be most useful for exploring the Rydberg physics discussed in Chapters 2 and 3, and in particular through its ability to create well-defined and reconfigurable excitation volumes and arbitrary lattice structures. These conclusions hold more general, and we expect SLMs to become an important tool in the toolchest of the experimentalist in the field of ultracold atoms. This latter statement would be especially true when the refresh rates would reach the timescales relevant for atomic physics.
Bibliography


[238] HoloEye. *PLUTO Phase Only Spatial Light Modulators, Device Operating Instructions and SLM Application Software Instructions*.


Chapter 4.


Abstract - We present a general method for obtaining the exact static solutions and collective excitation frequencies of a trapped Bose-Einstein condensate (BEC) with dipolar atomic interactions in the Thomas-Fermi regime. The method incorporates analytic expressions for the dipolar potential of an arbitrary polynomial density profile, thereby reducing the problem of handling non-local dipolar interactions to the solution of algebraic equations.

We comprehensively map out the static solutions and excitation modes, including non-cylindrically symmetric traps, and also the case of negative scattering length where dipolar interactions stabilize an otherwise unstable condensate. The dynamical stability of the excitation modes gives insight into the onset of collapse of a dipolar BEC. We find that global collapse is consistently mediated by an anisotropic quadrupolar collective mode, although there are two trapping regimes in which the BEC is stable against quadrupole fluctuations even as the ratio of the dipolar to s-wave interactions becomes infinite. Motivated by the possibility of a fragmented condensate in a dipolar Bose gas due to the partially attractive interactions, we pay special attention to the scissors modes, which can provide a signature of superfluidity, and identify a long-range restoring force which is peculiar to dipolar systems. As part of the supporting material for this Chapter we provide the computer program used to make the calculations, including a graphical user interface.

5.1 Introduction

Since the realization of atomic Bose-Einstein condensates (BECs) in 1995 \[248–250\], there has been a surge of interest in quantum degenerate gases \[251, 252\]. Despite the diluteness of these gases, interatomic interactions play an important role in determining their properties. In the majority of experiments the dominant interactions have been isotropic and asymptotically of the van der Waals type, falling off as \(1/r^6\). At ultracold temperatures this leads to essentially pure \(s\)-wave scattering between the atoms. An exception to this rule is provided by gases that have significant dipole-dipole interactions \[253–256\]. In comparison to van der Waals type interactions, dipolar interactions are longer range and anisotropic, and this introduces rich new phenomena. For example, a series of experiments that have revealed the anisotropic nature of dipolar interactions are those on \(^{52}\text{Cr}\) BECs in an external magnetic field. These have demonstrated anisotropic expansion of the condensate depending on the direction of polarization of the atomic dipoles \[257, 258\], collapse and \(d\)-wave explosion \[259\], and an enhanced stability against collapse in flattened geometries \[260\]. Meanwhile, an experiment with \(^{39}\text{K}\) atoms occupying different sites in a 1D optical lattice has demonstrated the long-range nature of dipolar interactions in BECs through dephasing of Bloch oscillations \[255\]. Dipolar interactions have also been shown to be responsible for the formation of a spatially modulated structure of spin domains in a \(^{87}\text{Rb}\) spinor BEC \[250\].

In order to incorporate atomic interactions into the Gross-Pitaevskii theory for the condensate one should use a pseudo-potential \[251, 252\]. In the presence of both dipolar and van der Waals interactions the pseudo-potential can be written as the sum of two terms \(U(r) = U_s(r) + U_{dd}(r)\) \[261–264\], where \(r\) is the relative interatomic separation. The long-range dipolar interaction can be treated accurately within the Born approximation providing one is not close to a scattering resonance \[263, 264\]. This first-order approximation means that the effective interaction is replaced by the potential itself. This is quite different to the shorter range van der Waals interaction, for which the Born approximation is not valid at low temperatures, and where one rather uses the contact potential

\[
U_s(r) = g\delta(r).
\]

The \(s\)-wave coupling constant \(g = 4\pi\hbar^2 a_s/m\) is given in terms of the scattering length \(a_s\) and the atomic mass \(m\). For the dipolar interaction, we consider two atoms whose dipoles are aligned by an external field pointing along the direction specified by the unit vector \(\hat{e}\). The potential is then given by

\[
U_{dd}(r) = \frac{C_{dd}}{4\pi} \hat{e}_i \hat{e}_j \left(\delta_{ij} - 3\hat{r}_i \hat{r}_j\right) \frac{1}{r^3},
\]

where \(C_{dd}\) parameterizes the strength of the dipolar interactions, \(\hat{r}\) is a unit vector in the direction of \(r\), and summation over repeated indices is implied. A key figure of merit
is the ratio of the two coupling strengths, defined as [265],

$$\epsilon_{dd} = \frac{C_{dd}}{3g}.$$  \hspace{1cm} (5.3)

Dipole-dipole interactions can be either magnetic or electric in origin. To date, the dipolar interactions seen in ultracold atom experiments [253–256] have all been magnetic dipolar interactions, for which

$$C_{dd} = \mu_0 d^2,$$

where \(d\) is the magnetic dipole moment and \(\mu_0\) is the permeability of free space. In terms of the Bohr magneton \(\mu_B\), the magnetic dipole moment of a \(^{52}\text{Cr}\) atom is \(d = 6\mu_B\) giving \(\epsilon_{dd} \approx 0.16\) [253]. Although this is 36 times larger than the typical value of \(\epsilon_{dd}\) found in the alkalis, it is still small. Thus, unless the system is in a configuration that makes it particularly sensitive [255], and/or is specially prepared [256], the magnetic dipolar interactions in the atomic gases made so far tend to be masked by stronger \(s\)-wave interactions. In order to make dipolar interactions in BECs more visible, the Stuttgart group have succeeded in implementing magnetic Feshbach resonances [266, 267] in \(^{52}\text{Cr}\) [260]. These allow \(g\) to be tuned from positive to negative and even to zero. Moreover, the sign and amplitude of the effective value of \(C_{dd}\) can also be tuned by rapidly rotating the external polarizing field [265]. Polar molecules can have huge electric dipole moments and these systems are now close to reaching degeneracy [268–273]. By appropriately tuning an external electric field a large degree of control can be exerted over these systems [273]. Combined with what has already been achieved in \(^{52}\text{Cr}\), a large parameter space of interactions can now be realistically explored in dipolar BECs.

**Figure 5.1:** Schematic illustration of the basic collective modes under consideration: the dipole mode \(D\) (shown here in the \(x\)-direction \(D_x\)), scissors mode \(Sc\) (shown here in \(x - z\) plane \(Sc_{xz}\)), the monopole mode \(M\) and the quadrupole modes \(Q_1\) and \(Q_2\). These modes are discussed in more detail in Section 5.3.
Chapter 5.

The ground state of a trapped dipolar BEC has already been investigated theoretically by a number of authors, e.g. [261–264, 274–281], with most studies focusing on the regime where $g \geq 0$ and $C_{dd} > 0$. The presence of dipolar interactions was widely predicted to lead to certain distinctive effects, some of which have recently been seen experimentally. For example, if the dipoles are aligned in the $z$-direction, then a condensate will elongate along $z$ and become more “cigar”-shaped, i.e. undergo magnetostriction, in order to benefit energetically from the attractive end-to-end interaction of dipoles. As $\epsilon_{dd}$ is increased, for example by reducing $g$ with a Feshbach resonance, the BEC eventually becomes unstable to collapse, and this striking behavior has been realized in the experiment [259]. Conversely, a condensate that is flattened by strong trapping along $z$ will be mostly composed of repulsive side-by-side dipoles and so this “pancake”-shaped geometry is more stable, as confirmed experimentally [260]. In the limit that $\epsilon_{dd}$ becomes large, but the BEC remains in the pancake configuration due to tight trapping, remarkable density wave structures have been predicted for certain regions of parameter space close to the collapse threshold [278–280].

In this Chapter we work in the Thomas-Fermi (TF) regime, which is of rather general interest because it is formally equivalent to the hydrodynamic regime of zero-temperature superfluids [282]. The TF regime may be viewed as the semiclassical approximation to the full Gross-Pitaevskii theory. A stationary condensate enters the TF regime when the zero-point kinetic energy of the atoms due to the confinement by the trap becomes negligible in comparison to the total interaction and trapping energies. For BECs with repulsive interactions in a harmonic trap this generally occurs in the large $N$ limit, where $N$ is the number of atoms. However, for dipolar BECs the picture is considerably complicated by the partially attractive and partially repulsive nature of the interactions. The question of the validity of the TF regime in static dipolar BECs has been addressed in [283]. The validity of results for collective excitations, which is the major theme of this Chapter, rests primarily on the validity of the underlying static solution about which they are a perturbation (we are interested in small amplitude excitations here). Theoretical results [281] demonstrate that if the static solution closely approximates the true Gross-Pitaevskii solution then the TF equations of motion (the superfluid hydrodynamic equations) give a remarkably robust account of the dynamics. For example, excellent agreement was observed between the TF model and full simulations of the Gross-Pitaevskii equation for a dipolar BEC even for extreme perturbations to the condensate, including the initial dynamics of collapse. Thus, for the small perturbations considered here we can be confident of consistent results, providing the underlying static solution is valid.

The TF regime is theoretically simpler to handle than the full Gross-Pitaevskii theory, thereby facilitating analytical results. For example, under harmonic trapping it can be shown that the exact density profile of a dipolar condensate in the TF regime is an inverted parabola [275, 276], similar to the usual $s$-wave case but distorted by the magnetostriction. Furthermore, the stability of the ground state to collapse can be estimated simply in the TF regime and reasonable agreement with experiment has been
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reported [260]. Rotational instabilities of dipolar BECs are also amenable to analysis in the TF regime [284, 285]. The current Chapter builds on these earlier works by applying the exact results available in the TF regime to collective excitations.

The excited states of a BEC can be accurately calculated within the TF regime provided they are of sufficiently long wavelength. The most basic collective excitations of a trapped BEC are the dipole (centre-of-mass), monopole (breathing), quadrupole and scissors modes, illustrated schematically in Fig. 5.1. Their characterization offers important opportunities to measure interaction effects, test theoretical models, and even detect weak forces [286]. Specifically, the scissors mode provides an important test for superfluidity [287, 290], while the quadrupole mode plays a key role in the onset of vortex nucleation in rotating condensates [284, 285, 291, 295]. An instability of the quadrupole mode is also thought to be the mechanism by which collapse of dipolar BECs proceeds when it occurs globally [263, 274, 281, 296] (rather than locally [281]). While the collective modes of a dipolar BEC have been studied previously [274, 275, 277, 297–301], key issues remain at large, for example, the regimes of $C_{dd} < 0$ and $g < 0$, and the behaviour of the scissors modes. This provides the motivation for the current work.

In this Chapter we present a general and accessible methodology for determining the static solutions and excitation frequencies of trapped dipolar BECs in the TF limit. We explore the static solutions and the low-lying collective excitations throughout a large and experimentally relevant parameter space, including positive and negative dipolar couplings $C_{dd}$, positive and negative $s$-wave interactions $g$, and cylindrically and non-cylindrically symmetric systems. Moreover, our approach enables us to unambiguously identify the modes responsible for global collapse of the condensate. We would like to point out that there is a freely available MATLAB implementation of the calculations presented in this Chapter, complete with a graphical user interface, which can be found in the supporting material [302].

Section 5.2 is devoted to the static solutions of the system. Beginning with the underlying Gross-Pitaevskii theory for the condensate mean-field, we make the TF approximation and outline the methodology for deriving the TF static solutions. We then use it to map out the static solutions with cylindrical symmetry, for both repulsive and attractive $s$-wave interactions, and then present an example case of the static solutions in a non-cylindrically-symmetric geometry. We compare to recent experimental observations where possible.

In Section 5.3 we present our methodology for deriving the excitation frequencies of a dipolar BEC. This is an adaptation of the method that Sinha and Castin applied to standard $s$-wave condensates [292] where one considers perturbations around the static solutions (derived in Section 5.2) and employs linearized equations of motion for these perturbations. At the heart of our approach is the exact calculation of the dipolar potential of a heterogeneous ellipsoidal BEC, performed by employing results from gravitational potential theory known in astrophysics [303–308] and detailed in appendix 5.A.

In Section 5.4 we apply this method to calculate the frequencies of the important
low-lying modes of the system, namely the monopole, dipole, quadrupole and scissors modes, for cylindrically-symmetric traps. We show how these frequencies vary with the key parameters of the system, $\epsilon_{dd}$ and trap ratio $\gamma$, and give physical explanations for our observations. In Section 5.5 we extend our analysis to non-cylindrically-symmetric traps. Although the parameter space of such systems is very large, we present pertinent examples. An important feature of non-cylindrically-symmetric ground states is that they support a family of scissors modes which can be employed as a test for superfluidity. As such, in Section 5.6 we focus on these scissors modes and show how they vary with key parameters. Finally, in Section 5.7 we summarise our findings.

Appendix 5.A outlines the method by which we calculate dipolar potentials due to arbitrary polynomial density distributions of atoms. This is the main technical advance of this work over our previous papers, which were limited to the dipolar potentials associated with strictly paraboloidal density distributions, i.e. those of the same symmetry class as the static solution.

5.2 Static solutions

5.2.1 Methodology for obtaining static solutions

At zero temperature the condensate is well-described by a mean-field order parameter, or “wave function”, $\psi(r, t)$. This defines an atomic density distribution via $n(r, t) = |\psi(r, t)|^2$. Static solutions, denoted by $\tilde{\psi}(r)$, satisfy the time-independent Gross-Pitaevskii equation (GPE) given by

$$\left[ -\frac{\hbar^2}{2m} \nabla^2 + V(r) + \Phi_{dd}(r) + g|\tilde{\psi}(r)|^2 \right] \tilde{\psi}(r) = \mu \tilde{\psi}(r) \tag{5.4}$$

where $\mu$ is the chemical potential of the system. The external potential $V(r)$ is typically harmonic with the general form,

$$V(r) = \frac{1}{2} m \omega_{\perp}^2 \left[ (1 - \epsilon) x^2 + (1 + \epsilon) y^2 + \gamma^2 z^2 \right]. \tag{5.5}$$

Here $\omega_{\perp}$ is the average trap frequency in the $x - y$ plane and the trap aspect ratio $\gamma = \omega_z / \omega_{\perp}$ defines the trapping in the axial ($z$) direction. The trap ellipticity $\epsilon$ in the $x - y$ plane defines the transverse trap frequencies via $\omega_x = \sqrt{1 - \epsilon} \omega_{\perp}$ and $\omega_y = \sqrt{1 + \epsilon} \omega_{\perp}$. When $\epsilon = 0$ the trap is cylindrically symmetric.

The $\Phi_{dd}$-term in Eq. (5.4) is the mean-field potential arising from the dipolar interactions

$$\Phi_{dd}(r) = \int n(r') U_{dd}(r - r') d^3r'. \tag{5.6}$$

This term is a non-local functional of the density and is the source of the difficulties
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associated with theoretical treatments of dipolar BECs: it turns the GPE into an integro-differential equation. A key feature of the approach taken by us in this Chapter is to calculate this term analytically. To this end we express the dipolar mean-field in terms of a fictitious electrostatic potential

$$\Phi_{dd}(r) = -C_{dd} \left( \frac{\partial^2}{\partial z^2} \phi(r) + \frac{1}{3} n(r) \right), \tag{5.7}$$

where

$$\phi(r) = \frac{1}{4\pi} \int \frac{n(r')}{|r - r'|} d^3r'. \tag{5.8}$$

\(\phi(r)\) satisfies Poisson’s equation \(\nabla^2 \phi(r) = -n(r)\). Note that in (5.7) we have taken the dipoles to be aligned along the \(z\)-direction. The term \(n(r)/3\) appearing on the right hand side of (5.7) cancels the Dirac delta-function which arises in the \(\partial^2 \phi(r) \partial z^2\) term \([309, 310]\). This means that \(\Phi_{dd}\) includes only the long-range \((r^{-3})\) part of the dipolar interaction, exactly as written in Equation (5.2).

We assume the TF approximation where the zero-point kinetic energy of the atoms in the trap is neglected. Dropping the relevant \(\nabla^2\)-term in Eq. (5.4) leads to

$$V(r) + \Phi_{dd}(r) + gn(r) = \mu. \tag{5.9}$$

For an \(s\)-wave BEC under harmonic trapping, the exact density profile in the TF approximation is known to be an inverted parabola \([252]\) with the general form,

$$n(r) = n_0 \left( 1 - \frac{x^2}{R_x^2} - \frac{y^2}{R_y^2} - \frac{z^2}{R_z^2} \right) \quad \text{for} \quad n(r) \geq 0 \tag{5.10}$$

where \(n_0 = 15N/(8\pi R_x R_y R_z)\) is the central density, and \(R_x, R_y, \text{and} \ R_z\) are the condensate radii. In order to obtain the dipolar potential arising from this density distribution, one must find the corresponding electrostatic potential of Eq. (5.8). References \([275, 276]\) follow this procedure, and arrive at the remarkable conclusion that the dipolar potential \(\Phi_{dd}\) is also parabolic. Therefore, a parabolic density profile is also an exact solution of the time-independent TF Equation (5.9) even in the presence of dipolar interactions. In Section 5.3 and Appendix 5.A we point out that this result can be extended using results from 19th century gravitational potential theory \([303, 304, 306]\) to arbitrary polynomial densities yielding polynomial dipolar potentials of the same degree. For the parabolic density profile at hand, the internal dipolar potential is given by \([276, 284]\)

$$\Phi_{dd}(r) = -g_{dd}n_0\kappa_x\kappa_y \times \left[ \beta_{001} - \left( \beta_{101}x^2 + \beta_{011}y^2 + 3\beta_{002}z^2 \right) R_z^{-2} \right] \tag{5.11}$$

where \(\kappa_x = R_x/R_z\) and \(\kappa_y = R_y/R_z\) are the aspect ratios of the condensate, and
\[ \beta_{ijk} = \int_0^\infty \frac{ds}{(\kappa_x^2 + s)^{i+\frac{1}{2}}(\kappa_y^2 + s)^{j+\frac{1}{2}}(1 + s)^{k+\frac{1}{2}}}, \quad (5.12) \]

where \( i, j, k \) are integers. Explicit expressions for \( \beta_{001}, \beta_{101}, \beta_{011}, \) and \( \beta_{002} \) can be given in terms of elliptic integrals. Note that for a cylindrically-symmetric trap \( \epsilon = 0 \), the static condensate profile is also cylindrically-symmetric with aspect ratio \( \kappa_x = \kappa_y =: \kappa \). In the cylindrically-symmetric case the integrals \( \beta_{ijk} \) of Eq. (5.12) can be evaluated in terms of the \( _2F_1 \) Gauss hypergeometric function [311, 312] for any \( i, j, k \)

\[ \beta_{ijk} = 2 \frac{_{2F_1} \left( k + \frac{1}{2}, 1; i + j + k + \frac{3}{2}, 1 - \kappa^2 \right)}{(1 + 2i + 2j + 2k)\kappa^{2(i+j)}}. \quad (5.13) \]

For the parabolic density profile of Eq. (5.10), the TF Eq. (5.9) becomes

\[ \mu = 3g \epsilon_{dd} \frac{n_0 \kappa_x \kappa_y}{2R_z^2} \left[ R_z^2 \beta_{001} - \beta_{101} x^2 - \beta_{011} y^2 - 3\beta_{002} z^2 \right] \]

\[ + \ V(r) + (1 - \epsilon_{dd}) \frac{g n_0}{R_z^2} \left( R_z^2 - \frac{x^2}{\kappa_x^2} - \frac{y^2}{\kappa_y^2} - z^2 \right). \quad (5.14) \]

Inspection of the coefficients of \( x^2, y^2 \) and \( z^2 \) leads to three self-consistency relations, given by

\[ \kappa_x^2 = \frac{\omega_x^2}{\omega_x^2} \frac{1 + \epsilon_{dd} \left( \frac{3}{2} \kappa_y^2 \beta_{011} - 1 \right)}{1 - \epsilon_{dd} \left( 1 - \frac{g \kappa_x \kappa_y}{2} \beta_{002} \right)}, \quad (5.15) \]

\[ \kappa_y^2 = \frac{\omega_y^2}{\omega_y^2} \frac{1 + \epsilon_{dd} \left( \frac{3}{2} \kappa_x^2 \beta_{101} - 1 \right)}{1 - \epsilon_{dd} \left( 1 - \frac{g \kappa_x \kappa_y}{2} \beta_{002} \right)}, \quad (5.16) \]

\[ R_z^2 = \frac{2 g n_0}{m \omega_z^2} \left[ 1 - \epsilon_{dd} \left( 1 - \frac{9 \kappa_x \kappa_y}{2} \beta_{002} \right) \right]. \quad (5.17) \]

Solving Eqs. (5.15)-(5.17) gives the exact static solutions of the system in the TF regime.

The energetic stability of the condensate is determined by the TF energy functional

\[ E = \int \left( V(r) + \frac{1}{2} \Phi_{dd}(r) + \frac{1}{2} g n(r) \right) n(r) d^3r. \quad (5.18) \]

Inserting the parabolic density profile (5.10) yields an energy landscape
Collective Excitations of Dipolar Bose-Einstein Condensates

\[ E = \frac{15N^2g}{28\pi\kappa_x\kappa_y R_z^3} \left[ (1 - \epsilon_{dd}) \right. \\
+ \frac{3}{8}\kappa_x\kappa_y\epsilon_{dd} \left( 7\beta_{001} - 3\beta_{002} - \kappa_x^2\beta_{101} - \kappa_y^2\beta_{011} \right) \right] \\
+ \frac{N}{14}mR_z^2 \left( \kappa_x^2\omega_x^2 + \kappa_y^2\omega_y^2 + \gamma^2 \right). \]  
(5.19)

Static solutions correspond to stationary points in the energy landscape. If the stationary point is a local minimum in the energy landscape, it corresponds to a physically stable solution. However, if the stationary point is a maximum or a saddle point, the corresponding solution will be energetically unstable. The nature of the stationary point can be determined by performing a second derivative test on Eq. (5.19) with respect to the variables \( \kappa_x, \kappa_y, \) and \( R_z \). This leads to 6 lengthy equations that will not be presented here. Note that this only determines whether the stationary point is a local minimum within the class of parabolic density profiles. In other words, with the three variables \( \kappa_x, \kappa_y, \) and \( R_z \) we are only able to determine stability against “scaling” fluctuations, so named because they correspond to a rescaling of the static solution \[313, 314\]. However, the class of scaling fluctuations includes important low-lying shape oscillations such as the monopole and quadrupole modes. Although higher order (beyond quadrupole) modes can become unstable in certain regimes, as a criterion of stability we will use the local minima of (5.19). This assumption is supported by the recent experiments by Koch et al. \[260\], where dipolar BECs were produced with \( \epsilon_{dd} > 1 \) that were stable over significant time-scales.

### 5.2.2 Cylindrically-symmetric static solutions for \( g > 0 \), and the critical trap ratios \( \gamma^+_{\text{crit}} \) and \( \gamma^-_{\text{crit}} \)

We have obtained the static solutions for a cylindrically-symmetric BEC by solving Eqs. (5.15) to (5.17) numerically. The solutions behave differently depending on whether the \( s \)-wave interactions are repulsive or attractive. We begin by considering the \( g > 0 \) case. The ensuing static solutions, characterised by their aspect ratio \( \kappa \), are presented in Fig. 5.2 as a function of \( \epsilon_{dd} \) with each line representing a different trap ratio \( \gamma \). While the TF solutions in the regime \( \epsilon_{dd} > 0 \) have been discussed previously \[273, 276\], the regime of \( \epsilon_{dd} < 0 \) has not been studied. Be aware that when we fix \( g > 0 \), the regime \( \epsilon_{dd} < 0 \) (left hand side of Fig. 5.2) corresponds to \( C_{dd} < 0 \) where the dipolar interaction is reversed, repelling along \( z \) and attracting in the transverse direction. This can be achieved by rapid rotation of the field aligning the dipoles about the \( z \)-axis \[265\].

Before we examine the question of stability, let us first interpret the structure of the solutions shown in Fig. 5.2. Imagine an experiment in which the magnitude of \( \epsilon_{dd} \) is slowly increased from zero. At \( \epsilon_{dd} = 0 \) we have purely \( s \)-wave interactions and all solutions have the same aspect ratio as the trap, i.e. \( \kappa = \gamma \). As \( \epsilon_{dd} \) is increased above zero \( \kappa \) decreases...
so that $\kappa < \gamma$ for all solutions. This is because standard magnetostriction causes dipolar BECs to be more cigar-shaped than their $s$-wave counterparts. Conversely, if $\epsilon_{dd}$ is made negative then $\kappa$ increases so that $\kappa > \gamma$ for all solutions. This is because when $C_{dd} < 0$ we have non-standard (reversed) magnetostriction which leads to a more pancake shaped BEC.

Consider now the stability of the solutions, beginning with the range $-1/2 < \epsilon_{dd} < 1$ [white region in Fig. (5.2)]. We find that the energy landscape (5.19) has only one stationary point, namely a global energy minimum, and it occurs at finite values of the radii $R_x (= R_y)$, and $R_z$. This global minimum persists for all trap ratios (outside of the range $-1/2 < \epsilon_{dd} < 1$ the existence of stable static solutions depends on $\gamma$). Thus, in the range $-1/2 < \epsilon_{dd} < 1$ the static TF solution is stable against scaling fluctuations. Other classes of perturbation could lead to instability, but there is good reason to believe that in this range the parabolic solution is stable against these too. Take, for example, phonons, i.e. local density perturbations. These have a characteristic that can be considered opposite to the global motion involved in scaling oscillations. The local character of phonons means that considerable insight can be gained from the limiting case of a homogeneous dipolar condensate. The energy of a plane wave perturbation (phonon) with momentum $p$ is given by the Bogoliubov energy $E_B$ [261],

$$E_B^2 = \left(\frac{p^2}{2m}\right)^2 + 2gn \left\{1 + \epsilon_{dd} (3 \cos^2 \theta - 1)\right\} \frac{p^2}{2m},$$

(5.20)

where $\theta$ is the angle between the momentum of the phonon and the polarization direction. The perturbation evolves as $\sim \exp(iE_Bt/\hbar)$ and so when $E_B^2 < 0$ the perturbations grow exponentially, signifying a dynamical instability. Dynamical stability requires that $E_B^2 > 0$ which, for $g > 0$, corresponds to the requirement that $[1 + \epsilon_{dd} (3 \cos^2 \theta - 1)] \geq 0$ in Eq. (5.20). This leads once again to precisely the stability condition $-1/2 < \epsilon_{dd} < 1$.

Outside of the regime $-1/2 < \epsilon_{dd} < 1$ the global energy minimum of the TF system is a collapsed state where at least one of the radii is zero, just like in the uniform dipolar BEC case. However, unlike the uniform case, in the presence of a trap the energy functional can also support a local energy minimum corresponding to a metastable solution [light gray region in Fig. 5.2]. The existence of a metastable solution means there must also be a saddle point connecting the metastable solution to the collapsed state and this is indicated by the dark gray region in Fig. (5.2).

In general, the occurrence of metastable solutions depends sensitively on $\epsilon_{dd}$ and $\gamma$. Remarkably, however, there are two critical trap ratios, $\gamma^+_{\text{crit}} = 5.17$ and $\gamma^-_{\text{crit}} = 0.19$, beyond which the BEC is stable against scaling fluctuations even as the strength of the dipolar interactions becomes infinite. First consider $\epsilon_{dd} > 1$, for which there is a susceptibility for collapse towards an infinitely narrow line of end-to-end dipoles ($R_x = R_y \to 0$). Providing $\gamma > \gamma^+_{\text{crit}}$, i.e. if the trap is pancake enough, condensate solutions metastable against scaling fluctuations persist even as $\epsilon_{dd} \to \infty$ [262, 263, 276]. Referring
Figure 5.2: Aspect ratio $\kappa$ of the $g > 0$ cylindrically symmetric static solutions as a function of $\epsilon_{dd}$ according to Eqs. (5.15)-(5.17). Note that $\epsilon_{dd} < 0$ corresponds to $C_{dd} < 0$. The solid lines indicate the static solutions for specific trap ratios $\gamma$ which are equally spaced on a logarithmic scale in the range $\gamma = [0.1, 10]$. The parameter space of global, metastable and unstable solutions is denoted by white, light gray and dark gray regions, respectively.

These curves are located in the upper right hand portion of the plot and asymptote to horizontal lines as $\epsilon_{dd}$ is increased (see Fig. 3 in [276] for a plot which extends $\epsilon_{dd}$ to much higher values than shown here so that this behavior is clearer). However, if the trap is not pancake-shaped enough, i.e. $\gamma < \gamma_{\text{crit}}^+$, then as $\epsilon_{dd}$ is increased from zero the local energy minimum eventually disappears and no stable solutions exist. Referring again to Fig. 5.2 these are the curves that turn over as $\epsilon_{dd}$ is increased, and in so doing enter the dark gray region. Second, consider $\epsilon_{dd} < -0.5$, for which the system is susceptible to collapse into an infinitely thin pancake of side-by-side dipoles ($R_z \to 0$). If the trap is sufficiently cigar-like with $\gamma < \gamma_{\text{crit}}^-$ collapse via scaling oscillations is suppressed even in the limit $\epsilon_{dd} \to -\infty$. These curves are located in the lower left hand portion of Fig. 5.2 and asymptote to horizontal lines. However, if the trap is not cigar-shaped enough, i.e. $\gamma > \gamma_{\text{crit}}^-$, then for sufficiently large and negative $\epsilon_{dd}$ the metastable solution disappears, bending upwards to enter the dark gray region on the left hand portion of Fig. 5.2 and the system becomes unstable to collapse.

In a recent experiment Lahaye et al. [259] measured the aspect ratio of the dipolar condensate over the range $0 \lesssim \epsilon_{dd} \lesssim 1$, using a Feshbach resonance to tune $g$, and found very good agreement with the TF predictions. Similarly, Koch et al. [260] observed the
threshold for collapse in a $\gamma = 1$ system to be $\epsilon_{dd} \approx 1.1$, in excellent agreement with the TF prediction of $\epsilon_{dd} = 1.06$. Using various trap ratios, it was also found that collapse became suppressed in flattened geometries and the critical trap ratio was observed to exist in the range $\gamma_{crit}^+ \approx 5 - 10$, which is in qualitative agreement with the TF predictions.

5.2.3 Cylindrically-symmetric static solutions for $g < 0$, and the nature of dipolar stabilization

We now consider the case of attractive $s$-wave interactions $g < 0$. Negative values of $g$ can be achieved using a Feshbach resonance. The static solutions are presented in Fig. 5.3. Be aware that because $g < 0$, $\epsilon_{dd} < 0$ ($\epsilon_{dd} > 0$) now corresponds to $C_{dd} > 0$ ($C_{dd} < 0$). The stability diagram differs greatly from the $g > 0$ case and, in particular, no TF solutions exist in the range $-1/2 < \epsilon_{dd} < 1$. Nevertheless, TF solutions can exist outside of this range in regions of parameter space determined by the two critical trap ratios $\gamma_{crit}$ and $\gamma_{crit}^+$ introduced in the previous section. We find that for $\epsilon_{dd} > 0$ solutions only exist for significantly cigar-shaped geometries with $\gamma < \gamma_{crit} = 0.19$, while for $\epsilon_{dd} < 0$ solutions only exist for significantly pancake-shaped geometries with $\gamma > \gamma_{crit}^+ = 5.17$. Furthermore, the attractive $s$-wave interactions always cause the global minimum to be a collapsed state. This means that static TF solutions are only ever metastable (light gray region in Fig. 5.3) when $g < 0$.

Although our TF model predicts that no solutions exist for $-1/2 < \epsilon_{dd} < 1$, it is well-known that stable condensates with attractive purely $s$-wave interactions (corresponding to $\epsilon_{dd} = 0$) can be made in the laboratory [315]. The zero-point energy of the atoms (ignored in the TF model) induced by the trapping potential stabilises the condensate up to a critical number of atoms $N_{crit} = k a_{ho}/|a_s|$, where $a_{ho} = \sqrt{\hbar/(m \omega)}$ is the harmonic oscillator length obtained from the mean trapping frequency $\omega = (\omega_x \omega_y \omega_z)^{1/3}$, and $k \approx 1/2$ is a constant [316]. One can expect, therefore, that for a finite number of atoms the presence of zero-point energy enhances the stability of the condensate beyond the TF solutions [281]. This will extend somewhat the light gray regions in both Figures 2 and 3 into the dark gray regions, the amount depending upon $N_{crit}/N$. The TF regime corresponds to the $N \to \infty$ limit and so is in a sense a universal regime that can always be realized with a large enough condensate.

Insight into the TF stability diagram shown in Fig. 5.3 can once again be gleaned from the Bogoliubov spectrum for a uniform system given by Eq. (5.20), this time with $g < 0$. Firstly, for the purely $s$-wave case we recall the well-known result [252] that a homogeneous attractive BEC is always unstable to collapse. With dipolar interactions the uniform system is stable to axial perturbations ($\theta = 0$) for $\epsilon_{dd} < -1/2$ and to radial perturbations ($\theta = \pi/2$) for $\epsilon_{dd} > 1$. This is the exact opposite of the $g > 0$ case and corroborates the lack of solutions given by the TF equations for $-1/2 < \epsilon_{dd} < 1$. Of course, $\epsilon_{dd} < -1/2$ and $\epsilon_{dd} > 1$ cannot be simultaneously satisfied and so a uniform dipolar system with $g < 0$ is always unstable. However, when the system is trapped
Figure 5.3: Aspect ratio $\kappa$ of the $g < 0$ cylindrically-symmetric static solutions as a function of $\epsilon_{dd}$. Note that the regime of $\epsilon_{dd} > 0$ corresponds to $C_{dd} < 0$. The solid lines denote static solutions for specific trap ratios $\gamma$, equally spaced on a logarithmic scale in the ranges $\gamma = [0.010, \gamma_{\text{crit}}]$ (lower right set of curves) and $\gamma = [\gamma_{\text{crit}}^+, 100]$ (upper left set of curves). Arrows indicate direction of increasing $\gamma$. The black lines in the light gray regions correspond to local minima (metastable) points in the energy landscape, while the red lines in the dark gray region correspond to saddle (unstable) points. At the extreme left and right hand sides of the figure the stable solutions become horizontal lines as they tend asymptotically to the trap aspect ratio $\kappa \rightarrow \gamma$ (see text).

The condensate can be stabilized even in the TF regime. The mean dipolar interaction depends on the condensate shape and can become net repulsive in cigar-shaped systems when $\epsilon_{dd} > 0$ (for which $C_{dd} < 0$), and in pancake-shaped systems when $\epsilon_{dd} < 0$ (for which $C_{dd} > 0$). Remarkably, in these cases it is the dipolar interactions that stabilize the BEC against the attractive $s$-wave interactions and lead to the regions of metastable static solutions observed in Fig. 5.3. Without the dipolar interactions the BEC would collapse.

The metastable TF solutions shown in Fig. 5.3 have a counter-intuitive dependence upon $\epsilon_{dd}$. Take, for example, the family of metastable solutions (black curves) in the lower right hand portion of the figure. We see that as $\epsilon_{dd}$ increases $\kappa$ decreases (condensate becomes more cigar-shaped). This is in contradiction to what one might naively expect because on this side of the figure $C_{dd} < 0$, and so the dipolar interaction has an energetic preference for dipoles sitting side-by-side not end-to-end! In order to appreciate what is happening in this region of Fig. 5.3 observe that for each value of $\epsilon_{dd}$ there is a
critical value of the condensate aspect ratio $κ$ below which the system is metastable, and above which it is unstable. As $ϵ_{dd}$ is increased from this point the net repulsive dipolar interactions favor elongating the BEC so that atoms sit further from each other, thereby lowering the interaction energy and decreasing $κ$. In the limit $κ \to 0$ one can show that the dipolar mean-field potential tends to $Φ_{dd} = −gϵ_{dd}n(r)$ \[^{283}\], i.e. it behaves like a spherically-symmetric contact interaction which is repulsive when $g < 0$ and $ϵ_{dd} > 0$. This means that when $ϵ_{dd}$ is increased in a strongly cigar-shaped configuration the condensate aspect ratio tends asymptotically towards that of the trap $κ \to γ$, as it must for a system with net-repulsive spherically-symmetric contact interactions. This behavior can be seen in Fig. 5.3 where the black curves all tend to straight lines as $ϵ_{dd}$ is increased, and the asymptotic value of $κ$ they tend to is exactly the trap aspect ratio $γ$.

A parallel argument holds for the upper left hand portion of Fig. 5.3 where the condensate is quite strongly pancake-shaped ($γ > γ_{\text{crit}}^+$): in the limit $κ \to ∞$ one can show that the dipolar mean-field potential tends to $Φ_{dd} = 2gϵ_{dd}n(r)$ \[^{283}\], i.e. it behaves like a spherically-symmetric contact interaction which is repulsive when $g < 0$ and $ϵ_{dd} < 0$. In this portion of the figure one therefore also finds that as $|ϵ_{dd}| \to ∞$ the condensate aspect ratio tends asymptotically towards that of the trap $κ \to γ$.

It is tempting to conclude that when $g < 0$ the collapse that occurs as the strength of the dipolar interactions is reduced relative to the $s$-wave interactions is an “$s$-wave collapse” of the type encountered in BECs with attractive purely $s$-wave interactions \[^{315}\], which typically occurs through an unstable monopole mode \[^{317}\]. However, from Fig. 5.3 we see that the magnitude of the dipolar interaction is always finite at the collapse point. Furthermore, we shall find in subsequent sections that it is always a quadrupole mode that is responsible for collapse in a TF dipolar BEC. Collapse via a quadrupole mode has a 1D or 2D character, depending on the sign of $C_{dd}$ \[^{281}\], and is distinct from collapse via the monopole mode which has a 3D character.

In the experiment by Koch et al. \[^{260}\], where the critical scattering length $a_{\text{crit}}$ at which collapse occurs in a dipolar BEC was measured for different trapping ratios $γ$, there are a few data points corresponding to negative values of $g$ for strongly oblate condensates, and are thus of relevance to this section. We infer from their Figure 3 that collapse occurred when $ϵ_{dd} \gtrsim −7$ in a trap with $γ = 10$. However, for this trap the TF static solutions only disappear at $ϵ_{dd} \gtrsim −1.5$ and the inclusion of zero-point motion cannot explain this discrepancy between theory and experiment since it should increase the critical value of $ϵ_{dd}$ above $−1.5$, not decrease it. Furthermore, including the zero-point motion by using a gaussian ansatz leads to an almost identical theoretical prediction \[^{260}\]. Possible explanations for the discrepancy include i) The errors bars on their data imply that $−5 \gtrsim a_{\text{crit}}/a_0 \gtrsim 1$ which, due to the inverse relation between $ϵ_{dd}$ and $g$, leads to a huge uncertainty in $ϵ_{dd}$; ii) They report an uncertainty in their trapping frequencies of $0.94 < ω_x/ω_y < 1.04$, which translates into an uncertainty in the ellipticity of the trap in the $x − y$ plane of $−0.04 < ε < 0.06$. As we shall see in Section 5.2.4 below, this can have an effect upon the stability; iii) Very close to collapse
the dominant dipolar interactions may lead to significant deviations of the density profile from a single-peaked inverted parabola/gaussian profile, for example, Ronen et al. \[278\] have predicted bi-concave density structures. These may alter the stability properties of the condensate.

Having indicated how the static solutions behave for attractive $s$-wave interactions $g < 0$, for the remainder of the Chapter we will concentrate (although not exclusively) on the more common case of repulsive $s$-wave interactions.

### 5.2.4 Non-cylindrically-symmetric static solutions

![Figure 5.4](image)

**Figure 5.4**: Stable static solutions, characterised by the aspect ratios $\kappa_x$ (dotted lines) and $\kappa_y$ (dashed lines), in a non-cylindrically-symmetric trap with ellipticity $\epsilon = 0.75$ and (a) $\gamma = 0.18$, (b) $\gamma = 0.333$, (c) $\gamma = 3$ and (d) $\gamma = 5.5$. Stable (unstable) static solutions are indicated by black (gray) lines. The corresponding static solutions for $\epsilon = 0$ are indicated by solid lines.

We now consider the more general case of a non-cylindrically-symmetric system for which the trap ellipticity $\epsilon$ is finite and $\kappa_x$ and $\kappa_y$ typically differ. Note that we perform our analysis of non-cylindrically-symmetric static solutions for repulsive $s$-wave interactions $g > 0$. In Fig. 5.4 we show how $\kappa_x$ and $\kappa_y$ vary as a function of $\epsilon_{dd}$ in a non-cylindrically-symmetric trap. Different values of trap ratio are considered and generic qualitative features exist. The splitting of $\kappa_x$ and $\kappa_y$ is evident, with $\kappa_x$ shifting upwards and $\kappa_y$ shifting downwards in comparison to the cylindrically-symmetric solutions. Furthermore, the branches become less stable to collapse. For example, for
\[ \gamma = 0.18 < \gamma_{\text{crit}}^- \] (Fig. 5.4(a)), in the cylindrically-symmetric system there exist stable solutions for \( \epsilon_{dd} \to -\infty \), but in the anisotropic case, stationary solutions only exist up to \( \epsilon_{dd} \simeq -11 \).

We already noted in the introduction that for a cylindrically symmetric dipolar BEC magnetostriction causes the radial vs axial aspect ratio \( \kappa = R_x/R_z \) to differ from the trap ratio \( \gamma \), in contrast to a pure s-wave BEC for which \( \kappa = \gamma \). It is therefore interesting to note that we find that when the trap is not cylindrically symmetric a dipolar BEC also has an ellipticity in the \( xy \)-plane which differs from that of the trap, although the deviation is generally small. This occurs despite the fact that dipolar interactions are radially symmetric.

### 5.3 Calculation of the excitation spectrum

Now that we have exhibited some of the features of the static solutions in the TF regime, we wish to determine their excitation spectrum. The methods which have been previously used for finding the excitation spectrum of a dipolar BEC include: i) A variational approach applied to a gaussian approximation for the BEC density profile \[263\, 274\, 297\, 318\]. This allows one to derive equations of motion for the widths of the gaussian. ii) Using the equations of dissipationless hydrodynamics, namely the continuity and Euler equations, to obtain equations of motion for the TF radii \[275\, 301\]. This method is exact in the TF limit (recall that the TF regime is mathematically identical to the hydrodynamics of superfluids at zero temperature). iii) Solving the full Bogoliubov equations \[277\, 298\, 300\]. iv) Solving for the time evolution of the full time-dependent GPE under well-chosen perturbations \[263\, 274\, 297\].

Methods i) and ii) are simple but yield only the three lowest energy collective modes (the monopole and two quadrupole modes). However, in the pure s-wave case these methods do have the advantage of giving analytic expressions for the frequencies, and in the dipolar case the frequencies are given by the solution of the algebraic equations \[5.15\, 5.17\], which are simple to solve. This is to be contrasted with the other methods which, although more general, require much more sophisticated numerical approaches. Furthermore, the non-local nature of the dipolar interactions make numerical calculations considerably more intensive than their s-wave equivalents. Therefore, the approach we adopt here is semi-analytic, incorporating analytic results for the non-local dipolar potential, thereby reducing the problem to the solution of (local) algebraic equations.

In our approach we generalize the methodology previously applied by Sinha and Castin \[292\] to pure s-wave BECs, where linearized equations of motion are derived for small perturbations about the mean-field stationary solution. One strength of this method, in contrast to some of those mentioned above, is that it is trivially extended to arbitrary modes of excitation and unstable modes/dynamical instability. For example, extension of the variational approach to higher-order modes (e.g., to consider the scissors...
modes of an s-wave BEC [319] requires that this is “built-in” to the variational ansatz itself. We outline our approach below.

The dynamics of the condensate wave function \( \psi(r, t) \) is described by the time-dependent Gross-Pitaevskii equation,

\[
i\hbar \frac{\partial \psi}{\partial t} = \left[-\frac{\hbar^2}{2m} \nabla^2 + V + \Phi_{dd} + g |\psi|^2 \right] \psi,
\]

where, for convenience, we have dropped the arguments \( r \) and \( t \). By expressing \( \psi \) in terms of its density \( n \) and phase \( S \) as,

\[
\psi = \sqrt{n} e^{iS},
\]

one obtains from Eq. (5.21) the well-known hydrodynamic equations,

\[
\frac{\partial n}{\partial t} = -\frac{\hbar}{m} \nabla \cdot (n \nabla S) \tag{5.22}
\]

\[
\hbar \frac{\partial S}{\partial t} = -\frac{\hbar^2}{2m} |\nabla S|^2 - V - gn - \Phi_{dd}. \tag{5.23}
\]

We have dropped the term \((\hbar^2/2m\sqrt{n})\nabla^2 \sqrt{n}\) arising from density gradients - this is synonymous with making the TF approximation [252]. Note that static solutions satisfy the equilibrium conditions \( \partial n/\partial t = 0 \) and \( \partial S/\partial t = -\mu/\hbar \).

We now consider small perturbations of the density \( \delta n \) and phase \( \delta S \) about the TF static solutions found in section 5.2. The static solutions for a non-rotating condensate have \( S = 0 \) and \( n = n_{eq} \), where \( n_{eq} \) is the parabolic density profile given in Eq. (5.10) and is obtained by solving Eqns (5.15–5.17). Linearizing the hydrodynamic equations (5.22) and (5.23) in the perturbations, we find that the dynamics are governed by

\[
\frac{\partial}{\partial t} \begin{bmatrix} \delta S \\ \delta n \end{bmatrix} = \mathcal{L} \begin{bmatrix} \delta S \\ \delta n \end{bmatrix}, \tag{5.24}
\]

where

\[
\mathcal{L} = -\begin{bmatrix} 0 & g(1 + \epsilon_{dd} K)/\hbar \\ \hbar m \nabla \cdot n_{eq} \nabla & 0 \end{bmatrix}. \tag{5.25}
\]

The operator \( K \) in Eq. (5.25) is defined via its action upon \( \delta n \) as

\[
(K\delta n)(r) = -3 \frac{\partial^2}{\partial z^2} \int \frac{\delta n(r') \text{d}^3r'}{4\pi |r - r'|} - \delta n(r). \tag{5.26}
\]

The integral in the above expression is carried out over the domain where the unperturbed density given by Eq. (5.10) satisfies \( n_{eq} > 0 \), that is, the general ellipsoidal domain with radii \( R_x, R_y, R_z \). Extending the integration domain to the region where \( n_{eq} + \delta n > 0 \)
would only add $O(\delta n^2)$ effects, since it is exactly in this extended domain that $n = O(\delta n)$, whereas the size of the extension is also proportional to $\delta n$. Clearly, to first order in $\delta n$, the quantity $\epsilon_{dd}K\delta n$ is the dipolar potential associated with the density distribution $\delta n$. To obtain the global shape excitations of the BEC one has to find the eigenfunctions $\delta n, \delta S$ and eigenvalues $\lambda$ of operator $L$ of Eq. (5.25). For such eigenfunctions equation (5.24) trivially yields an exponential time evolution of the form $\sim \exp(\lambda t)$. When the associated eigenvalue $\lambda$ is imaginary, the eigenfunction corresponds to a time-dependent oscillation of the BEC. However, when $\lambda$ possesses a positive real part, the eigenfunction represents an unstable excitation which grows exponentially. Such dynamical instabilities are an important consideration, for example in rotating condensates where they initiate vortex lattice formation [292, 294]. However, in the current study we will focus on stable excitations of non-rotating systems.

To find such eigenfunctions and eigenvalues we consider a polynomial ansatz for the perturbations in the coordinates $x, y, z$, of a total degree $\nu$, that is,

$$
\delta n = \sum_{p,q,r} a_{pqr} x^p y^q z^r, \quad \delta S = \sum_{p,q,r} b_{pqr} x^p y^q z^r, \quad (5.27)
$$

where

$$
\nu = \max_{\begin{array}{c}
  a_{pqr} \neq 0 \\
  b_{pqr} \neq 0
\end{array}} \{p + q + r\}. \quad (5.28)
$$

All operators in Eq. (5.25), acting on such polynomials of degree $\nu$, result again in polynomials of the same order. For the operator $K$ this property might not be obvious, but a remarkable result known from 19th-century gravitational potential theory states that the integral in Eq. (5.26) evaluated for a polynomial density $\delta n$, yields another polynomial in $x, y, z$. Its coefficients are given in terms of the integrals $\beta_{ijk}$ defined in Eq. (5.12), and the exact expressions are presented in Appendix 5.A. The degree of the resulting polynomial is $\nu + 2$, and taking the derivative with respect to $z$ twice yields another polynomial of degree $\nu$ again. Thus, operator (5.25) can be rewritten as a matrix mapping between scalar vectors of polynomial coefficients. Numerically finding the eigenvalues and eigenvectors of such a system is a simple task, which computational packages can typically perform.

We present only the lowest-lying shape oscillations corresponding to polynomial phase and density perturbations of degree $\nu = 1$ and $\nu = 2$. These form the monopole, dipole, quadrupole and scissors modes. These excitations are illustrated schematically in Fig. 5.1 and described below, where we state only the form of the density perturbation $\delta n$, since it can be shown that the corresponding phase perturbation $\delta S$ always contains the same monomial terms. Note that $a, b, c$ and $d$ are positive real coefficients.

- **Dipole modes** $D_x$, $D_y$ and $D_z$: A centre-of-mass motion along each trap axis [320]. The $D_x$ mode, for instance, is characterised by $\delta n = \pm ax$. 

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Monopole mode $M$: An in-phase oscillation of all radii with the form $\delta n = \pm a \pm (b x^2 + c y^2 + d z^2)$.

Quadrupole modes $Q_{xy}^1$, $Q_{xz}^1$ and $Q_{yz}^1$: The $Q_1$ modes feature two radii oscillating in-phase with each other (denoted in superscripts) and out-of-phase with the remaining radius. For example, the $Q_{xy}^{1y}$ mode is characterised by $\delta n = \pm a \pm (b x^2 + c y^2 - d z^2)$.

Quadrupole mode $Q_2$: This 2D mode is supported only in a plane where the trapping has circular symmetry. For example, in the transverse plane of a cylindrically-symmetric system the transverse radii oscillate out-of-phase with each other, with no motion in $z$, according to $\delta n = \pm a(x \pm iy)^2$.

Scissors modes $S_{xy}$, $S_{yz}$ and $S_{xz}$: Shape preserving oscillatory rotation of the BEC over a small angle in the $xy$, $xz$ and $yz$ plane, respectively. The $S_{xy}$ mode is characterised by $\delta n = \pm axy$. Note that a scissors mode in a given plane requires that the condensate asymmetry in that plane is non-zero otherwise no cross-terms exist. Furthermore, the amplitude of the cross-terms should remain smaller than the condensate/trap asymmetry otherwise the scissors mode turns into a quadrupole mode.

Note that, in order to confirm the dynamical stability of the solution, one must also check that positive eigenvalues do not exist. We have performed this throughout this Chapter and consistently observe that when $\text{Im}(\lambda) \neq 0$ that $\text{Re}(\lambda) = 0$ and that when $\text{Im}(\lambda) = 0$ that $\text{Re}(\lambda) \neq 0$. It is also possible to determine excitation frequencies of higher order excitations of the BEC by including higher order monomial terms. Such modes, for example, play an important role in the dynamical instability of rotating systems.

We would like to remind the reader that they can download the MATLAB program used to perform the calculations described in this section. It includes an easy to use graphical user interface.

5.4 Excitations in a cylindrically-symmetric trap

In this section we present the oscillation frequencies of the lowest lying stable excitations of a dipolar condensate in a cylindrically-symmetric trap. Through specific examples we indicate how they behave with the key experimental parameters, namely the dipolar interaction strength $\epsilon_{dd}$ and trap ratio $\gamma$. Note that we will discuss the scissors modes in more detail in Section 5.6. Here we will just point out that two scissors modes exist, corresponding to $S_{xz}$ and $S_{yz}$, while the $S_{xy}$ mode is non-existant due to the cylindrical symmetry of the system.
5.4.1 Variation with dipolar interactions $\epsilon_{dd}$ for $g > 0$

In Fig. 5.5 we show how the collective mode frequencies vary with the dipolar interactions for the case of $g > 0$. Although it would seem experimentally relevant to present these frequencies as a function of $\epsilon_{dd}$, we plot them as a function of the aspect ratio $\kappa$ instead. We do this for the following two reasons: i) plotting the frequencies as a function of $\epsilon_{dd}$ is problematic since two static solutions (metastable local minima and unstable saddle points) can exist for a given value of $\epsilon_{dd}$; ii) in the critical region of collapse at the turning point from stable to unstable, the excitation frequencies vary rapidly as a function of $\epsilon_{dd}$, but much more smoothly as a function of $\kappa$, and so it is easier to view the behavior as a function of $\kappa$. Also, analytic expressions for the frequencies of the $M$ and $Q_1$ modes in a cylindrically symmetric dipolar BEC in the TF regime can be found in [275].
Figure 5.5: Excitation frequencies as a function of condensate aspect ratio $\kappa$ for a cylindrically-symmetric trap with aspect ratio (a) $\gamma = 0.18$, (b) $\gamma = 1$ and (c) $\gamma = 5.5$. Shown are the results for the modes $M$ (orange, circles), $D$ (black, stars), $Q_1$ (red, diamonds), $Q_2$ (purple, squares) and $S_{c_{xz}} (= S_{c_{yz}})$ (green, triangles). (d) Static solutions $\kappa$ for $\gamma = 0.18$, 1 and 5.5. Vertical dashed lines mark the transition from stable to unstable for the static solution, and this coincides with the point at which one of the frequencies tends to zero. Vertical dotted lines mark the point at which the static solution ceases to exist altogether.
It is worth pointing out that the condensate shape accounts for a significant part of the physics of these systems, and so $\kappa$ is a good variable to work with. For example, in the problem of a rotating dipolar BEC, the critical rotation frequency at which a vortex becomes energetically favorable is exactly the same as that in a purely $s$-wave BEC providing one corrects for the change in the aspect ratio due to the dipolar interactions \[321\]. However, $\kappa$ alone does not contain all the physics. In the case of the calculation of the excitation frequencies this is clear from Eq. (5.25) which depends upon both $(\nabla \cdot n_{eq}\nabla)^{\delta S}$ and $\epsilon_{dd}K\delta n$. The former term has a direct dependence upon $\kappa$ via the equilibrium density profile $n_0(r)$, whereas the latter term does not.

We consider three values of trap ratio $\gamma$, which fall into three distinct regimes: (1) $\gamma < \gamma_{-\text{crit}}$ (2) $\gamma_{-\text{crit}} < \gamma < \gamma_{+\text{crit}}$ and (3) $\gamma_{+\text{crit}} < \gamma$. Recall that $\gamma_{+\text{crit}}(\gamma_{-\text{crit}})$ is the critical value above (below) which there exist stable solutions for $\epsilon_{dd} \to +\infty(-\infty)$, see also Fig. 5.2. In each case the aspect ratio of the stable solutions exists over a finite range $\kappa = [\kappa_{-}, \kappa_{+}]$. We will now discuss each regime in turn.

$\gamma < \gamma_{-\text{crit}}$

In Fig. 5.5(a) we present the excitation frequencies for $\gamma = 0.18$ as a function of $\kappa$. The corresponding static solutions are shown as the left hand curve in Fig. 5.5(d) and confirm that the stable static solutions (solid black part of curve) exist only over a range of $\kappa = [\kappa_{-}, \kappa_{+}]$, with $\kappa_{-} \approx 0.03$ and $\kappa_{+} \approx 0.25$ indicated by vertical lines (dashed and dotted, respectively). For $\kappa > \kappa_{+}$, no static solutions exist and so the excitation frequencies are not plotted beyond this point [dotted vertical line in Fig. 5.5(a) and most left hand dotted vertical line in 5.5(d)]. For $\kappa < \kappa_{-}$, the static solution is no longer a local energy minimum but becomes instead a saddle point/maximum that is unstable to collapse [transition marked with dashed, vertical line in Fig. 5.5(a) and most left hand dashed vertical line in 5.5(d)]. Although this solution is not stable we can still determine its excitation spectrum. Crucially, this will reveal which modes are responsible for collapse and which remain stable throughout.

Three dipole modes (stars) exist. Dipole modes, in general, are decoupled from the internal dynamics of the condensate \[252\] and are determined by the trap frequencies $\omega_x, \omega_y$, and $\omega_z$. This provides an important check on our code. For the cylindrically symmetric case, $\omega_x = \omega_y = \omega_{\perp}$, and hence only two distinct dipole modes are visible. For $\kappa < \kappa_{-}$ the dipole frequencies remain constant, indicating the dynamical stability of this mode.

In general, the remaining modes vary with the dipolar interactions. Perhaps the key mode here is the quadrupole $Q_1$ mode (diamonds). At the point of collapse the $Q_1$ frequency decreases to zero. This is connected to the dynamical instability of this mode since $\text{Re}(\lambda) > 0$ for $\kappa < \kappa_{-}$. The physical interpretation of this is that the $Q_1$ mode, which comprises of an anisotropic oscillation in which the condensate periodically elongates and then flattens, mediates the collapse of the condensate into an infinitely narrow cigar-
shaped BEC. In the energy landscape picture, this occurs because the barrier between the local energy minimum and the collapsed $R_{x,y} = 0$ state disappears for $\kappa < \kappa^-$. The $Q_2$ quadrupole mode (squares) decreases to zero, and becomes dynamically unstable, after one passes into the unstable regime as indicated in Fig. 5.5(d). The monopole $M$ mode (circles) remains stable for $\kappa < \kappa^-$ and increases with $\kappa$ above this point.

$\gamma^-_{\text{crit}} < \gamma < \gamma^+_{\text{crit}}$

In Fig. 5.5(b) we present the excitation frequencies for $\gamma = 1$ as a function of $\epsilon_{dd}$. Since $\gamma^-_{\text{crit}} < \gamma < \gamma^+_{\text{crit}}$, the solutions exist over a finite range of $\epsilon_{dd}$. In terms of $\kappa$, collapse occurs at both limits of its range, i.e., for $\kappa < \kappa^-$ and $\kappa > \kappa^+$, where $\kappa^- \approx 0.3$ and $\kappa^+ \approx 2.5$ (dashed vertical lines in Fig. 5.5(b) and (d)).

Since the trap is spherically-symmetric, the dipole modes (stars) all have identical frequency, i.e. $\omega_\perp$. The $Q_1$ quadrupole frequency (diamonds) decreases to zero at both points of collapse, $\kappa^-$ and $\kappa^+$. In the former case, this corresponds to the anisotropic collapse into an infinitely narrow BEC, while in the latter case, collapse occurs into an infinitely flattened BEC. In the low $\kappa$ regime, the $Q_2$ quadrupole mode (squares) becomes unstable just past the point of collapse, but shows no instability in the opposite limit for $\kappa > \kappa^+$.

It is interesting to note that the monopole mode (circles) shows no dependence on $\kappa$ and therefore the dipolar interactions, in agreement with [275]. Additionally, we find that the aspect ratio of the density perturbation remains fixed at precisely 1 for all values of the condensate aspect ratio $\kappa$. These observations are specific to the case of $\gamma = 1$.

$\gamma > \gamma^+_{\text{crit}}$

In Fig. 5.5(c) we plot the excitation frequencies for $\gamma = 5.5$. For $\kappa < \kappa^-$, no static solutions exist, and for $\kappa > \kappa^+$, no stable solutions exist. Here $\kappa^- \approx 3.3$ and $\kappa^+ \approx 54$ (dotted and dashed vertical lines, respectively, in Fig. 5.5(c) and (d)).

Again, the dipole modes are constant, while the remaining modes vary with dipolar interactions. Apart from the quadrupole $Q_1$ mode, all modes are stable past the point of collapse, including the $Q_2$ quadrupole mode. The $Q_1$ mode decreases to zero at the point when the condensate collapses to an infinitely flattened pancake BEC, which is again consistent with this mode mediating the anisotropic collapse.

In real experiments with a finite number of atoms the zero-point kinetic energy can be expected to extend the region of stability of the BEC. Therefore, in systems that deviate from the TF limit we expect the $Q_1$ frequency to go to zero at a smaller value of $\kappa$ than that shown in Fig. 5.5(a), to go to zero at smaller and larger values of $\kappa$ at the left and right hand sides, respectively, than those shown in Fig. 5.5(b), and to go to zero at a larger value of $\kappa$ than shown in Fig. 5.5(c). In this context, we note that previous calculations of collective excitations in dipolar BECs by Góral and Santos [274] using a gaussian ansatz (that takes account of zero-point energy and so is expected to be more
accurate close to collapse but less accurate in the TF regime) also found that instabilities of the $Q_1$ mode were responsible for collapse when $g > 0$.

### 5.4.2 Variation with dipolar interactions $\epsilon_{dd}$ for $g < 0$

We now consider the analogous case but with $g < 0$. As shown in Section 5.2.3 stable solutions only exist for $\gamma > \gamma_{\text{crit}}^+ = 5.17$ and $\gamma < \gamma_{\text{crit}}^- = 0.19$, with no stable solutions existing in the range $\gamma_{\text{crit}}^- < \gamma < \gamma_{\text{crit}}^+$. Hence we will only consider the two regimes of (1) $\gamma < \gamma_{\text{crit}}^-$ and (2) $\gamma > \gamma_{\text{crit}}^+$.

#### $\gamma < \gamma_{\text{crit}}^-$

In Fig. 5.6(a) we present the excitation frequencies in a highly elongated trap $\gamma = 0.18$. Stable static solutions exist only for $\kappa^- < \kappa < \kappa^+$ where $\kappa^- \approx 0.25$ and $\kappa^+ \approx 0.29$. In this regime we find that all collective frequencies are purely imaginary and finite, and therefore stable. At the critical point for collapse $\kappa \approx 0.29$ the $Q_1$ mode frequency passes through zero and becomes purely real, signifying its dynamical instability. This shows that, as for $g > 0$, the $Q_1$ mode mediates collapse and therefore collapse proceeds in a highly anisotropic manner due to the anisotropic character of the dipolar interactions. The remaining modes do not become dynamically unstable past the critical point, and only vary weakly over the range of $\kappa$ shown. It should also be remarked that higher order modes with polynomial degree $\nu > 2$ also become unstable within the range $\kappa^- < \kappa < \kappa^+$ where no stable parabolic solutions lie, further highlighting the metastability of the $g < 0$ states and confirming the relevance of the predictions made by the uniform-density Bogoliubov spectrum (5.20) for a system in the TF regime.

#### $\gamma > \gamma_{\text{crit}}^+$

Figure 5.6(b) shows the mode frequencies in a highly flattened trap $\gamma = 5.5$, for which stable static solutions exist only in the regime $\kappa^- < \kappa < \kappa^+$ where $\kappa^- \approx 2.7$ and $\kappa^+ \approx 3.3$. Similarly, at the point of collapse $\kappa \approx 2.7$ the $Q_1$ mode has zero frequency and is dynamically unstable. Well below the critical point the $Q_2$ mode frequency also becomes zero and dynamically unstable.
Figure 5.6: Excitation frequencies as a function of condensate aspect ratio $\kappa$ for a $g < 0$ cylindrically-symmetric trap with aspect ratio (a) $\gamma = 0.18$ and (b) $\gamma = 5.5$, with corresponding static solutions shown in figure (c). Included are the results for the modes $M$ (orange, circles), $D$ (black, stars), $Q_1$ (red, diamonds), $Q_2$ (purple, squares) and $Sc$ (green, triangles). Dashed vertical lines indicate the critical point at which the stable static solutions turn into unstable ones, dotted vertical lines indicate endpoints of branches where static solutions cease to exist.
5.4.3 Variation with trap ratio $\gamma$

Having illustrated in the previous section how the excitation frequencies behave for $g < 0$, from now on we will limit ourselves to the case of $g > 0$. In Fig. 5.7 we plot the excitation frequencies as a function of $\gamma$ for various values of $\epsilon_{dd}$. A common feature is that the dipole frequencies scale with their corresponding trap frequencies, such that $\omega_{Dx} = \omega_{Dy} = \omega_{\perp}$ and $\omega_{Dz} = \gamma \omega_{\perp}$. We now consider the three regimes of zero, negative and positive $\epsilon_{dd}$.

$\epsilon_{dd} = 0$

For $\epsilon_{dd} = 0$ stable solutions exist for all $\gamma$ and the corresponding mode frequencies are plotted in Fig. 5.7(a). Our results agree with previous studies of non-dipolar BECs where analytic expressions for the mode frequencies can be obtained, see, e.g., [251] and [252]. The $Q_2$ quadrupole mode has fixed frequency $\omega_{Q_2} = \sqrt{2} \omega_{\perp}$. The scissors mode frequency corresponds to $\omega_{Sc_{xz}} = \omega_{Sc_{yz}} = \sqrt{1 + \frac{\gamma^2}{2}} \omega_{\perp}$, and the remaining modes obey the equation [252],

$$\omega^2 = \omega_{\perp}^2 \left( 2 + \frac{3}{2} \gamma^2 \pm \frac{1}{2} \sqrt{16 - 16 \gamma^2 + 9 \gamma^4} \right), \tag{5.29}$$

where the “+” and “-” solutions correspond to $\omega_M$ and $\omega_{Q_1}$, respectively.

$\epsilon_{dd} < 0$

For $\epsilon_{dd} = -0.75$ (Fig. 5.7(b)) stable solutions, and collective modes, exist up to a critical trap ratio $\gamma_{\text{max}} \approx 0.56$. Beyond that the attractive nature of side-by-side dipoles (recall $C_{dd} < 0$) makes the system unstable to collapse.

For all of the modes except the $Q_1$ quadrupole mode we see the same qualitative behaviour as for the non-dipolar case (gray lines) with the modes extending right up to the point of collapse with no qualitative distinction from the non-dipolar case. The $Q_1$ quadrupole mode, on the other hand, initially increases with $\gamma$, like the non-dipolar case, but as it approaches the point of collapse, it rapidly decreases towards zero. Above $\gamma_{\text{max}}$, the $Q_1$ mode is dynamically unstable.

$\epsilon_{dd} > 0$

For $\epsilon_{dd} = 1.5$ (Fig. 5.7(c)) stable solutions exist only above a lower critical trap ratio $\gamma_{\text{min}} \approx 2.3$. For $\gamma < \gamma_{\text{min}}$ the attraction of the end-to-end dipoles becomes dominant and induces collapse. Indeed, we find that the frequency of the $Q_1$ mode passes through zero and is dynamically unstable for $\gamma < \gamma_{\text{min}}$. Above this, the $Q_1$ and $Q_2$ frequencies increase towards the limiting values of the non-dipolar frequencies of $1.82 \omega_{\perp}$ and $\sqrt{2} \omega_{\perp}$ because in a very pancake-shaped trap the atoms cannot sample the anisotropy of the interactions. The remaining modes behave qualitatively like the non-dipolar modes for $\gamma > \gamma_{\text{min}}$. 

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Figure 5.7: Excitation frequencies in a cylindrically-symmetric trap as a function of the trap aspect ratio $\gamma$ for (a) $\epsilon_{dd} = 0$, (b) $\epsilon_{dd} = -0.75$ and (c) $\epsilon_{dd} = 1.5$. Shown are the results for the modes $M$ (orange, circles), $D$ (black, stars), $Q_1$ (red, diamonds), $Q_2$ (purple, squares) and $Sc$ (green, triangles). In figures (b) and (c) the frequencies for $\epsilon_{dd} = 0$ are included as dashed, gray lines.
5.5 Excitations in a non-cylindrically-symmetric trap and relevance to rotating-trap systems

In this section we will apply our approach to the most general case of non-cylindrically-symmetric traps. An important experimental scenario where this occurs is when condensates are rotated in elliptical harmonic traps. This has provided a robust method for generating vortices and vortex lattices in condensates (see Ref. [322] for a review). Whilst the trap ellipticity in the $x-y$ plane is typically small (in most experiments it is of the order of a few percent), the rotation accentuates the ellipticity induced in the condensate. Indeed, one can derive effective harmonic trap frequencies for the condensate which show that the effective ellipticity can be orders of magnitude greater than the static ellipticity [284, 291, 292].

The $Q_2$ mode can be pictured as a surface wave traveling around the edge of the condensate. It has a similar shape to the rotating elliptical deformation of the trap, and when the trap is rotated at frequencies close to that of the $Q_2$ mode then even a perturbatively small trap deformation strongly couples to this mode. When viewed from the frame of reference rotating with the trap, the excitation of the $Q_2$ mode appears as a bifurcation of the stationary condensate into a new stationary state which mixes in some of the $Q_2$ mode and the condensate therefore develops an elliptical shape in the $x-y$ plane. For some ranges of rotation speeds this new stationary state is in turn dynamically unstable to the excitation of higher order modes [284, 285, 292].

This dynamical instability disrupts the condensate and is the first step in the process by which vortices enter. Although this process is complex, the dynamical instability that initiates it is accurately described within the TF approximation because the modes which are initially excited are of sufficiently long wavelength. The predictions obtained within the TF approximation are in excellent agreement with both experiments [293] and numerical simulations of the GPE [294, 295]. Although we will not specifically consider rotation further here, our methodology can be easily extended to this scenario [284].

As in Section 5.2.4, we consider finite trap ellipticity $\epsilon$ in the $x-y$ plane. In Fig. 5.8 we present the mode frequencies as a function of ellipticity $\epsilon$ for three different examples. There are some important generic differences to the cylindrical case. Due to the complete anisotropy of the trapping potential the dipole mode frequencies (stars) all differ, and are equal to the corresponding trap frequencies $\omega_x = \sqrt{1-\epsilon\omega_{\perp}}$, $\omega_y = \sqrt{1+\epsilon\omega_{\perp}}$ and $\omega_z = \gamma\omega_{\perp}$. The monopole mode is present (circles) and its frequency increases with $\epsilon$. Strictly speaking the $Q_2$ mode is no longer present due to the breakdown of cylindrical symmetry. Instead we find a new $Q_1$ mode appearing (upper diamonds) which corresponds to the $Q_1^{yz}$ mode for $\epsilon_{dd} > 0$ and the $Q_1^{xz}$ mode for $\epsilon_{dd} \leq 0$. The usual quadrupole mode $Q_1^{xy}$ is also present. The reader is reminded that the superscript in the $Q_1$ mode notation refers to the in-phase radii, the remaining radius oscillates out of phase with the other two. Although there are actually three permutations of $Q_1$, only two appear for any

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Figure 5.8: Excitation frequencies in a non-cylindrically-symmetric trap as a function of the trap ellipticity $\epsilon$ in the $x - y$ plane for the cases of (a) $\epsilon_{dd} = 0$ and $\gamma = 1$, (b) $\epsilon_{dd} = -0.6$ and $\gamma = 0.8$ and (c) $\epsilon_{dd} = 1.25$ and $\gamma = 2$. Shown are the modes $D$ (black, stars), $M$ (orange, circles), $Q_1$ (red, diamonds), $S_{c_{xy}}$ (blue, triangles pointing down), $S_{c_{yz}}$ (green, triangles pointing up, upper branch) and $S_{c_{xz}}$ (green, triangles pointing up, lower branch).
given value of $\epsilon_{dd}$ since linear combinations of these and the monopole mode can form the remaining $Q_1$ mode.

We will now consider the specific features for the cases presented in Fig. 5.8. For $\epsilon_{dd} = 0$ and $\gamma = 1$ (Fig. 5.8(a)), the solutions are stable right up to $\epsilon = 1$. At this limit the $x$-direction becomes untrapped and this causes the system to become unstable with respect to the dipole $D_x$ mode, as well the $Q_1^{xy}$ mode which can now expand freely along the $x$-axis. For $\epsilon_{dd} = -0.6$ and $\gamma = 0.8$ (Fig. 5.8(b)) the solutions become unstable to collapse at $\epsilon \approx 0.425$. Only the lower $Q_1^{xy}$ mode becomes dynamically unstable at this point, indicating that it is the mode responsible for collapse, which is towards a pancake shaped system. For $\epsilon_{dd} = 1.25$ and $\gamma = 0.2$ (Fig. 5.8(c)) the solutions become unstable to collapse at $\epsilon \approx 0.45$. We again observe that the same $Q_1$ mode mediates the collapse, only this time the collapse is towards a cigar shaped system. The other modes remain stable.

5.6 Scissors modes

In this section we study the so-called scissors modes of the condensate, which describe an oscillatory rotation of the condensate in response to a sudden rotation of an elliptic trap about a small angle. The importance of the scissors modes is that they clearly distinguish between superfluid and non-superfluid systems and can thus be used to gain information about the nature of the quantum state of a trapped ultracold gas [287]. The special relevance of this to ultracold dipolar Bose gases is that when the attractive portion of the dipolar interactions becomes important, such as in cigar-shaped systems aligned along the external polarizing field, it is possible that simple Bose-Einstein condensation (macroscopic occupation of a single quantum state) is no longer energetically favored. Some time ago [323, 324], it was realized that due to quantum exchange effects repulsive interactions in spatially uniform Bose gases favor simple Bose-Einstein condensation over fragmented Bose-Einstein condensation (macroscopic occupation of two or more quantum states) [325]. The converse is expected to be true in the presence of attractive interactions, although attractive interactions in a uniform system lead to mechanical collapse. The experimental realization of trapped ultracold gases with attractive $s$-wave interactions [315] was therefore a significant event: fragmentation could potentially be studied in systems with attractive interactions which are stabilized by their zero-point energy. However, the consensus now seems to be that in attractive $s$-wave systems the magnitude of the interactions required to see significant fragmentation is too great for the zero-point energy to stabilize the system [326]. Dipolar interactions, on the other hand, are partially attractive and partially repulsive, the net balance being tunable via the shape of the atomic cloud. Recent work by Bader and Fischer [327] suggests that fragmentation can occur before collapse in low-dimensional trapped systems with anisotropic interactions. A comprehensive investigation of fragmentation in dipolar BECs is beyond the
scope of the current Chapter, but below we take a step in this direction by calculating the properties of scissors modes of a dipolar BEC.

Experimentally, one of the simplest indicators of whether or not an atomic cloud is Bose condensed is to examine the momentum distribution following free expansion after the trap is turned off \cite{248–250}. For example, according to the equipartition theorem, a gas at thermal equilibrium will expand isotropically even if the trap was anisotropic. This is not true for a BEC which, due to its zero-point energy, expands most rapidly in the direction which was most tightly confined. However, for dipolar BECs the situation is complicated by the anisotropy of the long-range interactions which continue to act at some level even as the gas expands \cite{257, 258}. Quantized vortices are another “smoking gun” indicating the presence of a BEC, but these are not easy to controllably generate in the cigar-shaped systems which would be of primary interest (although they might be useful in cases where $C_{dd} < 0$, for which pancake-shaped BECs have dominant attractive interactions). Furthermore, in cigar-shaped systems with $C_{dd} > 0$, the rotation speed at which a vortex becomes energetically favorable diverges as $\epsilon_{dd}$ increases \cite{321}. Scissors modes, on the other hand, offer an alternative vehicle for the investigation of superfluidity in dipolar systems which does not suffer from the difficulties mentioned above.

A detailed account of the scissors mode in a pure $s$-wave BEC can be found in \cite{287}. The scissors mode of a trapped atomic cloud (thermal or Bose condensed) is excited by suddenly rotating the anisotropic trapping potential over a small angle. Consequently, the atomic cloud will experience a restoring force exerted by the trap, and provided the angle of rotation is small, it will exhibit a shape preserving oscillation around the new equilibrium position. The exact response of the atomic cloud to the torque of the rotated trapping potential depends strongly on the moment of inertia of the cloud. Since a superfluid is restricted to irrotational flow, it will have a significantly different moment of inertia compared to a thermal cloud. In particular, when the trap anisotropy vanishes the moment of inertia of a superfluid also vanishes, whereas in a thermal cloud this is not the case. The superfluid scissors mode frequency will approach a finite value, whereas in a thermal cloud it will vanish as the trap anisotropy approaches zero \cite{287}. A measurement of the scissors mode frequency therefore constitutes a direct test for superfluidity \cite{282, 287}, as has been verified experimentally for non-dipolar BECs \cite{288, 290}.

In the following, we will consider the scissors mode to be excited by rotating the trapping potential as well as the external aligning field of the dipoles (except for the $Sc_{xy}$ mode which does not require a rotation of the external field) simultaneously and abruptly through a small angle, such that the condensate suddenly finds itself in a rotationally displaced configuration. Three scissors modes now appear due to the three distinct permutations of this mode, namely $Sc_{xy}$ (triangles pointing down in Fig. 5.8), $Sc_{yz}$ (triangles pointing up), and $Sc_{xz}$ (triangles pointing up). Clearly, from Fig. 5.8 the oscillation frequencies of the scissors modes are affected by the dipolar interactions. The effect of the dipolar interactions is two-fold. Firstly, since the dipolar interactions change the aspect ratio of the condensate, both the moment of inertia of the condensate and the
Figure 5.9: Schematic illustration of the dipolar restoring force for the $S_{c_{xz}}$ mode. When the condensate is rotated with respect to the dipole alignment axis $\hat{z}$ as in situation (b), the dipoles will on average be more side-by-side than in the aligned case, situation (a). Since this is an energetically unfavourable configuration compared to the aligned case, there will consequently be a dipolar restoring force present in (b) trying to re-align the condensate, illustrated by arrows.

As a result, there will be a dipolar restoring force trying to re-align the dipoles, which in turn is expected to affect the scissors mode frequencies. Figure 5.9 schematically illustrates this process for the $S_{c_{xz}}$ mode. For a pancake shaped condensate the effect is opposite. Since the dipolar interaction potential is rotationally invariant in the $x-y$ plane, the dipolar restoring force is absent for the $S_{c_{xy}}$ mode.

Explicit expressions for the scissors frequencies can be obtained by performing the procedure outlined in Section 5.3 analytically, rather than numerically. We start with the frequency $\omega_{s_{xy}}$ of the $S_{c_{xy}}$ mode, in which case we only expect an influence of dipolar interactions through changes in the geometry, and find

$$\omega_{s_{xy}}^2 = 2\omega_1^2\epsilon \left(\frac{\kappa_x^2 - \kappa_y^2}{\kappa_y^2 + \kappa_x^2}\right)^{-1},$$

where it should be noted that the quantity in brackets is precisely the ellipticity of the condensate. The $S_{c_{xy}}$ frequency does not depend explicitly on the strength of the dipolar interactions $\epsilon_{dd}$, but merely on the condensate ellipticity, which is an indication of the absence of a dipolar restoring force as discussed above. The condensate ellipticity
turns out to be approximately proportional to the trap ellipticity, where the constant of proportionality is dependent on the dipolar interaction strength $\epsilon_{dd}$ and axial trapping strength $\gamma$. As a result, the $S_{cxy}$ scissors frequencies shown in Fig. 5.8 are (almost) independent of the trap ellipticity for fixed values of $\epsilon_{dd}$ and $\gamma$. Figure 5.10(a) shows the $S_{cxy}$ frequency as a function of the axial trapping strength $\gamma$, for various dipolar interaction strengths $\epsilon_{dd}$.

In the presence of dipolar interactions, the condensate ellipticity deviates from the trap ellipticity $\epsilon$ (see Section 5.2.4), and hence the $S_{cxy}$ frequency also changes when dipolar interactions are switched on. In the absence of dipolar interactions [dashed line in Fig. 5.10(a)], the trap and condensate ellipticity are equal and the $S_{cxy}$ frequency is independent of the condensate size, trap ellipticity, as well as the $s$-wave interaction strength $[287]$. For very prolate ($\gamma \ll 1$) and very oblate ($\gamma \gg 1$) traps, the dipolar interactions become either mainly attractive or mainly repulsive and lose their anisotropic character. The dipolar potential becomes contact-like and can be renormalized into the $s$-wave interactions (see Section 5.2.3 or [283]), which do not influence the scissors mode frequency. This effect is visible in Fig. 5.10(a) in the form of the scissors frequency returning to the non-dipolar value for extremal values of $\gamma$. Finally, we would like to point out a remarkable similarity between the scissors frequencies shown in Fig. 5.10(a), and the trap rotation frequencies at which the static solution diagram of a rotating dipolar BEC shows a bifurcation point, as investigated in reference [285] (see figure 1(b) therein). For all values of $\gamma$ and $\epsilon_{dd}$ the scissors frequency is precisely twice the bifurcation frequency. Presumably, the underlying connection is the fact that the scissors mode $S_{cxy}$ has the same superfluid field as a stationary state of a BEC in a rotating trap.

Turning our attention to the $S_{cxz}$ and $S_{cyz}$ frequencies, the analytical calculation yields

$$\frac{\omega_{sxz}^2}{\omega_z^2} = \left( \frac{1}{\kappa_x^2} + \frac{1}{\kappa_y^2} \right) \frac{1 - \epsilon_{dd} \left( 1 - \frac{9}{2} \kappa_x^3 \kappa_y \beta_{102} \right)}{1 - \epsilon_{dd} \left( 1 - \frac{9}{2} \kappa_x \kappa_y \beta_{002} \right)},$$

(5.31)

$$\frac{\omega_{syz}^2}{\omega_z^2} = \left( \frac{1}{\kappa_x^2} + \frac{1}{\kappa_y^2} \right) \frac{1 - \epsilon_{dd} \left( 1 - \frac{9}{2} \kappa_x \kappa_y^3 \beta_{012} \right)}{1 - \epsilon_{dd} \left( 1 - \frac{9}{2} \kappa_x \kappa_y \beta_{002} \right)}.$$

(5.32)

Here, the quantity $\epsilon_{dd}$ appears explicitly and as such the frequencies depend directly on the strength of the dipolar interactions, an effect we attribute to the dipolar restoring force. Figure 5.10(b) shows the above frequencies for a cylindrically symmetric trap and as a function of the axial trapping strength $\gamma$, for various values of $\epsilon_{dd}$. There are two distinct effects to be noted. Firstly, when $\epsilon_{dd} > 0$ ($\epsilon_{dd} < 0$) the scissors frequencies go up (down) for cigar shaped systems and down (up) for pancake shaped systems. This behaviour is consistent with what one would expect in the presence of a dipolar restoring force. Secondly, for $\gamma \ll 1$ and $\gamma \gg 1$ we see that the $\omega_{sxz}$ frequency approaches the non-dipolar value again. For the $\omega_{sxy}$ frequency this effect could be explained solely by the fact that for such values of $\gamma$ the condensate aspect ratios return to the non-dipolar values. However, for the $S_{cxz}$ and $S_{cyz}$ modes we have to account for the apparent
Figure 5.10: Scissors frequencies as a function of the axial trapping strength $\gamma$ for various values of $\epsilon_{dd}$, with $-0.45 \leq \epsilon_{dd} \leq 0.9$ and increasing in the direction of the arrow in steps of 0.15. The dashed line indicates $\epsilon_{dd} = 0$. (a) Frequency of $S_{cxy}$ mode for fixed trap ellipticity of $\epsilon = 0.1$. For very prolate ($\gamma \ll 1$) or very oblate ($\gamma \gg 1$) systems, the dipolar interactions renormalize into the $s$-wave interactions and $\omega_{sxy}$ returns to the non-dipolar value. (b) Frequency $\omega_{sz}$ of the $S_{cz}$ mode as a function of $\gamma$ for a cylindrically symmetric trap, scaled to the non-dipolar frequency $\omega_{sz}^{(0)}$. 


vanishing of the dipolar restoring force as well. To see why it plays no part here, we have to analyze the expectation values of the quantity \( R = \sqrt{x^2 + y^2 + z^2} \). For \( \gamma \ll 1 \) we have \( \langle R \rangle \approx \langle |z| \rangle \to \infty \), and for \( \gamma \gg 1 \) we have \( \langle R \rangle \approx \langle \sqrt{x^2 + y^2} \rangle \to \infty \). Although in both cases the torque exerted by the dipolar restoring force is proportional to \( \langle R \rangle \) and in principle approaches infinity, it vanishes relative to the other two quantities contributing to the scissors frequencies, namely the moment of inertia of the condensate and the torque exerted by the trap, which both scale as \( \langle R^2 \rangle \) \cite{289}. In Fig. 5.10(b) this behaviour can be observed for the extremal values of \( \gamma \), where the scissors frequencies approach those of the non-dipolar case.

5.7 Conclusions

In this Chapter we have performed an investigation into the static and dynamic states of trapped dipolar Bose-Einstein condensates in the Thomas-Fermi regime. We have extended our previous work in this area by examining new regimes of dipolar and \( s \)-wave interactions (namely, positive and negative values of \( C_{dd} \) and \( g \)), non-cylindrically symmetric traps, and different classes of collective excitation, including the scissors modes. Our approach is based upon the analytic calculation of the non-local dipolar mean-field potential inside the condensate, for an arbitrary polynomial density profile, and exploiting the fact that this potential is again of polynomial form. Using this method, we have examined the stability of static states and collective excitations, including the behavior of the collective excitations as a function of the trap aspect ratio and ellipticity, and as a function of the relative strength of the dipolar and \( s \)-wave interactions. We consistently find that an instability of the \( Q_1 \) quadrupole mode mediates global collapse of a dipolar BEC whether \( g > 0 \) or \( g < 0 \). However, there are two critical trap ratios, \( \gamma^+_{\text{crit}} = 5.17 \) and \( \gamma^-_{\text{crit}} = 0.19 \), beyond which the BEC is stable against scaling fluctuations (monopole and quadrupole excitations) even as the strength of the dipolar interaction overwhelms the \( s \)-wave one, i.e. when \( \epsilon_{dd} \to \pm \infty \). In the case of attractive \( s \)-wave interactions \( (g < 0) \), where the dipolar interactions can stabilize an otherwise unstable condensate, the magnetostriction seems to act counter-intuitively (see Figure 5.3), although upon closer examination the behavior can be explained by understanding how dipolar interactions behave in highly confined geometries.

We have paid special attention to the scissors modes because of their sensitivity to superfluidity, which we identify as an issue of particular interest in cigar-shaped dipolar condensates due to the possibility of fragmentation when the attractive part of the dipolar interaction becomes significant. Our expressions for the frequencies of the scissors modes include a term due to a restoring force which is not present in the pure \( s \)-wave case, and which we identify as arising due to an anisotropic dipolar re-alignment force. A freely available MATLAB implementation of the calculations outlined in this Chapter, including a graphical user interface, can be obtained online \cite{302}.
Appendix 5.A   Calculating the dipolar potential inside a heterogeneous ellipsoidal BEC

In this appendix we will concern ourselves with the calculation of integrals of the form

\[ \phi[\rho](r) = \frac{1}{4\pi} \int \frac{\rho(r')}{|r' - r|} dx'dy'dz', \]

(5.33)

where the domain of integration is a general ellipsoid with semi-axes \( R_x, R_y, R_z \), and the point \( r = (x, y, z) \) is an internal point of the ellipsoid. The square brackets indicate a functional dependence. When computing the dipolar potential in Eqs. (5.7), (5.8) and (5.26) we need to evaluate this integral in order to obtain the fictitious electrostatic potential \( \phi[\rho_{ijk}](r) \) arising from a particle density of the form

\[ \rho_{ijk} = x^i y^j z^k, \]

(5.34)

with \( i, j, k \) nonnegative integers. By taking linear combinations of the general term \( \rho_{ijk} \) we can calculate the internal dipolar potential created by an arbitrary density distribution because Eq. (5.33) defines a linear integral operator acting upon \( \rho(r) \).

The physically relevant density distributions naturally can be divided into two classes:

1. The inverted parabola \( n(r) \) given by Eq. (5.10) which corresponds to the static ground state of the BEC.

2. The excitations \( \delta n(r) \) and \( \delta S(r) \) given by Eq. (5.27) which can be written as linear combination of terms \( x^i y^j z^k \).

Actually, these two classes have some overlap because certain low lying excitations (monopole, quadrupole, and scissors modes), which would otherwise seem to fall into class 2, can be described in terms of the parabolic density profile of class 1 but with time-oscillating radii (in the case of monopole and quadrupole modes), or time-oscillating symmetry axes (in the case of the scissors modes). In this appendix we give the general theory which works for all density distributions of the form (5.34).

In the context of calculating gravitational potentials in astrophysics one encounters exactly the same integrals as here and as such, the problem attracted considerable interest even in the 19\textsuperscript{th} century. Among others, significant contributions to the topic were made by MacLaurin, Ivory, Green, Poisson, Cayley, Ferrers, and Dyson. A detailed historical overview can be found, for instance, in references [307, 308]. However, for our purposes the most important contribution came from N. M. Ferrers, who showed that the potential (5.33) associated with the density (5.34) evaluates exactly to a polynomial in the coordinates \( x, y, \) and \( z \). As a matter of general interest we will outline the method employed by Ferrers to arrive at this remarkable result.

In his 1877 paper, Ferrers first shows how the internal (gravitational) potential of an
ellipsoid with semi-axes $R_x$, $R_y$, and $R_z$, with a density of the form

$$\rho = \rho(s) = s^n, \quad n = 1, 2, 3, \ldots$$

(5.35)

with

$$s = 1 - \frac{x^2}{R_x^2} - \frac{y^2}{R_y^2} - \frac{z^2}{R_z^2}$$

(5.36)

can be calculated, using integration over so-called homoeoidal shells, to be

$$\phi[s^n](x, y, z) = \frac{R_x R_y R_z}{4(n+1)} \int_0^\infty Q^{n+1} \frac{d\sigma}{\Delta},$$

(5.37)

where

$$Q = 1 - \frac{x^2}{R_x^2 + \sigma} - \frac{y^2}{R_y^2 + \sigma} - \frac{z^2}{R_z^2 + \sigma},$$

and

$$\Delta = \sqrt{(R_x^2 + \sigma)(R_y^2 + \sigma)(R_z^2 + \sigma)}.$$  

(5.38)

Homoeoidal shells are shells situated inside the ellipsoid, bounded by equidensity surfaces of the density $\rho$.

Using infinitesimally thin homoeoidal shells, the triple integral can be reduced to a single integral over the variable $\sigma$. For a detailed account on this integration process, see for instance references [276, 305, 307], or of course the original work by Ferrers [304]. Notably, the right hand side of Eq. (5.37) evaluates to a polynomial in $x, y,$ and $z$.

Next, Ferrers noted that whenever $\rho = 0$ at the boundary of the ellipsoid, then differentiation of the potential with respect to any of the cartesian coordinates, for example $x$, yields

$$\frac{d}{dx} \phi[\rho](x, y, z) = \phi \left[ \frac{d\rho}{dx} \right] (x, y, z),$$

(5.39)

which can easily be checked with integration by parts. Finally, he noted that any monomial, such as $\rho_{ijk}$ defined in Eq. (5.34), can be expressed by means of a series of differential coefficients of powers $s^m$ of the variable $s$ defined in (5.36),

$$\rho_{ijk} = \sum_m \sum_{p+q+r \leq m} A_{mpqr}^{(ijk)} \frac{d^{p+q+r}}{dx^p dy^q dz^r} s^m,$$

(5.40)

where the $A_{mpqr}^{(ijk)}$ are constants and whereby it should be noted that the order of differentiation never exceeds $m$. By virtue of the latter observation, we can calculate the potential of the above density by repeatedly applying the step (5.39) in the opposite direction, transferring all differential operators appearing in (5.40), from inside the integral $\phi[\rho_{ijk}]$ to the outside, since at each step we are ensured that the density being integrated over contains at least a factor of $s$, and hence is always equal to 0 on the boundary. Thus,
we arrive at the following result

\[ \phi[\rho_{ijk}](x, y, z) = \phi \left[ \sum_{mpqr} A_{mpqr}^{(ijk)} \frac{d^{p+q+r}}{dx^p dy^q dz^r} \phi[s^m](x, y, z) \right] (x, y, z) \]

\[ = \sum_{mpqr} A_{mpqr}^{(ijk)} \frac{d^{p+q+r}}{dx^p dy^q dz^r} \phi[s^m](x, y, z) \]

in which the \( \phi[s^m] \) terms are known through Eq. (5.37). Recalling that the potentials \( \phi[s^m] \) are in fact polynomial in \( x, y, \) and \( z, \) and hence also any differential quotient, we can conclude that the potential \( \phi[\rho_{ijk}] \) is also a polynomial. Crucial point in the above derivation is the observation expressed in equation (5.40), that any monomial can be written as a series of differential coefficients of a function of the homoeoidal shell index variable \( s, \) which is specific to ellipsoids only. In different geometries, some monomial densities might yield polynomial potentials, but in general this is not the case.

It remains to determine the precise coefficients of this polynomial, a task undertaken by F.W. Dyson who found a compact and elegant general expression \[393\]. Through the efforts of Ferrers and Dyson, the triple integral of (5.33) which depended on the coordinate \( r, \) is reduced to a finite number of single integrals appearing in the coefficients of a polynomial only.

Although Dyson’s formula in principle solves the problem, it is not particularly suited to numerical computation because it still contains differential operators. We therefore employ results from a more recent paper by Levin and Muratov \[306\], which computes the polynomial coefficients of the potential \( \phi[\rho_{ijk}] \) explicitly. Levin and Muratov make use of generalised depolarisation factors defined as

\[ M_{lmn} = (2l-1)!!(2m-1)!!(2n-1)!! \frac{K_x K_y \beta_{lmn}}{2R_z^{2(l+m+n-1)}}, \]  

(5.41)

where \( m, l, n = 0, 1, 2 \ldots, \) and \( \beta_{lmn} \) is defined in Eq. (5.12). Next, we write the exponents of \( \rho_{ijk} = x^i y^j z^k \) as

\[ i = 2\lambda + \delta_\lambda, j = 2\mu + \delta_\mu, k = 2\nu + \delta_\nu, \]

with \( \lambda, \mu, \nu \) positive integers such that the \( \delta_\mu, \delta_\nu, \delta_\lambda \) are either 0 or 1 for \( i, j, k \) even or odd (respectively), and define

\[ \sigma = \lambda + \mu + \nu + 1. \]

In the particular case of calculating the dipolar potential we are interested in the second derivative with respect to \( z, \) rather than the potential \( \phi[\rho_{ijk}] \) itself. Using the results of Levin and Muratov then, this quantity is given by
\[
\frac{\partial^2}{\partial z^2} \phi[p_{ijk}](x,y,z) = \frac{2R_x^i R_y^j R_z^k}{4^\rho} \sum_{p=0}^{\sigma-p} \sum_{q=0}^{\sigma-q} \sum_{r=1}^{\sigma-p-q} S_{pqr} (2r + \delta_\nu)(2r + \delta_\nu - 1)x^{2p+\delta_\lambda} y^{2q+\delta_\mu} z^{2r+\delta_\nu - 2} \frac{(\sigma - p - q - r)!}{(2p\delta_\lambda + 1)(2q\delta_\mu + 1)(2r\delta_\nu + 1)} \Gamma_{pq}^{(i,j,k)},
\]

(5.42)

where

\[
\Gamma_{pq}^{(i,j,k)} = \sum_{l=0}^{\lambda} \sum_{m=0}^{\mu} \sum_{n=0}^{\nu} S_{lmn} R_x^{2l+\delta_\lambda} R_y^{2m+\delta_\mu} R_z^{2n+\delta_\nu} M_{l+p+\delta_\lambda,m+q+\delta_\mu,n+r+\delta_\nu} \frac{(\lambda - l)!(\mu - m)!(\nu - n)! (2l\delta_\lambda + 1)(2m\delta_\mu + 1)(2n\delta_\nu + 1)}{(2l)!(2m)!(2n)!},
\]

(5.43)

Bibliography


Chapter 5.


Collective Excitations of Dipolar Bose-Einstein Condensates


Chapter 5.


[302] See supplementary material at: http://pra.aps.org/supplemental/10.1103/physreva.82.033612 for a Matlab implementation of the excitation frequency calculations with a graphical user interface.


Collective Excitations of Dipolar Bose-Einstein Condensates


[320] For rotating systems the directions of oscillation will differ from the trap axes, but these systems are not considered in this work.


Synthetic magneto-hydrodynamics in Bose-Einstein condensates and routes to vortex nucleation

Abstract - Engineering of synthetic magnetic flux in Bose-Einstein condensates [Lin et al., Nature 462, 628 (2009)] has prospects for attaining the high vortex densities necessary to emulate the fractional quantum Hall effect. We analytically establish the hydrodynamical behaviour of a condensate in a uniform synthetic magnetic field, including its density and velocity profile. Importantly, we find that the onset of vortex nucleation observed experimentally corresponds to a dynamical instability in the hydrodynamical solutions and reveal other routes to instability and anticipated vortex nucleation.
Chapter 6.

One of the driving forces behind quantum degenerate gas research is the emulation of many-body condensed matter phenomenon \[328\]. For quantum degenerate Bose gases, achievements include the observation of Bloch oscillations \[329\] and the Mott Insulator superfluid transition \[330–335\]. Considerable attention has also been applied to the achievement of the fractional quantum Hall (FQH) regime in rotating Bose-Einstein condensates (BECs) \[336–344\]. To obtain quantum Hall physics time reversal symmetry must be broken. In solid state devices this is done through the relatively simple process of applying a magnetic field. Since the atoms in a BEC are neutral an alternative method needs to be applied to break time reversal symmetry. In the context of FQH physics considerable focus has been applied to the breaking of time reversal symmetry through rotation.

For BECs in rotating traps the nucleation of vortices has been observed by several groups \[345–347\]. Theoretical superfluid hydrodynamical studies, in the Thomas-Fermi (TF) regime, have proved effective in calculating the rotation frequency at which vortices are nucleated by dynamical instability \[348, 349\]. Such studies, which have been extended to dipolar BECs \[350, 351\], agree with numerical predictions from the Gross-Pitaevskii equation \[348, 349, 352–355\]. However, to reach the FQH regime the number of vortices needs to be significantly larger than the number of bosons in the BEC. Typically, experiments are carried out in parabolic traps, defined by average in-plane trapping frequency \(\omega_\perp\). When the rotation frequency, \(\Omega_z\), is equal to \(\omega_\perp\) the BEC becomes untrapped. Hence, the attainment of the FQH regime, which requires \(\Omega_z \to \omega_\perp\), is an extremely challenging task. An alternative approach is to generate a synthetic vector potential \[356\], which breaks the time reversal symmetry of the problem, producing a synthetic magnetic field \[357–363\].

The recent work of Lin et al. \[363\] has demonstrated the effectiveness of such an approach, in a \(^{87}\)Rb BEC, through the experimental realization of a synthetic vector potential in the Landau gauge: \(A^\ast = A^\ast \hat{x}\), corresponding to a synthetic magnetic field \(B^\ast = \nabla \times A^\ast\). The authors showed that vortices were nucleated at a critical synthetic magnetic field. Here we generalize the TF methodology used to successfully calculate the onset of vortex nucleation in rotating systems \[348, 349\] to the case of synthetic magnetic fields in harmonically confined BECs. We analytically determine stationary solutions, including the aspect ratios, of a BEC and determine the critical synthetic magnetic field at which the stationary solutions become unstable. In the rotating case this instability corresponds to the onset of vortex nucleation. We find that this analysis predicts the synthetic magnetic field at which vortex nucleation occurred in the experiments of Lin et al. \[363\].

Our starting point is a generalized form of the Gross-Pitaevskii equation, which provides a mean-field description of the condensate wave function, \(\psi \equiv \psi(r, t)\):

\[
i \hbar \frac{\partial \psi}{\partial t} = \left\{ \frac{[-i \hbar \nabla - q^* A^\ast]^2}{2m} + V(r, t) + g |\psi|^2 \right\} \psi,
\] (6.1)
where \( m \) is the atomic mass, \( V(\mathbf{r}, t) \) is the confining potential and \( g = 4\pi\hbar^2 a_s/m \) defines the contact interactions, via the s-wave scattering length \( a_s \).

Expressing \( \psi = \sqrt{\rho} \exp[i\theta] \) in the TF limit we can rewrite the Gross-Pitaevskii equation as:

\[
\frac{\partial \rho}{\partial t} = -\nabla \cdot [\rho \mathbf{v}] \quad (6.2)
\]

\[
\frac{\partial \mathbf{v}}{\partial t} = -\nabla \left[ \frac{\nu^2}{2} + \frac{1}{m} (V + g\rho) \right]. \quad (6.3)
\]

where the generalized velocity is \( \mathbf{v} = \mathbf{v} - q^* \mathbf{A}^*/m \), with \( \mathbf{v} = \hbar/m\nabla\theta \).

To investigate stationary solutions we set \( \partial \rho/\partial t = \partial \mathbf{v}/\partial t = 0 \) and solve for \( \rho \) and \( \mathbf{v} \). Having found static solutions, they are not necessarily stable and so we analyze their dynamical stability. To do this we consider small perturbations (\( \delta\rho \) and \( \delta\theta \)) to the stationary solutions. Then, by linearizing the hydrodynamical equations, the dynamics of such perturbations are described by

\[
\frac{\partial}{\partial t} \begin{bmatrix} \delta\theta \\ \delta\rho \end{bmatrix} = - \begin{bmatrix} \mathbf{v} \cdot \nabla \\ \frac{\hbar}{m} \nabla \cdot \rho_0 \nabla \end{bmatrix} \begin{bmatrix} \frac{2}{\hbar} \\ \left( \nabla \cdot \mathbf{v} \right) + \mathbf{v} \cdot \nabla \end{bmatrix} \begin{bmatrix} \delta\theta \\ \delta\rho \end{bmatrix}. \quad (6.4)
\]

To investigate the stability of the BEC we look for eigenfunctions and eigenvalues of the above operator: dynamical instability arises when one or more eigenvalues possess a real positive part. The size of the real eigenvalues dictates the rate at which the instability grows. Imaginary eigenvalues relate to stable collective modes of the system, i.e., sloshing and breathing. In order to find such eigenfunctions we consider a polynomial ansatz for the perturbations in the coordinates \( x, y \) and \( z \) of total degree \( n \). For the examples considered below, all operators in Eq. (6.4), acting on polynomials of degree \( n \), result in polynomials of (at most) the same degree. Therefore, Eq. (6.4) can be rewritten as a scalar matrix operator, acting on vectors of polynomial coefficients, for which finding the eigenvectors and eigenvalues is a trivial task.

Below we consider the static solutions to Eqs. (2) and (3) for a BEC in a static elliptical harmonic trap under the influence of a synthetic magnetic field in the \( z \)-direction. The harmonic trapping potential has the form \( V(\mathbf{r}, t) = m\omega_\perp^2 (x^2 (1 - \epsilon) + y^2 (1 + \epsilon) + z^2 \gamma^2) /2 \). In the \( x - y \) plane the trap has a mean trap frequency \( \omega_\perp \) and ellipticity \( \epsilon \). The trap strength in the axial direction is specified by \( \gamma = \omega_z / \omega_\perp \). Throughout this Chapter we restrict our discussion to the case where \( \omega_\perp \) and \( \omega_z \) are greater than zero. Since the synthetic magnetic field is defined in terms of the synthetic vector potential there is more than one possible choice of \( \mathbf{A}^* \). We consider the two most popular choices, the Landau and symmetric gauges. In each case we find the following form of the generalized velocity provides exact stationary solutions to Eqs. (2) and (3): \( \mathbf{v} = \alpha \omega_\perp (\hat{y} + \hat{x}) - q^* \mathbf{A}^*/m \). For the symmetric (S) and Landau (L) gauges the synthetic vector potential has the forms:
\[A^* = -\hat{i}B_z y/2 + \hat{j}B_z x/2 \ (S)\] and \[A^* = -\hat{i}B_z y \ (L),\] such that \[B^* = \hat{k}B^*_z.\]

Setting \(\partial \rho / \partial t = \partial v / \partial t = 0\) in Eqs. (6.2,6.3) the stationary solutions for the respective gauges are:

\[
0 = \left( \alpha + \frac{\tilde{B}^*_z}{2} \right) \tilde{\omega}^2_x + \left( \alpha - \frac{\tilde{B}^*_z}{2} \right) \tilde{\omega}^2_y \quad (S) \tag{6.5}
\]

\[
0 = \left( \alpha + \tilde{B}^*_z \right) \tilde{\omega}^2_x + \alpha \tilde{\omega}^2_y \quad (L) \tag{6.6}
\]

where the dimensionless effective trap frequencies are

\[
\tilde{\omega}^2_x = (1 - \epsilon) + \alpha^2 - \alpha \tilde{B}^*_z + \left( \frac{\tilde{B}^*_z}{2} \right)^2, \quad (S) \tag{6.7}
\]

\[
\tilde{\omega}^2_x = (1 - \epsilon) + \alpha^2, \quad (L) \tag{6.8}
\]

\[
\tilde{\omega}^2_y = (1 + \epsilon) + \alpha^2 + \alpha \tilde{B}^*_z + \left( \frac{\tilde{B}^*_z}{2} \right)^2, \quad (S) \tag{6.9}
\]

\[
\tilde{\omega}^2_y = (1 + \epsilon) + \alpha^2 + 2\alpha \tilde{B}^*_z + \left( \tilde{B}^*_z \right)^2, \quad (L) \tag{6.10}
\]

and \(\tilde{B}^*_z = q^* B^*_z / m \omega_\perp\). Eqs. \(6.5,6.7,6.9\) [Eqs. \(6.6,6.8,6.10\)] can be solved analytically to determine \(\alpha\) in the symmetric [Landau] gauge as a function of \(\epsilon\) and \(\tilde{B}^*_z\), with the solutions being independent of the axial trapping strength, \(\gamma\) \([348, 349, 365]\).

In Fig. 1(a) we plot the solutions \(\alpha\) for the symmetric and Landau gauges respectively, for \(\epsilon = 0\). In each case for \(\tilde{B}^*_z < 2\) only one solution exists with two additional solutions, referred to as the upper and lower branch solutions, bifurcating at \(\tilde{B}^*_z = 2\). Despite the quantitative differences in the solutions for \(\alpha\) the generalized velocities, relevant in determining the physical properties of the BEC, are gauge invariant.

The solutions for \(\alpha\) [Fig. 1(a)] have qualitatively similar properties to the solutions obtained in the TF limit for a BEC in a rotating trap. This similarity is best observed through a comparison of the symmetric gauge solutions with solutions for a BEC in a rotating trap \([348, 349, 365]\). In a rotating trap \(\alpha\) is determined by Eq. (6.5), with \(\tilde{B}^*_z/2\) replaced by the dimensionless rotation frequency about the \(z\)-axis (\(\tilde{\Omega}_z = \Omega_z/\omega_\perp\)), with terms proportional to \((\tilde{B}^*_z)^2\) in Eqs. \(6.7,6.9\) set to zero. This difference emphasizes the key advantage a synthetic magnetic field has over physical rotation. For physical rotation, when \(\tilde{\Omega}_z \to 1\), \(\tilde{\omega}_x \ (\tilde{\omega}_y)\) tends to zero for the upper (lower) branch solution, hence the BEC is no longer confined. In the synthetic magnetic field case, the term proportional to \((\tilde{B}^*_z)^2\) prevents \(\tilde{\omega}_x,y\) tending to zero and the BEC remains confined.

We now consider how the synthetic magnetic field alters the TF condensate density profile:
Figure 6.1: (a) For a spherically symmetric trap, $\alpha$ as a function of the synthetic magnetic field strength, in the symmetric (solid black curve) and Landau (dashed green curve) gauges. (b) $\kappa_{x,y}$ as a function of $\tilde{B}_z^s$, in the symmetric and Landau gauges [366]. (c) $\alpha$ as a function of the synthetic magnetic field (symmetric gauge). (d) the condensate ellipticity, $(\kappa_x^2 - \kappa_y^2)/(\kappa_x^2 + \kappa_y^2)$, for the upper branch solutions in Fig. 1(c) as a function of the synthetic magnetic field strength [366]. In (c) and (d) $\epsilon = 0.01$ (solid black curve), $\epsilon = 0.05$ (dashed green curve) and $\epsilon = 0.1$ (red dotted curve).

$$\rho = \rho_0 \left(1 - \frac{x^2}{R_x^2} - \frac{y^2}{R_y^2} - \frac{z^2}{R_z^2}\right), \quad (6.11)$$

with $\rho_0 = 15N/(8\pi R_x R_y R_z)$. In Fig. 1(b) we plot the aspect ratios of the condensate, $\kappa_{x,y}^2 = R_{x,y}^2/R_z^2 = \gamma^2/\tilde{\omega}_{x,y}^2 [R_z^2 = 2g\rho_0/(m\omega_z^2)]$, as a function of the synthetic magnetic field, for both the symmetric and Landau gauges, with $\gamma = 1$ and $\epsilon = 0$. The solutions for $\kappa_{x,y}$ are gauge independent, with the solutions bifurcating at $\tilde{B}_z^s = 2$. For low synthetic magnetic fields, $\tilde{\omega}_z < 2$, the aspect ratios of the condensate in the $x$ and $y$ directions are reduced by $> 25\%$, with $\kappa_x = \kappa_y$. For $\tilde{B}_z^s > 2$, consistent with the solutions obtained for $\alpha$, we have three solutions for $\kappa_{x,y}$. Since the solutions are gauge independent, with no loss of generality, we describe the dependence of the aspect ratios, for $\tilde{B}_z^s > 2$, in terms of the solutions for $\alpha$ in the symmetric gauge [Fig. 1(a), solid black curve]. For the $\alpha = 0$ solution the aspect ratios of the condensate continue to reduce with $\kappa_x = \kappa_y$. For the upper (lower) branch solutions for $\alpha \kappa_x$ follows the upper (lower) branch solutions and $\kappa_y$ follows the lower (upper) branch solutions in Fig. 1(b), with $\kappa_x(\alpha) = \kappa_y(-\alpha)$.

For $\epsilon \neq 0$ the solutions for $\alpha$ significantly change [Fig. 1(c)]. For the symmetric gauge, the $\alpha = 0$ solution no longer exists and the plot has two distinct branches. The upper branch ($\alpha \geq 0$) is single valued and exists over all $\tilde{B}_z^s$. For this branch the ellipticity of the BEC in the $x - y$ plane is plotted as a function of the synthetic magnetic field, in
Fig. 1(d). We find that the BEC ellipticity monotonically increases from $\epsilon$ with increasing $\tilde{B}_z^*$. The lower branch ($\alpha < 0$) is double valued and exists only when $\tilde{B}_z^*$ is greater than the backbending magnetic field: $\tilde{B}_z^*(\epsilon)$. This lower branch backbending point shifts to higher $\tilde{B}_z^*$ as the ellipticity is increased [solid green in Fig. 2(a)] \[366\]. In Fig. 2(a) we have characterized the stability of the upper branch solutions through the evaluation of Eq. \[6.4\] for polynomial perturbations up to order $n = 5$ \[366\]. The instability region consists of a series of crescents \[348\]. Each crescent corresponds to a single value of the polynomial degree $n$, where higher values of $n$ add extra crescents, from above. At the high synthetic field end these crescents merge to form a main region of instability, characterized by large eigenvalues. At low synthetic magnetic fields the crescents become vanishingly small, with eigenvalues several orders of magnitude smaller than in the main instability region. These regions induce instability if they are traversed very slowly \[367\]. $\tilde{B}_z^*(\epsilon)$ and the stability of the stationary solutions of the upper branch are key to understanding the response of the BEC to the adiabatic introduction of $\epsilon$ or $\tilde{B}_z^*$.

For a fixed synthetic magnetic field as the ellipticity of the trap is introduced adiabatically, from zero, the BEC can follow two routes, depending on the value of $\tilde{B}_z^*(\epsilon)$ relative to $\tilde{B}_z^*(0)$. For $\tilde{B}_z^* < \tilde{B}_z^*(0)$ the solutions follow the upper branch until these solutions become dynamically unstable. This route to instability is schematically indicated by the vertical black arrow in Fig. 2(a). For $\tilde{B}_z^* > \tilde{B}_z^*(bif)(0)$ the BEC follows the lower branch from $\alpha = 0$ to negative $\alpha$. However, as $\epsilon$ is increased $\tilde{B}_z^*(\epsilon)$ shifts upwards [solid green curve in Fig 2(a)]. When $\tilde{B}_z^* > \tilde{B}_z^*(\epsilon)$ the lower branch no longer exists. This route to instability is schematically indicated by the vertical white arrow in Fig. 2(a). For fixed ellipticity as the synthetic magnetic field is introduced adiabatically, from zero, the BEC follows the upper branch solutions for $\alpha$, which become dynamically unstable. This route to instability is schematically indicated by the horizontal black arrow in Fig. 2(a).

In the case of rotation \[349\] the analogous instabilities due to: (i) the dynamical instability of the upper branch solutions or (ii) the shifting of the backbending point lead to the nucleation of vortices. As such we expect that for: (i) the adiabatic introduction of a synthetic magnetic field, for a fixed trap ellipticity or (ii) the adiabatic introduction of trap ellipticity, for a fixed synthetic magnetic field, the onset of vortex nucleation can be determined by the onset of instability in the upper branch solutions and the evolution of the backbending point.

To test this we consider the recent experiment of Lin et al. \[363\] where the synthetic magnetic field was generated via an engineered, Landau gauge, vector potential \[361\]. To engineer the vector potential an $^{87}$Rb BEC was illuminated with a pair of Raman laser beams with a momentum difference along the $x$-direction, producing three tunable dressed states. The dressed state with the lowest energy corresponds to the following Hamiltonian along the $x$-direction $H_x = (\hbar k_x - q^*A_x^*)^2/2m^*$. In this Hamiltonian $A_x^*$ is the synthetic vector potential, controlled by the Zeeman shift for the atoms with synthetic charge $q^*$ and an effective mass for the atoms along the $x$-direction $m^*$. To
produce the desired spatially synthetic vector potential the Zeeman shift varies linearly in the \( y \)-direction, resulting in an effective vector potential: \( A_x^* = -B_x^* y \). Including the effective mass into the hydrodynamical analysis:

\[
\nu = \left( \frac{\hbar}{m^*} \partial_x - \frac{q^* B_z^* y}{m^*} \right) \hat{i} + \frac{\hbar}{m} \partial_y \hat{j} = \left( \frac{m}{m^*} \alpha \omega_\perp y + \frac{q^* B_y^* y}{m^*} \right) \hat{i} + \alpha \omega_\perp x \hat{j},
\]

(6.12)

the stationary solutions are determined by

\[
0 = \left( \frac{m}{m^*} \alpha + \tilde{B}_z^* \right) \tilde{\omega}_x^2 + \alpha \tilde{\omega}_y^2
\]

(6.13)

with \( \tilde{B}_z^* = q^* B_z^*/(m^* \omega_\perp) \), \( \tilde{\omega}_x^2 \) defined by Eq. (6.8) and

\[
\tilde{\omega}_y^2 = (1 + \epsilon) + \frac{m}{m^*} \alpha^2 + 2 \alpha \tilde{B}_z^* + \frac{m^*}{m} \left( \tilde{B}_z^* \right)^2.
\]

(6.14)

In the experiments the synthetic magnetic field is ramped up over 0.3s. Thus as the synthetic field is increased the solution starting at \( \alpha = 0 \) when \( \tilde{B}_z^* = 0 \) will be tracked, until it becomes dynamically unstable. Experimentally vortex nucleation started when the synthetic magnetic flux \( (\Phi_B^z = \pi R_x R_y B_z^*) \), in units of the flux quantum \( (\Phi_0 = \hbar/q^*) \), passing through the BEC

\[
\frac{\Phi_B^z}{\Phi_0} = \frac{\tilde{B}_z^*}{2} \left( 15 N \tilde{a}_s \gamma \right)^{\frac{2}{3}} \left( \tilde{\omega}_x \tilde{\omega}_y \right)^{-\frac{2}{3}} \approx 10,
\]

(6.15)
see Fig. 2(g) of Ref. [363]. In Eq. (6.15) we have redefined the contact interactions to include the effective mass difference in the $x$-direction, such that $\tilde{a}_s = a_s/(2.5^{1/3}l_{xy})$, with $l_{xy} = \sqrt{\hbar/m\omega_\perp}$. Using the experimental parameters [368] the stationary solution becomes unstable when $\tilde{B}_z^* = 0.74$ [Fig. 2(b)], corresponding to $\frac{\Phi_B}{\Phi_0} = 11.1$, in close agreement with the experimental observation of the onset of vortex nucleation. This strongly suggests that the observed onset of vortex nucleation arises from a dynamical instability as calculated from the stationary TF solutions [369].

The engineering of synthetic magnetic fields, in BECs, offers a new and exciting route into the emulation of FQH physics. Such an arrangement also provides a new route to investigate the FQH regime in lattice systems, without the need to rotate the lattice. By adapting the techniques developed by Sinha and Castin [348] we have shown that the nucleation of vortices, the first step in obtaining high density vortex systems, due to adiabatic changes in the synthetic field and ellipticity of the trap can be calculated in the TF regime. This enables us to analytically determine the aspect ratios of the BEC for fields below the nucleation point and show that the experimental observation of the onset of vortex nucleation is due to dynamical instabilities in the stationary state of the BEC.

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Bibliography


Synthetic magneto-hydrodynamics in Bose-Einstein condensates


[366] This result is independent of the chosen gauge.


[368] N = 1.4 × 10^5, a_s = 5 × 10^{-9} m, m^* = 2.5 m, \omega_x = 80\pi rad/s, \omega_y = 60\pi rad/s (\omega_\perp = 320 rad/s and \epsilon = 0.38) and \gamma = 1.37. These parameters correspond to an experimental detuning gradient \delta of 0.2 kHz \mu m^{-1}. For precise details of the dependence of the effective mass and trap parameters on the detuning gradient see Refs. [361, 363].

[369] Experimental parameters, \(N\tilde{a}_s = 350\), imply the system is in the TF regime.
Quantum Engineering with Ultracold Atoms

The quantum laws governing the microscopic world present many opportunities for designing new technologies and developing new materials. Currently a large research effort is directed at developing a quantum computer. Powered by intrinsic quantum phenomena such as the superposition principle and entanglement, this computer would be able to do calculations no other computer can. The closely related quantum simulator would be able to simulate the physics of other complex quantum systems, aiding in the development and understanding of novel materials with unique properties such as high-Tc superconductivity. For these future technologies, precise control over the quantum state of many-particle systems is a necessity.

Perhaps the best way to study, engineer and control quantum systems is using ultracold atomic gases. In this highly active field of research, atoms are cooled to temperatures in the micro- or nano-Kelvin regime. When the atoms reach these temperatures, they reveal their quantum nature in ways directly observable to us. Systems of ultracold atoms provide very clean quantum systems which can often be described from first principles, allowing the effects of interest to be studied and understood in isolation from other complicating factors. Additionally, ultracold atoms offer a high degree of experimental control. Using a combination of lasers and electromagnetic fields, experimentalists are able to modify system parameters such as trapping geometries, dimensionality and even interaction strengths. Theorists can put their models to the test over wide parameter ranges, and exotic regimes can be probed. In this thesis, we have studied aspects of two important ultracold quantum systems: Rydberg atoms and Bose-Einstein condensates.

Rydberg atoms are atoms with one electron excited to an orbital with very high principal quantum number, giving rise to exaggerated properties such as high sensitivity to external fields and extremely strong and tunable interactions. In this thesis, the interaction strengths between rubidium atoms occupying either of two Rydberg S-states are calculated, in the Van der Waals regime, and in the presence of electric fields. When the principal quantum numbers of the two atoms differ by one, an interaction process can occur in which the two atoms swap states. It is shown how quantum simulations of...
several paradigmatic spin systems known from condensed matter physics can be realised, by arranging many interacting Rydberg atoms on a lattice. Besides a previously known quantum Ising model, novel Rydberg implementations are found of a spin-1 system and the famous Heisenberg XXZ model. External electric fields allow tuning through a large parameter regime. The inclusion of a second Rydberg level is essential; it provides a key ingredient in the form of the swapping interaction.

Next, a theoretical investigation is performed for creating crystalline states of interacting Rydberg atoms in a disordered gas, using carefully crafted laser pulses. Such Rydberg crystals are a model for dilute metallic solids with tunable lattice parameters, and provide an interface between matter and light, with applications in quantum computing and quantum cryptography. By adiabatically varying the laser detuning and intensity, it is shown how 5 or 6 regularly spaced excitations in a 1D geometry can be created in a few microseconds, for four different experimental systems. The systems considered are atoms in optical lattices, in a magneto-optical trap, in a high density dipole trap, and a Bose-Einstein condensate. The crystalline structures arising in two and three dimensions are also computed.

Ultracold atom experiments often require precisely controlled optical potentials. This is particularly true for the Rydberg experiments discussed in this thesis. As a first step towards performing these Rydberg experiments, the creation of arbitrary optical potentials is investigated, both theoretically and experimentally, with the use of a spatial light modulator (SLM). An SLM is a commercially available device, able to imprint a spatially varying phase onto a laser light field and thus controlling the light intensity in the focal plane of a lens. Existing techniques and algorithms are explored for finding appropriate phase patterns, and improved upon with a novel feedback algorithm. Furthermore, experimental issues such as aberrations in the optical path are addressed and corrected. Finally, the SLM is implemented in an existing ultracold experiment, demonstrating the capability to shape excitation and ionisation lasers and imprint spatial structures inside an atomic cloud.

The second ultracold quantum system studied in this thesis is a Bose-Einstein condensate (BEC), where large numbers of atoms all condense into a single ground state, forming a giant matter wave. BECs provide a unique window on the quantum world, where for instance aspects of superfluidity can be clearly observed. In this thesis, a general method for obtaining the exact static solutions and collective excitation frequencies is presented, for a BEC in the Thomas-Fermi regime, and in the presence of long-range and anisotropic dipolar interactions. The method incorporates analytic expressions for the dipolar potential of an arbitrary polynomial density profile, obtained from 19th century astrophysics, thereby reducing the problem of handling nonlocal dipolar interactions to the solution of algebraic equations. The parameter space is extensively explored for all possible trap configurations and interaction parameters. Particular attention is being paid to the scissors modes, which can provide a signature of superfluidity.

The methodology of finding stationary states and collective excitations is also useful
for a different Bose-Einstein system. Recent experiments have demonstrated the engineering of synthetic magnetic flux in BECs, introducing a Lorentz-like force for neutral atoms. This has prospects for quantum simulation of superconductors, and attaining the high vortex densities necessary to emulate the fractional quantum Hall effect. In this thesis, the hydrodynamical behaviour and stationary states of such a BEC in the presence of a synthetic magnetic field are calculated. The experimentally observed onset of vortex nucleation is found to correspond to a dynamical instability in the collective excitation spectrum. Using this knowledge, new routes towards vortex nucleation are predicted.
Samenvatting

Quantum Engineering met Ultrakoude Atomen

De quantum wetten die het gedrag van de allerkleinste deeltjes bepalen, bieden veel mogelijkheden en perspectieven voor het ontwerpen van nieuwe technologïën en nieuwe materialen. Zo wordt er momenteel grootschalig onderzoek gedaan naar het ontwikkelen van een zogenaamde quantum computer. Deze computer ontleent zijn kracht aan intrinsieke quantum fenomenen zoals het superpositie-principe en verstrengeling, en kan zo berekeningen doen die geen enkele andere computer kan. De nauw-verwante quantum simulator zou de fysica van complexe quantum systemen kunnen simuleren, om zo een bijdrage te leveren aan het begrijpen en ontwikkelen van nieuwe materialen met unieke eigenschappen, zoals bijvoorbeeld supergeleiding bij hoge temperaturen. Deze toekomstige technologieën vereisen een hoge graad van controle over de quantum toestanden van veel-deeltjes systemen.

Wellicht de beste manier om quantum systemen te leren beheersen en begrijpen, is door gebruik te maken van ultrakoude atomaire gassen. In dit zeer actieve onderzoeksgebied worden atomen afgekoeld tot temperaturen in de orde van micro- of nano-Kelvins. Wanneer atomen zover worden afgekoeld, onthullen zij hun quantum eigenschappen zodanig dat wij ze direct kunnen waarnemen. Ultrakoude atomen vormen fysische systemen die goed met elementaire theorieën te beschrijven zijn waarvoor weinig tot geen benaderingen nodig zijn. Hierdoor is het mogelijk interessante effecten te isoleren en te bestuderen, zonder hinder van complicerende factoren. Bovendien bieden koude atomen de mogelijkheid experimentele vrijheidsgraden zeer precies in te stellen en over een groot bereik te reguleren. Met behulp van lasers en electromagnetische velden kunnen experimentatoren parameters manipuleren zoals de geometrie van de opsluiting van de atomen, de dimensionaliteit van het systeem en zelfs de sterkte van de wisselwerkingen tussen de deeltjes. Theoreten kunnen hun modellen toetsen over een groot bereik van de parameters, en exotische regimes kunnen worden onderzocht. In dit proefschrift zijn diverse aspecten onderzocht van twee belangrijke ultrakoude, atomaire quantum systemen: Rydberg atomen en Bose-Einstein condensaten.

Rydberg atomen zijn atomen waarvan een electron is geëxciteerd naar een orbitaal
met een zeer hoog principieel quantum getal. Het atoom krijgt hierdoor extreme eigen-
schappen, zoals grote gevoeligheid voor externe velden en zeer sterke, reguleerbare wissel-
werkingen. In dit proefschrift zijn de sterktes van de wisselwerkingen tussen twee rubid-
ium atomen berekend, in het Van der Waals regime, wanneer de atomen naar (mogelijk
verschillende) Rydberg \( S \) toestanden zijn geëxciteerd. Wanneer de principiële quantum
getallen precies met één verschillen, kan er een wisselwerkingproces optreden waarbij de
atomen van toestand verwisselen. Er wordt aangetoond hoe op deze manier quantum
simulaties kunnen worden gerealiseerd van belangrijke modellen uit de gecondenseerde
materie, door vele Rydberg atomen in een rooster-geometrie te plaatsen. Behalve een
reeds bekend quantum Ising model, zijn ook nieuwe Rydberg implementaties gevonden
van een spin-1 systeem en het gevierde Heisenberg XXZ model. De systemparameters
can over een groot bereik gevarieerd worden door middel van het aanleggen van
externe elektrische velden. De mogelijkheid om twee verschillende Rydberg niveaus te
bezetten is essentieel voor de quantum simulatie; het brengt een onmisbaar ingrediënt
met zich mee in de vorm van het verwisselings-proces.

Een volgend theoretisch onderzoek in dit proefschrift richt zich op het creëren van
tussentraden van wisselwerkende Rydberg atomen in een ongeordend gas. Hier-
bij wordt gebruik gemaakt van zorgvuldig vormgegeven laser pulsen. Dergelijke Rydberg
traden vormen onder meer een quantum simulatie van metalen vaste stoffen, met
instelbare rooster parameters. Verder kunnen Rydberg kristallen fungeren als interface
tussen licht en materie, met applicaties in quantum computers en quantum cryptografie.
In dit proefschrift wordt aangetoond hoe 5 of 6 Rydberg excitaties met regelmatige on-
derlinge afstand kunnen worden gemaakt op een lijn, door de verstemming en intensiteit
de van de excitatielasers adiabatisch te variëren. Dit is mogelijk in een tijdschane van enkele
microseconden, en wordt onderzocht voor vier relevante experimentele systemen, in een
eén-dimensionale geometrie. De systemen in kwestie zijn atomen in optische roosters, in
een magneto-optische val, in een dipool-val met hoge dichtheden, en een Bose-Einstein
condensaat. Tot slot worden de kristalleine structuren berekend in het geval van twee of
drie dimensionele systemen.

Experimenten met ultrakoude atomen vereisen vaak zeer precieze optische poten-
tialen. Dit is in het bijzonder zo voor de voorgestelde Rydberg experimenten die in
dit proefschrift zijn behandeld. In dit proefschrift wordt een eerste stap richting het
daadwerkelijk uitvoeren van deze experimenten gezet in een theoretisch en experimenteel
onderzoek naar het creëren van willekeurige optische potentialen, met behulp een Spa-
tial Light Modulator (SLM). Een SLM is een commercieel verkrijgbaar apparaat, dat
een ruimtelijk variërende fase kan toevoegen aan een laser-lichtveld. Hiermee kan de
intensiteit in het brandvlak van een lens worden gereguleerd. Bestaande technieken en
algoritmes voor het vinden van een geschikt fase-patroon worden onderzocht. Vervolgens
worden verbeteringen doorgevoerd in de vorm van een nieuw terugkoppeling-algoritme.
Vervolgens worden experimentele complicaties zoals aberraties in het optisch systeem
verholpen met hulp van de SLM. Tot slot wordt de SLM ingebouwd in een bestaand
experiment met ultrakoude gassen, waarbij het succesvol vormgeven van de excitatie en ionisatie lasers wordt gedemonstreerd en ruimtelijke structuren in een atomenwolk worden geprojecteerd.

Het tweede ultrakoude quantum systeem dat in dit proefschrift wordt bestudeerd is een Bose-Einstein condensaat (BEC), bestaande uit grote aantallen atomen die allen in een enkel grondtoestand zijn gecondenseerd. Deze atomen vormen een grote materiegolf die een uniek venster op de quantumwereld verschaf, waarmee bijvoorbeeld het fenomeen van superfluiditeit bestudeerd kan worden. Een eerste onderzoek richt zich op het beschrijven van stationaire toestanden en collectieve excitaties van een BEC in het Thomas-Fermi regime, in de aanwezigheid van lange-drachts, anisotrope dipolaire wisselwerkingen. De methode ontwikkelt exacte uitdrukkingen voor de dipolaire potentiaal van ellipsoïde met een willekeurige polynomiale dichtheid, gebruik makend van resultaten uit de 19e eeuwse astrofysica. Problemen met niet-lokale dipolaire interacties worden zo gereduceerd tot het oplossen van algebraïsche vergelijkingen. De parameter-ruimte voor het systeem wordt grondig onderzocht, waarbij alle mogelijke geometrieën van de atomaire opsluiting en alle mogelijke interactie sterktes aan bod komen. Speciale aandacht wordt besteed aan de zogenaamde 'scissors'-mode, welke als indicator gebruikt kan worden voor superfluiditeit van een BEC.

De methode voor het vinden van stationaire toestanden en collectieve excitaties is tevens bruikbaar voor een ander Bose-Einstein systeem. Recente experimenten hebben laten zien dat het mogelijk is een synthetische magnetische flux te genereren in een BEC. Hiermee kan een 'Lorentz-kracht' worden gegenereerd voor ladings-neutrale atomen, hetgeen toepassingen heeft in de quantum simulatie van supergeleiders en het realiseren van de hoge vortex-dichtheden benodigd voor het bestuderen van het Quantum Hall effect. In dit proefschrift worden het hydrodynamisch gedrag en stationaire toestanden van een BEC berekend in de aanwezigheid van een synthetisch magnetisch veld. Er wordt aangetoond dat de aanvang van vortex-nucleatie, zoals geobserveerd in experimenten, correspondeert met een dynamische instabiliteit in het collectieve excitatiespectrum. Gebruik makend van deze kennis kunnen nieuwe manieren van vortex-nucleatie worden voorspeld.
List of Publications


(continued on next page)


Curriculum Vitae

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Rydberg Atomen", Max Planck Institute for the
Physics of Complex Materials, Dresden, Duitsland
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