Dipole - dipole interaction between cold Rydberg atoms

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On the cover is depicted a Stark map: the energy levels of Rydberg states with principal quantum numbers between 32 and 35 in a static electric field. The rainbow color scheme was inspired by work from the artist Jen Stark.

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1 Introduction

1.1 Quantum computing

The combination of quantum mechanics and technology has many promises of which the quantum computer might be the most spectacular one. Despite this claim, the quantum computer is not existing yet. The reason is that there are competing requirements from quantum mechanics and from technology. The bit of a quantum computer, the qubit, can have the value $|0\rangle$ and $|1\rangle$ at the same time, whereas the classical computer bit is either 0 or 1. This is called superposition. Secondly, the qubits are entangled, which means their values are connected. The advantage of the quantum computer lies in the combination of entanglement and superposition: complicated calculations are performed with all qubits at the same time, while they also have all possible values at the same time. This makes the quantum computer much faster than a classical computer.

The qubits in a quantum computer should be realized with quantum mechanical objects and they should be able to have an unperturbed coherent evolution. In other words, they should be light, cold and isolated. Hardware implementation requires, on the other hand, a system that is sufficiently large and sufficiently strongly coupled to a measuring device. This conflict is very generic and there are various proposals for a solution from a surprisingly wide range of physics. Quantum information can be encoded in, for instance, the various spins of electrons in a molecule (the NMR approach) [96], the spins of an electron in a solid state [53] or the internal state of a trapped ion [15]. But there are many more proposals [44], including ones that seem very exotic at first sight, such as a quantum bit on basis of the topology of an $N$-particle configuration in a 2D system [75, 8]. This thesis investigates the idea to use the states of gas-phase Rydberg atoms as qubits, which are atoms in highly excited states.

A quantum computer requires operations involving multiple qubits, notably the XOR-operation, which requires interaction between qubits. A system of interacting Rydberg atoms can perform this task and has some unique advantages:

- The interaction between particles is unusually strong for quantum systems.
- The interaction between particles can be switched on and off, e.g. by exciting an atom to a Rydberg state and de-exciting back to the ground state, or by tuning an electric field.
- A system can have typical dimensions of micrometers instead of nanometers, which makes it technologically easily accessible.
The building blocks (Rydberg atoms) are neutral, and have therefore a relatively weak interaction with the environment.

The possible relevance of Rydberg atoms for quantum information was first pointed out by the Zoller group in Innsbruck [46, 65]; they sketched a potential solution for the XOR gate, which makes use of the so-called dipole-blockade; alternative proposals followed [81, 84]. This thesis focuses on the dipole-dipole interaction between Rydberg atoms, which is the cause of the dipole-blockade. In short, the goal of the research presented in this thesis is to investigate the interaction between Rydberg atoms and to explore the possibilities and limitations of the system of interacting Rydberg atoms in its future contribution to quantum information processing.

The dipole-dipole interaction occurs in many places and systems in nature, but it is most easily studied and manipulated in Rydberg atoms, because of the accessible distance scale. A well-known form of the dipole-dipole interaction is the Van der Waals-interaction. This interaction is very weak due to the non-resonant character, and only significant at smaller distances; it drops off with the sixth or seventh power of distance. In the familiar case of droplet formation in liquids, the interaction occurs between particles at distances comparable to their own size. Here we focus on a resonant and thus stronger type of dipole-dipole interaction. In addition, the Rydberg atoms are a thousand times larger than regular atoms and therefore have an interaction that is a million times stronger. Due to the unusually strong interaction we can work with distances that are four orders of magnitude larger compared to the normal distance in a liquid or a solid. This makes the dipole-dipole interaction twelve orders of magnitude smaller - the interaction decreases with the third power of the atom-atom distance. Still, the interaction energy is Planck’s constant times hundreds of kHz; interactions that are detectable within 10’s of microseconds. Two dipole-interacting Rydberg atoms are schematically depicted in figure 1.1.

**Figure 1.1:** A schematic representation of two Rydberg atoms that are used as qubits (not to scale). Dipole-dipole interaction ($V_{dd}$) causes the left atom to make a transition from $|0\rangle$ to $|1\rangle$ while the right atom makes a transition from $|1\rangle$ to $|0\rangle$.

The dipole-dipole interaction occurs in many places and systems in nature, but it is most easily studied and manipulated in Rydberg atoms, because of the accessible distance scale. A well-known form of the dipole-dipole interaction is the Van der Waals-interaction. This interaction is very weak due to the non-resonant character, and only significant at smaller distances; it drops off with the sixth or seventh power of distance. In the familiar case of droplet formation in liquids, the interaction occurs between particles at distances comparable to their own size. Here we focus on a resonant and thus stronger type of dipole-dipole interaction. In addition, the Rydberg atoms are a thousand times larger than regular atoms and therefore have an interaction that is a million times stronger. Due to the unusually strong interaction we can work with distances that are four orders of magnitude larger compared to the normal distance in a liquid or a solid. This makes the dipole-dipole interaction twelve orders of magnitude smaller - the interaction decreases with the third power of the atom-atom distance. Still, the interaction energy is Planck’s constant times hundreds of kHz; interactions that are detectable within 10’s of microseconds. Two dipole-interacting Rydberg atoms are schematically depicted in figure 1.1.

**1.2 Rydberg atoms**

The name "Rydberg atoms” honors the Swedish physicist Johannes Rydberg. He introduced the Rydberg formula, which gives the wavenumber of the spectral lines in hydro-
1.2 Rydberg atoms

gen, but also other atoms, especially alkali atoms. For hydrogen, the Rydberg formula is
\[ \frac{1}{\lambda} = \text{Ry} \left( \frac{1}{n_1^2} - \frac{1}{n_2^2} \right) \],
(1.1)
with \( 1/\lambda \) the wavenumber, \( n_1 \) and \( n_2 \) integer values and \( \text{Ry} \) the Rydberg constant. For high values of \( n \) equation 1.1 is most accurate, which is why atoms with high \( n \) are called Rydberg atoms.

Rydberg atoms have highly exaggerated properties compared to ground-state atoms. They are large, very sensitive to electromagnetic fields and, as we study in this thesis, they interact with each other at large distances. Some of their properties can easily be deduced from the Bohr model. The Bohr model is not as accurate as quantum mechanics, but it is much easier and it works as a first approximation for the hydrogen atom. Quantization of angular momentum \((mvr = n\hbar)\) is combined with classical mechanics – the Coulomb force equals the centripetal force on the electron:
\[ \frac{1}{4\pi\epsilon_0} e^2 \frac{r^2}{r^2} = \frac{mv^2}{r} \].
(1.2)
This leads to an expression of the radius of the electron orbit
\[ r = \frac{4\pi\epsilon_0 m^2 \hbar^2}{e^2 m} = a_0 n^2 \]
(1.3)
where \( a_0 \) is the Bohr radius. The energy of such an electron is negative, because the electron is bound to the positively charged nucleus
\[ E = -\frac{e^4 m}{(4\pi\epsilon_0)^2 \hbar^2 2n^2} = -\frac{\text{Ry}}{n^2} \].
(1.4)
Bohr had hereby expressed the Rydberg constant in terms of already known constants.
We see that with increasing \( n \), the radius increases as \( n^2 \) and the binding energy decreases as \( 1/n^2 \). Also note that the energy levels of subsequent \( n \) states come increasingly closer together for higher \( n \); the energy difference between two adjacent \( n \) states scales like \( 1/n^3 \).

The larger distance of the electron to the nucleus makes the atom more sensitive to external electric fields; an external electric field can easily overcome the Coulomb field of the nucleus far away. It can easily be shown from the Bohr atom that the electron can escape the atom for an external field of
\[ F_{\text{ion}} = \frac{1}{4\pi\epsilon_0} \frac{\text{Ry}^2}{4n^4} = \frac{1}{16n^4} \text{ a.u.} \],
(1.5)
where the right hand side is the more usual form in atomic units (see section 2.1).

The round trip time of the electron around the nucleus scales as \( n^3 \), simply from \( T = 2\pi r/v \) for uniform circular motion. The radiative lifetime of the Rydberg state also scales like \( n^3 \). This is explained by the fact that the electron spends most of its time far away from the nucleus, where it is almost a free electron. Free electrons can not radiate, so the electron can only radiatively decay when it’s close to the core, which occurs once every round trip.
The dipole moment $\mu$ is given by the separation of charge, so this scales equally as the electron-core distance: $\mu \sim n^2$. The dipole-dipole interaction energy between two similar dipoles is deduced from the energy of a dipole in the electric field of the other dipole: $V_{dd} = -\mu \cdot F_d$. The field of a dipole at distance $R$ is the sum of the field of the nucleus $1/R^2$ and the field of the electron $-1/(R + n^2)^2$, which approximately adds up to $F_d \sim n^2/R^3$. The dipole-dipole interaction energy therefore becomes $V_{dd} \sim n^4/R^3$.

All these scaling laws for $n$ show that for highly excited states (high $n$) many properties are exaggerated. This enables interesting experiments, which are not possible with ground state atoms. The long lifetime gives the opportunity to study the atoms for a long time. The sensitivity to external fields makes it relatively simple to study their behavior in very modest fields. The large dipole moments make it possible to study interactions between atoms at very large distances. Some values for $n = 50$ are given in table 1.1.

<table>
<thead>
<tr>
<th>Property</th>
<th>n-dependence</th>
<th>value for $n = 50$</th>
</tr>
</thead>
<tbody>
<tr>
<td>radius</td>
<td>$n^2$</td>
<td>0.2 $\mu$m</td>
</tr>
<tr>
<td>binding energy</td>
<td>$n^{-2}$</td>
<td>5.4 meV</td>
</tr>
<tr>
<td>natural lifetime</td>
<td>$n^3$</td>
<td>$\sim$0.2 ms</td>
</tr>
<tr>
<td>ionization field</td>
<td>$n^{-4}$</td>
<td>51 V/cm</td>
</tr>
<tr>
<td>maximum permanent dipole moment</td>
<td>$n^2$</td>
<td>9300 debye</td>
</tr>
<tr>
<td>dipole-dipole interaction</td>
<td>$n^4/R^3$</td>
<td>$2\pi$ 1.6 MHz (R=20 $\mu$m)</td>
</tr>
</tbody>
</table>

1.3 Experimental context

In the 20th century many experiments with Rydberg atoms have been performed already, but the development of the magneto-optical trap has set off a new wave of Rydberg atom experiments in the 21st century. In the magneto-optical trap ground-state atoms are cooled down to $\sim100$ $\mu$K by means of laser cooling. This provides more control over the position of the atoms, since they move slower. Having obtained such a cold cloud of atoms, the atoms can be excited to Rydberg states with a laser [7, 69]. Another approach is deceleration and trapping of Rydberg atoms by static electric fields [97, 42]. The main focus is on interactions between Rydberg atoms, in most cases electric dipole-dipole interactions.

Dipole-dipole interactions can occur between static dipoles, but in Rydberg atom experiments often transition dipoles are used, where both atoms undergo a transition between two angular momentum states.Due to energy conservation, the energy that one atom gains with the transition equals the energy that the other atom loses. This is called a Förster resonance. In Rydberg atoms, there is a lot of choice for initial energy levels and, furthermore, the energy levels can easily be tuned with an electric field, so a resonant two-atom transition is easily found. Resonant dipole-dipole interactions have been studied with room-temperature Rydberg atoms in the past, where the transition was brought into resonance with a static electric field [85, 90] or an oscillating field, i.e. electro-magnetic radiation [34, 16, 73].
Several interesting effects are observed in a sample of ultracold Rydberg atoms. While in the older "hot" experiments the interactions were two-atom collisions, in the new "frozen gases" many-body interactions play a role [7, 69]. Secondly, mechanical effects could be observed now, due to the dipolar forces [31, 61, 104], as well as the angular dependence of the dipole-dipole interaction [21]. The dipole forces lead to spontaneous plasma formation for high densities of cold atoms [61], and also the other way round: starting from an ultracold plasma, Rydberg atoms can be spontaneously formed [70]. Such an ultracold plasma [82] is a new and exotic type of matter and could give insights in plasma’s that occur naturally in astrophysical systems. Another possibility is that two cold Rydberg atoms can form an enormous molecule [36, 18, 26, 30, 11].

The already mentioned dipole-blockade proposal [46, 65] has received a lot of interest. Dipole-blockade is the inhibition of optical transitions from ground state atoms to Rydberg states due to the presence of a nearby Rydberg atom. This dipole blockade effect is observed by several groups [92, 86, 62, 98, 49, 103]. It is suggested that using oscillating fields instead of static fields to tune the interaction into resonance has some benefits for quantum information processing [4, 17]. However, all these experiments are performed with randomly organized "frozen" gases, without direct control over the interatomic distances. In this thesis we aim for interactions over a fixed and controlled distance. During the final stages of writing this thesis two groups achieved a large amount of control of resonant dipole-dipole interaction [94, 33], where coherent evolution was observed between two Rydberg atoms fixed in space.

Quantum information with dipole-interacting particles is also investigated in other fields of physics. For example, molecules can be used [41] or self-assembled quantum dots, instead of real atoms [20]. In nanophotonics, coherent dipole-dipole coupling between carefully placed polarizable plasmonic nanoparticles is pursued as a tool to create ultrasmall optical circuits [76, 19, 40, 52]. Resonant dipole-dipole interactions even occur in biological systems: in bacterial light harvesting complexes, responsible for photosynthesis, resonant dipole-dipole interactions mediate the ultrafast energy flow [95, 39, 28, 59]. Dipole-dipole interactions between fluorophores, as first described by Förster [32], are now a workhorse tool in biological imaging to measure nanoscale distances [47]. We believe that our research might help in understanding these complex biological systems.

In most of these systems the dipole moments are so small that the interaction can only be observed if the separation of the dipoles is at a nanometer scale. Moreover, the interactions typically occur on a femtosecond timescale. These properties make these experiments technologically very challenging. With the ability to control the distance between Rydberg atoms, which have an interaction over a distance of tens of micrometers and an interaction time of several microseconds, the study of coherent dipole-dipole interaction is more feasible.

1.4 This thesis

In the experiments described in this thesis we use Rydberg states between \( n = 40 \) and 50 which have dipole moments of the order of \( 1000a_0e \) and radiative lifetimes around
100 µs. The dipole-dipole interaction time is then typically 15 µs for a distance of 40 µm. The gas-phase atoms must have such a low speed that they move only a small fraction of their distance (∼40 µm) during the interaction time (∼20 µs). To achieve such low speeds (≪2 m/s) we use laser-cooling of the ground-state atoms prior to the experiments. Once we have prepared a cold cloud of ground state atoms - in a magneto-optical trap - we excite atoms to two different Rydberg states with the use of two separate lasers. Each laser is focused in the cloud to create a narrow region of Rydberg atoms (∼15 µm in diameter). Rydberg atoms from one volume are allowed to interact resonantly with atoms in the other volume, tens of micrometers away, for a controlled amount of time, typically several microseconds, after which they are detected.

In the following chapter, chapter 2, we will provide the theoretical background of the experiments described in chapters 4 - 7. We describe calculations of experimental parameters, such as the energy levels of the Rydberg states in an electric field, needed to find an appropriate dipole-dipole resonance, and relevant transition dipole moments. The analytic version of this is a new generalization of the Gordon expression [12]. Furthermore we describe simulations, which provide basic understanding of the experiments.

In chapter 3 we describe the experimental setup we have build for our experiments, and we investigate some of its properties, such as the density in the cold atom cloud, the size of the Rydberg volumes and the homogeneity of the electric field.

In chapter 4 we present a novel technique that we developed, with which we can simultaneously detect the quantum state of the Rydberg atoms as well as their position. The technique combines state-selective field ionization to deduce the principal quantum number and time-of-flight of the ionization products (electrons) to deduce the original position of the atoms with an accuracy of about 10 µm.

Chapter 5 is in our view the most important chapter of this thesis. It describes the first realization of dipole-dipole interaction between Rydberg atoms in separate volumes. The resonant interaction 41d+49s ↔ 42p+49p is observed for volume separations up to 50 µm. We measured the amount of interaction products as a function of static electric field, volume separation and time. The data fit the extensive many-body simulations.

In the last two chapters we investigated the possibility to manipulate the dipole-dipole interaction 41d+49s ↔ 42p+49p with radio-frequency fields. Instead of using a static electric field, the interaction can be tuned into resonance with an RF-field. In this case multiple resonances are observed due to multi RF-photon transitions. In chapter 6 we show that the dipole-dipole interaction can be switched on and off by rapidly switching the field. Secondly, we show that RF-fields can be used to perform sub-MHz spectroscopy on the involved Rydberg states and we observe strong AC-Stark shifts and multi-photon transitions with up to 5 photons.

In chapter 7 we use an RF-field to demonstrate that the interacting atoms are coherent for at least 0.6 µs, by making use of so-called Stückelberg oscillations. We present two different methods to calculate the interaction strength as a function of static field and RF-field. This calculation is essentially different for states with a linear Stark shift as for states with a quadratic Stark shift, which is the case for the angular momentum states involved here. The variations of interaction strength are in fact Stückelberg oscillations and the measured interaction strength fits the calculations very well. These calculations culminate in a rather unusual result: the generalized Bessel function.
Calculations of Rydberg wavefunctions and Stark shifts enable us to choose a suitable resonant dipole-dipole transition for our experiments: $41d + 49s \leftrightarrow 42p + 49p$ is resonant at a field of only $\sim 0.4 \text{ V/cm}$ and the relevant transition dipole moments are of the order of $\sim 1000 \text{ a.u.}$ Both the calculation of the wavefunctions as well as the Stark shift are covered extensively in this chapter. Simulations of dipoles distributed over two parallel elongated volumes show that the short-time energy transfer probability scales like $P \propto \frac{t^2}{d^5}$, with $d$ the distance between the volumes. In a simulation of excitation diffusion in a randomly organized system we observe localization resulting from the dipole blockade effect.
2.1 Introduction

This chapter describes the theoretical background and the calculations we have performed. They both provide us with physical insight as well as the necessary experimental parameters. The main goal was to find a suitable set of states for dipole-dipole interaction among Rydberg states. We aimed for a strong interaction, allowing us to measure at large atomic distances. To find resonant interactions, one needs to know the exact energies of the atoms as a function of electric field, for which the wavefunctions of the atoms should be known. The calculation of the Rydberg atom wavefunctions are described in section 2.2. The potential for the electron is not exactly known, but by making use of the experimentally known binding energies an approximate wavefunction can be obtained. We present two methods, one numerical and one analytical solution of the Schrödinger equation. The following section, section 2.3, deals with the Stark effect: the shift of atomic energy levels in an electric field. The Stark shift can be calculated by means of perturbation theory; in contrast to the situation with lower excited states, this perturbation couples many states and the calculation involves quite large matrices.

In section 2.4 dipole-dipole interactions are explained. Here we also pick a suitable resonant transition, that we will use in our experiments. The transition of our preference is 41d+49s ↔ 42p+49p. For this transition we calculated the resonance fields (0.3807 V/cm and 0.4112 V/cm) and the transition dipole moments of both involved atomic transitions 41d ↔ 42p and 49s ↔ 49p. Both dipole moments are about 1000 $a_0 e$, leading to a dipole-dipole interaction strength of $V_{dd} = 2\pi 35$ kHz at a particle distance of 40 $\mu$m.

Table 2.1: CODATA ("Committee on Data for Science and Technology") values of atomic units (a.u.) from [68], with $\hbar$ the reduced Planck constant, $\mu_B$ the Bohr magneton and $\epsilon_0$ the electric constant.

<table>
<thead>
<tr>
<th>Name</th>
<th>Origin</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>length</td>
<td>Bohr radius $a_0$</td>
<td>0.529 177 208 59(36)$\cdot 10^{-10}$ m</td>
</tr>
<tr>
<td>time</td>
<td>$\hbar/E_h$</td>
<td>2.418 884 326 505(16)$\cdot 10^{-17}$ s</td>
</tr>
<tr>
<td>mass</td>
<td>electron mass $m_e$</td>
<td>9.109 382 15(45)$\cdot 10^{-31}$ kg</td>
</tr>
<tr>
<td>energy</td>
<td>Hartree energy $E_h = -\frac{e^2}{4\pi\epsilon_0 a_0}$</td>
<td>4.359 743 94(22)$\cdot 10^{-18}$ J</td>
</tr>
<tr>
<td>charge</td>
<td>elementary charge $e$</td>
<td>1.602 176 487(40)$\cdot 10^{-19}$ C</td>
</tr>
<tr>
<td>electric dipole moment</td>
<td>$a_0 e$</td>
<td>8.478 352 81(21)$\cdot 10^{-30}$ C m</td>
</tr>
<tr>
<td>electric field</td>
<td>$E_h/a_0 e$</td>
<td>5.142 206 32(13)$\cdot 10^9$ V/cm</td>
</tr>
<tr>
<td>magnetic dipole moment</td>
<td>$2\mu_B = \hbar e/m_e$</td>
<td>1.854 801 830(46)$\cdot 10^{-27}$ J/G</td>
</tr>
<tr>
<td>magnetic field</td>
<td>$\hbar/a_0^2 e$</td>
<td>2.350 517 382(59)$\cdot 10^9$ G</td>
</tr>
</tbody>
</table>

The last section 2.5 presents some simulations with dipole-dipole interacting atoms. In the experiments described in this thesis, we don’t have two single Rydberg atoms, but two very thin and long cylinder-like volumes with tens of Rydberg atoms. Straightforward two-particle simulations predict the experimental data from chapter 5 quite well. Furthermore we analytically derive a scaling law for the transition probability for short times as a function of distance $d$: $P \sim t^2/d^5$. Another simulation we performed describes excitation diffusion through dipole-dipole interaction. This simulation compares the diffusion in an ordered lattice of particles to a randomly organized particle configuration, where the latter
2.2 Rydberg wavefunctions

Table 2.2: Energy conversions used in this thesis. The full precision and uncertainty can be found in [68].

<table>
<thead>
<tr>
<th></th>
<th>MHz</th>
<th>cm⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 MHz</td>
<td>1 MHz</td>
<td>3.335 641·10⁻⁵ cm⁻¹</td>
</tr>
<tr>
<td>1 cm⁻¹</td>
<td>2.997 925·10⁴ MHz</td>
<td>1 cm⁻¹</td>
</tr>
<tr>
<td>1 eV</td>
<td>2.417 989 5·10⁸ MHz</td>
<td>8.065 545·10³ cm⁻¹</td>
</tr>
<tr>
<td>1 a.u.</td>
<td>6.579 683 920 7·10⁹ MHz</td>
<td>2.194 746 313 7·10⁵ cm⁻¹</td>
</tr>
<tr>
<td>1 J</td>
<td>1.509 190 5·10²⁷ MHz</td>
<td>5.034 117·10²² cm⁻¹</td>
</tr>
</tbody>
</table>

Table 2.2: continued

<table>
<thead>
<tr>
<th></th>
<th>eV</th>
<th>a.u.</th>
<th>J</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 MHz</td>
<td>4.135 667·10⁻⁹ eV</td>
<td>1.519 829 846 0·10⁻¹⁰ a.u.</td>
<td>6.626 069·10⁻²⁸ J</td>
</tr>
<tr>
<td>1 cm⁻¹</td>
<td>1.239 841 9·10⁻⁴ eV</td>
<td>4.556 335 252 8·10⁻⁶ a.u.</td>
<td>1.986 446·10⁻²³ J</td>
</tr>
<tr>
<td>1 eV</td>
<td>1 eV</td>
<td>3.674 932 5·10⁻² a.u.</td>
<td>1.602 176 5·10⁻¹⁹ J</td>
</tr>
<tr>
<td>1 a.u.</td>
<td>27.211 384 eV</td>
<td>1 a.u.</td>
<td>4.359 744·10⁻¹⁸ J</td>
</tr>
<tr>
<td>1 J</td>
<td>6.241 510·10¹⁸ eV</td>
<td>2.293 713·10¹⁷ a.u.</td>
<td>1 J</td>
</tr>
</tbody>
</table>

case shows an interesting result: a localization effect which is due to a dipole-blockade mechanism.

In this thesis atomic units are used unless stated otherwise. Atomic units form a convenient system of units in atomic physics; all quantities are measured relative to the properties of atomic hydrogen. In practice it means that the values for the electron charge, the electron mass, the reduced Planck constant are all set to unity as well as $1/4\pi\varepsilon_0$ and $2\mu_B$. This leads to a significant simplification of all relevant equations.

In table 2.1 the conversion factors from atomic units to SI units are given for a selection of quantities relevant in this thesis. Values are taken from the ‘2006 CODATA recommended values’ as published in [68].

The energy unit Joule is usually impractical for atomic physics. Apart from atomic units we often use MHz or cm⁻¹, also eV are common. In table 2.2 some energy conversions are given.

2.2 Rydberg wavefunctions

Alkali atoms are effectively one-electron atoms, just as hydrogen. Especially when the electron of the atom is in a highly excited state, or Rydberg state, there are many similarities. For rubidium in its ground state we have a nucleus of charge +37, and the first 36 electrons around it form a very stable configuration around the nucleus, as in the noble gas krypton, where all electron shells up to 4p are filled. The last electron is on his own in a new shell, the 5s state. When the atom is (laser) excited to a Rydberg state, this single electron is moved to a shell with a much higher principal quantum number $n$. Already in the 5s ground state, but especially in the higher excited states, we can consider the inner part of the atom as a point particle with charge +1, equivalent with the hydrogen nucleus. Therefore we will first discuss the hydrogen wavefunctions, which can be cal-
Theoretical background

culated exactly (subsection 2.2.1). Then we will make some small adjustments for the wavefunctions that enter the core region of the atom: the low angular momentum states. These wavefunctions are not exact and we will discuss two different methods to calculate these wavefunctions, an analytical and a numerical method (resp. subsections 2.2.2 and 2.2.3). The last part of this section describes how the expectation values of powers of the electron position are obtained (subsection 2.2.4). The general expression for these expectation values enables us to calculate the interaction with a static electric or magnetic field as well as with electromagnetic radiation. Especially the interaction with an electric field is important, because this leads to an energy shift of the states resulting in multiple resonances of the dipole-dipole interaction. The expectation values will also be necessary to calculate the interaction strength between two Rydberg atoms.

2.2.1 Hydrogen wavefunctions

The potential for the electron is the Coulomb potential \[ V = -\frac{1}{r} \] due to the attraction to the nucleus. To compute the wavefunctions one needs to solve the time-independent Schrödinger equation

\[
-\frac{1}{2} \nabla^2 \psi(r) - \frac{1}{r} \psi(r) = E \psi(r).
\]

(2.1)

Given the spherically symmetric potential, we can use spherical coordinates and separate the Schrödinger equation in a radial and an angular part:

\[
\psi(r) = R(r) Y_{\ell m}(\theta, \phi).
\]

(2.2)

The spherical harmonics \( Y_{\ell m}(\theta, \phi) \) are independent of the exact potential as long as it is spherically symmetric. The solution of the angular part of the Schrödinger equation is given by [12]

\[
Y_{\ell m}(\theta, \phi) = \sqrt{\frac{(2\ell + 1)(\ell - m)!}{4\pi(\ell + m)!}} P_{\ell}^m(\cos \theta) e^{im\phi},
\]

(2.3)

with \( P_{\ell}^m \) the associated Legendre function [2]. From boundary conditions for \( \theta \) and \( \phi \) follows \( \ell \geq 0 \) and \( |m| \leq \ell \). \( \ell \) is the orbital angular momentum quantum number and \( m \) is the magnetic quantum number.

For the radial part of the Schrödinger equation it is practical to rewrite \( u(r) = rR(r) \) and we obtain

\[
\left( -\frac{1}{2} \frac{d^2}{dr^2} + \frac{\ell(\ell + 1)}{2r^2} - \frac{1}{r} \right) u(r) = Eu(r).
\]

(2.4)

The solutions [12]* are in the form of an exponent of \( r \), a power of \( r \) and a generalized Laguerre polynomial \( L_n^\ell \) [2]

\[
R_{\ell n}(r) = \sqrt{\left(\frac{2}{n}\right)^\ell \frac{(n - \ell - 1)!}{2n(n + \ell)!}} e^{-r/n} \left(\frac{2r}{n}\right)^\ell L_{2\ell+1}^{2\ell+1}\left(\frac{2r}{n}\right)
\]

(2.5)

*Note that Bethe and Salpeter [12] use a different definition of the generalized Laguerre polynomial. We use here the Abramowitz and Stegun [2] definition, resulting in a slightly different expression for \( R_{\ell n} \).
and the energy is
\[ E_n = -\frac{1}{2n^2}, \]
with \( n \) the principal quantum number. The orbital angular momentum quantum number \( \ell \) can not get larger than \( n - 1 \), or \( 0 \leq \ell \leq n - 1 \). The \( n^{-2} \) dependence of the binding energy (Eq. 2.6) was first noted in the 1880’s by Johann Balmer and Johannes Rydberg and further developed by Niels Bohr in 1913 [105]. The associated wavenumber of a photon that just ionizes the atom is given by the Rydberg constant \( R_y \). It is equivalent to half an atomic unit of energy (in units of \( \text{cm}^{-1} \)), given in table 2.2. So the binding energy can also be written as \( E_n = -R_y/n^2 \).

2.2.2 Analytical wavefunctions for alkali atoms

The adjustments for alkali atoms heavier than hydrogen lie in the low angular momentum states. Of all states, these low-\( \ell \) states have the largest probability near the core of the atom, and here, the nucleus is not fully screened by the core electrons. Also electron-electron interaction is largest close to the core. The potential near the core is not exactly known, but gives rise to a change in energy, which is known experimentally. Already noted by Rydberg, the binding energy Eq. 2.6 is modified by replacing \( n \) by \( n^* = n - \delta_{n\ell j} \)
\[ E_{n\ell j} = -\frac{1}{2(n - \delta_{n\ell j})^2}; \]
\( \delta_{n\ell j} \) is called the quantum defect. It depends mainly on \( \ell \), but the spin orbit coupling gives rise to a small dependence on \( j \); the number changes only slightly with \( n \). The quantum defects we use in our calculations are based on measurements in 2003 and 2006 [60, 38, 3]. They are given by
\[ \delta_{n\ell j} = \delta_0 + \frac{\delta_2}{(n - \delta_0)^2}, \]
where \( \delta_0 \) and \( \delta_2 \) depend on \( \ell \) and \( j \). The experimentally obtained values for \( \delta_0 \) and \( \delta_2 \) for \(^{85}\text{Rb} \) are given in table 2.3.

\begin{table}[h]
\centering
\begin{tabular}{|l|c|c|}
\hline
State & \( \delta_0 \) & \( \delta_2 \) \\
\hline
ns\(_{1/2}\) & 3.1311804(10) & 0.1784(6) \\
np\(_{1/2}\) & 2.6548849(10) & 0.2900(6) \\
np\(_{3/2}\) & 2.6416737(10) & 0.2950(7) \\
n\(_{3/2}\) & 1.34809171(40) & -0.60286(26) \\
n\(_{5/2}\) & 1.34646572(30) & -0.59600(18) \\
nf\(_{5/2}\) & 0.0165192(9) & -0.085(9) \\
nf\(_{7/2}\) & 0.0165437(7) & -0.086(7) \\
n\(_{g7/2}\) & 0.00405(6) & \\
\hline
\end{tabular}
\caption{The \(^{85}\text{Rb} \) quantum defects \( \delta_{n\ell j} = \delta_0 + \delta_2/(n - \delta_0)^2 \) from [60, 38, 3].}
\end{table}

The radial wavefunction is also modified by replacing \( n \) by \( n^* \) in equation 2.5. However, the Laguerre polynomials need an integer bottom index. Therefore, we also replace...
\( \ell \) by \( \ell^* = \ell - \delta_{n lj} + I(\ell) \) \cite{10, 54}. This modifies the radial potential in such a way that the binding energy is an exact solution of the Schrödinger equation again. In this approach the modification of the potential is proportional to the centrifugal potential, keeping the changes localized near the core. \( I(\ell) \) is an integer fixing the number of radial nodes, for which we have the restrictions \( \delta_{n lj} - \frac{1}{2} < I(\ell) \leq n_{\text{min}} - \ell - 1 \) with \( n_{\text{min}} \) the principal quantum number of the ground state. For rubidium we use for \( I(\ell) \) the nearest integer below or equal to \( \delta_{n lj} \), which gives the best agreement between the calculated and experimentally known value for the lifetime of the 5s – 5p transition. We have

\[
R_{n^* \ell^*}(r) = \sqrt{\left( \frac{2}{n^*} \right)^3 \left( n^* - \ell^* - 1 \right)!} \frac{2r^\ell}{n^*} \frac{L_{n^* - \ell^* - 1}^{2\ell + 1}}{(n^*)^\ell}.
\]  

(2.9)

The angular part of the wavefunction is equal to the one for hydrogen (Eq. 2.3), i.e. not \( \ell^* \), but just \( \ell \) should be used.

We have depicted the radial wavefunctions of \( n = 9 \) in figure 2.1. Note that the number of radial zero-crossings is given by \( n^* - \ell^* - 1 \). Furthermore it is visible that for higher \( \ell \) the wavefunction starts further away from the core, due to the centrifugal part of the potential (the second term in equation 2.4).

### 2.2.3 Numerical calculation of the radial wavefunctions

A numerical method to calculate the radial wavefunctions of heavy alkali atoms is described by Zimmerman et al. \cite{109}. Here the Numerov method is used to numerically solve the radial Schrödinger equation (Eq. 2.4). Numerov’s method is a numerical method to solve second-order differential equations, where the first-order term does not appear, like we have here. Zimmerman’s method uses a logarithmic scaling for \( r \), which is most efficient, because the wavefunction oscillates more and more when going inward. The very inner part of the potential and of the wavefunction are unknown, but the wavefunction can be calculated using the semi-empirically known energy (Eq. 2.7) and simply the hydrogen potential – the Coulomb potential and the centrifugal barrier, respectively the third and the second term in equation 2.4. The calculation is started from the outside and uses the fact that the outer part of the wavefunction (beyond the classical outer turning point \( r_{\text{out}} = 2n^2 \)) decreases exponentially. The starting point for the calculation is at \( r_s = 2n(n + 15) \) and from there the wavefunction is calculated step-by-step by going inwards with a logarithmic step size of \( h = 0.001 \) until the classical inner turning point \( r_{\text{in}} = n^* \left( n^* - \sqrt{\left( n^* \right)^2 - (\ell + 1/2)^2} \right) \) is reached.

Specifically, with \( x = \ln r \) and \( X = \sqrt{r} R \) the radial Schrödinger equation (Eq. 2.4) becomes

\[
\frac{d^2 X}{dx^2} = g(x)X,
\]

(2.10)

where

\[
g(x) = 2e^{2x} \left( -\frac{1}{e^x} + \frac{1}{2(n^*)^2} \right) + (\ell + \frac{1}{2})^2
\]

(2.11)
2.2 Rydberg wavefunctions

\begin{align*}
\ell = 0 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15 \\
\ell = 1 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15 \\
\ell = 2 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15 \\
\ell = 3 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15 \\
\ell = 4 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15 \\
\ell = 5 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15 \\
\ell = 6 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15 \\
\ell = 7 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15 \\
\ell = 8 & \quad -0.15 \quad -0.1 \quad -0.05 \quad 0 \quad 0.05 \quad 0.1 \quad 0.15
\end{align*}

**Figure 2.1:** The radial wavefunctions of \( n = 9 \) for rubidium-85. We have plotted \( rR_{\ell} \) (Eq. 2.9) versus the radius in units of the Bohr radius \( a_0 \), such that we get the total wavefunction for the whole spherical surface with radius \( r \). The insets show a small part of the analytical wavefunction together with the numerically calculated wavefunction in red, where for small \( \ell \) a small phase difference is visible.

and the Numerov algorithm is

\[
X_{i+1} = \frac{(10T_i + 2)X_i + (T_{i-1} - 1)X_{i-1}}{1 - T_{i+1}},
\]

where we used \( T_i = (h^2/12)g_i \). The index \( i \) is the number of the step \( x_i = \ln r_s - ih \). The normalization constant \( N \) is obtained by

\[
N^2\hbar \sum_i X_i^2r_i^2 = 1.
\]

The resulting wavefunctions \( \Xi = NX \) resemble the analytical wavefunctions quite well, especially for higher \( \ell \) states. In figure 2.1 a small phase difference is visible between the
numerically calculated and the analytical wavefunction of the p and d state, due to the different assumptions of both methods near the atom core. In the analytical version the phase of the radial wavefunction is chosen such that the function nicely goes to zero at \( r = 0 \), whereas the numerical calculation starts on the outside and stops abruptly near the core. It is not clear which method reflects the reality better, but the numerical method is used most often. In most calculations that follow in this thesis, we have used the numerical method to calculate the wavefunctions and expectation values.

### 2.2.4 Expectation values

The wavefunctions are used for the calculation of expectation values. The expectation value of some parameter \( A \) is defined as \( \langle \psi | A | \psi \rangle \). Often, \( \psi \) is written as a composition of several eigenfunctions \( \psi_j \) and the atom can make a transition from one eigenstate \( \psi_i \), the initial state, to another state, the final state \( \psi_f \). Here we discuss the expectation values of powers of the electron position.

\[
\langle \psi_f | r^d | \psi_i \rangle = \int \psi_f^*(r) r^d \psi_i(r) \, dr. \tag{2.14}
\]

We separate this integral in an angular and a radial part. For the numerically calculated radial wavefunctions the expectation value becomes a simple summation:

\[
X_{n\ell,n'\ell'} = \langle n'\ell' | r^d | n\ell \rangle = \hbar \sum_j \Xi_j \zeta_j^{r^2+d}, \tag{2.15}
\]

where the accents refer to the final state. For the analytical wavefunction we get for the radial part

\[
R_{i,f} = \langle n_f\ell_f | r^d | n_i\ell_i \rangle = \int R_{n_f\ell_f} r^{d+2} R_{n_i\ell_i} \, dr. \tag{2.16}
\]

Inspired by [54, 74] we have worked out this integral as

\[
\frac{1}{2^{1+d}} \sqrt{\frac{(n_i^*-\ell_i^*-1)! (n_f^*+\ell_f^*-1)!}{(n_i^*+\ell_i^*)!(n_f^*+\ell_f^*)!}} \left( \frac{2}{n_i^*+n_f^*} \right)^{\ell_f^*+2+d} (n_i^*)^{\ell_f^*+1+d} (n_f^*)^{\ell_f^*+1+d} \left( \frac{n_i^*+\ell_i^*}{n_f^*+\ell_f^*} \right) \left( \frac{n_f^*+\ell_f^*}{n_i^*+\ell_i^*} \right) \times (\ell_f^*+\ell_f^*+2+2d)! F_2\left( \ell^*_f+\ell^*_f+3+2d; -n_i^*+\ell_i^*+1, -n_f^*+\ell_f^*+1; 2\ell^*_f+2, 2\ell^*_f+2; \frac{2n_f^*}{n_i^*+n_f^*}, \frac{2n_i^*}{n_i^*+n_f^*} \right), \tag{2.17}
\]

where \( F_2 \) is the Appell hypergeometric function of the second kind [29]

\[
F_2(\alpha; \beta, \beta'; \gamma, \gamma'; x, y) = \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} \frac{(\alpha)_{p+q} (\beta)_p (\beta')_q}{p! q!} x^p y^q. \tag{2.18}
\]

The symbol \( (\alpha)_p = \Gamma(\alpha + p)/\Gamma(\alpha) \) is the Pochhammer symbol. For the upper limit of the summations in the Appell function in 2.17 we use \( p = n_f^* - \ell_f^* - 1 \) and \( q = n_f^* - \ell_f^* - 1 \) (the factors \( \beta \) and \( \beta' \) are zero for larger \( p \) and \( q \)). This result is in fact a generalization of

\[\text{Note that } \sum_j R_j \zeta_j^{r^2+d} \Delta r_j \text{ can be rewritten as } \hbar \sum_j \Xi_j \zeta_j^{r^2+d}. \]
the Gordon formula \[12\] and this Appell hypergeometric function is a generalization of the hypergeometric function.

The angular part of equation 2.14 is given by \[27\]

\[
A_{i,f} = \sqrt{\frac{4\pi}{3}} \int_0^\infty \sin(\theta) d\theta \int_0^{2\pi} d\phi \ Y_{\ell_f m_f}(\theta, \phi) Y_{1q}(\theta, \phi) Y_{\ell_i m_i}(\theta, \phi). \tag{2.19}
\]

Note that \(Y_{1q}(\theta, \phi)\) originates from the conversion to Cartesian coordinates

\[
\sqrt{\frac{4\pi}{3}} Y_{10}(\theta, \phi) = \cos \theta = \hat{z}, \tag{2.20}
\]

\[
\sqrt{\frac{4\pi}{3}} Y_{1\pm 1}(\theta, \phi) = \mp e^{\pm i\phi} \frac{\sin \theta}{\sqrt{2}} = \mp \hat{x} \pm i \hat{y}. \tag{2.21}
\]

In other words \(q\) is related to the polarization of the external electromagnetic wave; \(q = \pm 1\) for circularly polarized radiation and \(q = 0\) for linearly polarized radiation. This light can be either light that pumps the transition, or light that is spontaneously emitted from the atom that makes the transition from the initial to the final state. The integral Eq. 2.19 can be written in terms of a Wigner 3j-symbol

\[
A_{i,f} = (-1)^{\ell_f - m_f} \sqrt{\max(\ell_i, \ell_f)} \left( \begin{array}{ccc} \ell_f & 1 & \ell_i \\ -m_f & q & m_i \end{array} \right); \tag{2.22}
\]

this Wigner 3j-symbol is zero unless \(\Delta \ell = \ell_f - \ell_i = 0, \pm 1\) and \(\Delta m = m_f - m_i = q\).

Due to spin-orbit interaction we get a splitting of the \(\ell\) states into states with the new quantum number \(j\), which denotes the total electronic angular momentum. Possible values for \(j\) run from \(\ell - s\) to \(\ell + s\) in steps of 1. The angular part of the wavefunction becomes

\[
|\ell jm_j\rangle = \sum_i C_i Y_{\ell m_i}(\theta, \phi), \tag{2.23}
\]

with \(C_i\) the Clebsch-Gordan coefficients. The angular integral, now including the spin-orbit interaction, can be worked out as follows

\[
\mathcal{A}_{i,f} = (-1)^{\ell_f - m_f + 1/2} \sqrt{\max(\ell_i, \ell_f)} \sqrt{(2j_i + 1)(2j_f + 1)} \left( \begin{array}{ccc} \ell_f & j_f & s \\ j_i & \ell_i & 1 \end{array} \right) \left( \begin{array}{ccc} j_i & 1 & j_f \\ m_i & q & -m_f \end{array} \right), \tag{2.24}
\]

where \(m_i\) and \(m_f\) refer now to the value of \(m_j\) of the initial, respectively final state. Equivalently we have \(\Delta j = 0, \pm 1\) and \(\Delta m = q\) from the Wigner 3j-symbol. The array between the curly braces is the Wigner or Racah 6j-symbol. This 6j-symbol is nonzero for \(\Delta \ell = 0, \pm 1\).

As a test case of both methods we calculate the spontaneous emission rate of the 5p state. The spontaneous emission rate is given in a.u. by \[64\]

\[
\Gamma_{i,f} = \frac{4}{3} \alpha^3 (\Delta E_{i,f})^3 |\langle \psi_f | \mathbf{r} | \psi_i \rangle|^2, \tag{2.25}
\]

where \(\alpha\) is the fine structure constant. The final state is in this case the ground state 5s. For \(\Delta E_{i,f}\) we use \(1/\lambda\) with the known wavelength of the transition; \(\lambda = 780\ \text{nm}\) for the
§Note that if $A_{5p,5s}$ (Eq. 2.24) is used, the total spontaneous emission rate is the sum over both $\Gamma_{i,f}$'s for the two possible $m_j$ states of the 5s state. This gives the same result.

5P_{3/2} state and $\lambda = 795$ nm for 5P_{1/2}. The angular integral $A_{5p,5s}$ (Eq. 2.22) is $1/\sqrt{3}$ for all three possibilities of the magnetic quantum number $m_l$ of the 5p state§. For the radial part we can compare the analytical version $R_{5p,5s}$ (Eq. 2.17) with the numerical one $X_{5p,5s}$ (Eq. 2.15). As a result we get for the 5P_{1/2} state a lifetime of $\tau_{1/2} = 1/\Gamma_{5p,5s_{1/2}} = 30.9$ ns if we use the analytical calculation and $\tau_{1/2} = 26.1$ ns if we use the Numerov method. For the 5P_{3/2} state we get $\tau_{3/2} = 29.7$ ns analytically and $\tau_{3/2} = 24.8$ ns numerically. All numbers are a few nanoseconds off from the experimentally known values $\tau_{1/2} = 27.70(4)$ $\tau_{3/2} = 26.24(4)$ [99]. The difference is due to the different phase of the radial wave-functions, visible in figure 2.1, as already mentioned at the end of subsection 2.2.3. For high lying Rydberg states, the difference becomes much smaller; we will see in table 2.5 that the difference in the transition dipole moments is only about 0.6% for $n$ between 40 and 50. This is because the high-$n$ states are much less influenced by the – unknown – properties of the core region.

In the next section, section 2.3, we use the expectation value of $z$ to calculate the energy levels in the presence of an externally applied electric field in the $z$-direction. In section 2.4 we calculate the transition dipole moments $a_0 e \langle \psi_f | r | \psi_i \rangle$, which are the subjects in the resonant dipole-dipole interaction.

2.3 Rydberg atoms in a static electric field

In Rydberg experiments the coupling with an electrical field is usually important, because it is very strong. Due to this coupling the energy levels shift, which is called the Stark effect. This shift enables for resonances in dipole-dipole interactions. First we will give a general description of the Stark effect (subsection 2.3.1). The energy shift in rubidium can be calculated with perturbation theory (subsections 2.3.2 and 2.3.3). In the last subsection we describe an experiment we performed, which maps out a part of a Stark map, making use of dipole-dipole interacting atoms (subsection 2.3.4).

2.3.1 The Stark effect

The coupling with the external field occurs for dipoles. A dipole $\mu$ in an external electric field $F$ has an interaction potential of

$$V_{\text{dip}} = -\mu \cdot F = -\mu_z F,$$

where we have chosen the field to be in the $z$-direction. The dipole moment of an atom $\mu$ is in fact the expectation value of $r$ times the charge of the electron $-e$, or, if $r$ is in atomic units, $\mu = -a_0 e \langle r \rangle$ and $\mu_z = -a_0 e \langle z \rangle$, with $a_0$ the Bohr radius. For pure angular momentum states the expectation value of $r$ is always zero. However, states with a well defined angular momentum are not stable in an electric field. The atomic states are coupled to each other by means of transition dipole moments. The transition dipole
moments are given by $\mu_{1,2} = -a_0 e \langle \psi_1 | r | \psi_2 \rangle$. This second-order coupling (the second-order Stark effect) gives rise to a quadratic behavior of the energy levels.

$$\Delta E = -\frac{1}{2} \alpha F^2,$$

with $\alpha$ the polarizability. This polarizability can be calculated with perturbation theory described in the next subsection.

For states that are degenerate at zero field, the Stark effect will cause a splitting of the states. The energy shift of these states is linear, in other words, these states have a permanent dipole moment. In hydrogen this is the case for all angular momentum states with the same principal quantum number $n$ and in alkali’s for all higher angular momentum states with equal $n$. For the case of hydrogen it is convenient to replace the angular momentum quantum number $\ell$ by the parabolic quantum number $k$, which originates from the conversion to a parabolic coordinate system. The state $|nkm\rangle$ can be expressed as a linear superposition of $|n\ell m\rangle$ states \[72, 43\]

$$|nkm\rangle = \sum_\ell C_{k\ell} |n\ell m\rangle,$$

with the Clebsch-Gordan coefficient being

$$C_{k\ell} = (-1)^m \sqrt{2\ell + 1} \left( \begin{array}{ccc} \frac{1}{2}(n - 1) & \frac{1}{2}(n - 1) & \ell \\ \frac{1}{2}(m + k) & \frac{1}{2}(m - k) & m \end{array} \right).$$

The number $k$ runs from $n - |m| - 1$ to $-n + |m| + 1$ in steps of 2. The Stark shift of such a state in a field is given by \[12\]

$$\Delta E = \frac{3}{2} nkF.$$

So the quantum number $k$ is directly related to the permanent dipole moment $\mu = -\frac{3}{2} nk$ (in a.u.). In the heavier alkali atoms we will see the same linear effect in the high angular momentum states which are degenerate at zero field. However, there is no simple expression in terms of a the quantum number $k$. So for the case of heavier alkali’s it is most practical to use perturbation theory to calculate the energies in an electrical field.

### 2.3.2 Perturbation theory

Perturbation theory provides us with an approximate method to calculate a complicated quantum system in terms of a simpler one. The solution of the unperturbed Schrödinger equation is already known, namely we have the unperturbed energies $E_1, E_2, \ldots, E_N$ and the unperturbed wavefunctions $\psi_1, \psi_2, \ldots, \psi_N$ from section 2.2. We use these energies and wavefunctions and then we simply add the perturbation $H'$ to the unperturbed Hamiltonian $H_0$ (from Eq. 2.1)

$$H = H_0 + H' = \begin{pmatrix}
  E_1 & 0 & \ldots & 0 \\
  0 & E_2 & \ldots & 0 \\
  \vdots & \vdots & \ddots & \vdots \\
  0 & 0 & \ldots & E_N
\end{pmatrix} + \begin{pmatrix}
  H'_{11} & H'_{12} & \ldots & H'_{1N} \\
  H'_{21} & H'_{22} & \ldots & H'_{2N} \\
  \vdots & \vdots & \ddots & \vdots \\
  H'_{N1} & H'_{N2} & \ldots & H'_{NN}
\end{pmatrix}.$$
The matrix elements of the perturbation are $H'_{ij} = \langle \psi_i | H' | \psi_j \rangle$. The elements on the diagonal are the first-order perturbations, the off-diagonal matrix elements are the second-order perturbations. With this Hamiltonian we can calculate the energies of the perturbed states $E'$ and the new wavefunctions $\Psi'$ in terms of the old ones $\Psi$. We have the Schrödinger equation $HH' = E'\Psi'$ and we can solve it by diagonalizing the Hamiltonian. $E'$ is the set of eigenvalues and $\Psi'$ is the set of eigenfunctions, obtained from the matrix product of $C$, the set of eigenvectors, and $\Psi$, the set of the original wavefunctions. Specifically, we have

$$
\begin{pmatrix}
E_1 + H'_{11} & H'_{12} & \cdots & H'_{1N} \\
H'_{21} & E_2 + H'_{22} & \cdots & H'_{2N} \\
\vdots & \vdots & \ddots & \vdots \\
H'_{N1} & H'_{N2} & \cdots & E_N + H'_{NN}
\end{pmatrix}
\begin{pmatrix}
\psi'_1 \\
\psi'_2 \\
\vdots \\
\psi'_N
\end{pmatrix}
= 
\begin{pmatrix}
E'_1 & 0 & \cdots & 0 \\
0 & E'_2 & \cdots & 0 \\
\vdots & \vdots & \ddots & \vdots \\
0 & 0 & \cdots & E'_N
\end{pmatrix}
\begin{pmatrix}
\psi'_1 \\
\psi'_2 \\
\vdots \\
\psi'_N
\end{pmatrix}
$$

(2.32)

and

$$\Psi' = C\Psi = \begin{pmatrix}
c_{11}\psi_1 + c_{12}\psi_2 + \cdots + c_{1N}\psi_N \\
c_{21}\psi_1 + c_{22}\psi_2 + \cdots + c_{2N}\psi_N \\
\vdots \\
c_{N1}\psi_1 + c_{N2}\psi_2 + \cdots + c_{NN}\psi_N
\end{pmatrix}.$$  

(2.33)

### 2.3.3 Calculation of a Stark map

For the calculation of the energy of rubidium Rydberg levels in an external electric field the perturbation is $H' = zF$. The zero-field energies $E_1, E_2, \ldots, E_N$ are given by equation 2.7. All terms $H_{ij}'$ on the diagonal in the second matrix of equation 2.31 are zero, since we use the angular momentum wavefunctions to start with – from either subsection 2.2.2 or 2.2.3. The off-diagonal matrix elements are

$$H'_{ij} = F\langle \psi_i | z | \psi_j \rangle.$$  

(2.34)

The angular part of this integral is given by Eq. 2.24 with $q = 0$ (see Eq. 2.20). So we have only non-zero values for $\Delta m = 0$. Therefore, we only have to use states with the same value for $m$ in our calculation (in this case we work with $m_\ell$). Furthermore we have the selection rule $\Delta \ell = \pm 1$. At equation 2.24 we mentioned that $\Delta \ell = 0, \pm 1$, but parity considerations require odd $\Delta \ell$ for electric dipole transitions.

We need to work with quite a lot of states. For one principal quantum number $n$ and $m_\ell = 1/2$ we have $2n-1$ states – there are $n$ different $\ell$ states and each one is split in two $j$-sublevels, except for the $s$ state, which is not split. Secondly, coupling occurs between states several $n$-manifolds apart; we usually work with two or three manifolds above and below the state we’re interested in. For larger $\Delta n$ the coupling strengths become negligible. For example, if we’re interested in a state in the $n = 40$ manifold, we have a Hamiltonian of approximately $550 \times 550$ elements. Fortunately, most elements are zero, due to the selection rule $\Delta \ell = \pm 1$, which saves calculation time.

In figure 2.2 we depicted the result of such a calculation, a so-called Stark map. Depicted are the binding energies versus electric field around $n = 21$. Clearly visible are the fan-like structures of high angular momentum states for each $n$-manifold, due to the linear Stark effect. Note that these states are no longer pure angular momentum states, but they
are strongly mixed, like the \( k \)-states in hydrogen. The three lowest angular momentum states, \( s, p \) and \( d \), are clearly visible, due to their large quantum defect. These states experience a purely quadratic and therefore weak Stark shift until they come close to other states. The point where two fan-like structures – or manifolds – of subsequent \( n \) start to cross each other is given by the Inglis-Teller limit. It can easily be shown using the shift of the hydrogen \( k \)-states that this occurs at a field of approximately

\[
F_{\text{IT}} = \frac{1}{3n^5}.
\]  

(2.35)

When the external field is as strong as the Coulomb field from the core, the atom will be able to ionize. This occurs for a potential of \( V_{\text{ion}} = -2 \sqrt{F} \), depicted as a blue line in figure 2.2. With a binding energy of \(-1/2n^2\) we obtain an ionization field of

\[
F_{\text{ion}} = \frac{1}{16n^4}.
\]  

(2.36)

This is called the classical ionization limit. In quantum mechanics, states with positive \( k \)-values can live beyond this limit, when the \( k \)-states are not coupled (as in hydrogen) [35]. However, for rubidium, these \( k \)-states are coupled and they all have short lifetimes.

If the Stark map is examined closely, no two lines will be found that cross each other. Due to the coupling between all states the crossings are avoided crossings. This is however only true for a static field. If the field would be swept quickly over such a crossing the two levels now do cross each other and the coupling is "ignored". This is called the
diabatic case, opposed to the adiabatic case, where the eigenenergies are followed and the crossing is an avoided crossing, which occurs for a slow sweep of the field. This has implications for the method of State-selective Field Ionization (SFI), which uses the \( n \) dependence of the ionization field \( F_{\text{ion}} \) (Eq. 2.36) to detect the state of the atoms (see chapter 3, section 3.5). In this method the field is swept and the different \( n \)-states are detected subsequently when they ionize. Due to the (a)diabaticity, the exact ionization field depends on the sweep speed, which is examined in [9]. More about diabaticity and adiabaticity is explained in chapter 7, subsection 7.2.5.

### 2.3.4 Measurement of dipole-transitions to the manifold states

As an illustration of the Stark map we have performed a measurement, which shows the structure of the Stark-manifold, as first performed by [21]. In our setup we cannot resolve the energy of the manifold states with our laser, however, we can resolve the s- and d-states, especially at zero field, since they are the only allowed laser transitions (section 3.3.3). We make use of the resonant two atom transitions in rubidium like

\[
\begin{align*}
n d + n d &\leftrightarrow (n-2)m + (n+2)p, \quad (2.37a) \\
n d + n d &\leftrightarrow (n-1)m + (n+1)p, \quad (2.37b)
\end{align*}
\]

where the \( m \) refers to the manifold states. Both transitions are already close to resonant at zero field and if we scan the electric field, we see quite a number of manifold states. With these transitions we can now partly resolve the manifold states, after having created the d-states with our laser. The transitions are caused by dipole-dipole interaction, which is explained in the next section (section 2.4).

In the experiment we excite about 50 laser-cooled atoms up to the 32d-state, using one dye-laser beam (see chapter 3). We studied the process

\[
32d + 32d \leftrightarrow 34p + 30m \quad (2.38)
\]

as a function of an externally applied static electric field. We let the atoms interact for 3 \( \mu s \) in this field and afterward we detected the atoms, resolving the 34p state from the 32d state. In figure 2.3 we have depicted the fraction of 34p atoms (a fraction of the total number of atoms) versus the applied electric field.

In figure 2.3 we observe multiple peaks, which correspond well to the resonances shown in the upper panel of the figure. This gives us the trust that our Stark map calculation is accurate. The upper panel is constructed from a Stark map for \( m_j = \frac{1}{2} \) and shows the binding energy of the manifold states as well as the difference of the binding energies of the other involved states \( E_{32d} + E_{32d} - E_{34p} \), such that a resonance occurs of two lines cross. In reality more values of \( m_j \) can occur, which we did not take into account in the upper panel for clarity reasons.

In the calculation in the lower panel, we did take into account all relevant \( m_j \) values; with the limitation of \( \Delta m_j = 0, \pm 1 \) for each atom we obtain about 5000 resonances between 0 and 4 V/cm. The theoretical spectrum shown in the figure is a sum of Lorentzian peaks as given in equations 2.53 and 2.54. For the interaction strength, which determines the height and/or width of the peak, we used \( V = |\mu_A| |\mu_B| / R^3 \). This is a simplified form of the
2.3 Rydberg atoms in a static electric field

Dipole-dipole interaction, without angular dependence (section 2.4). $\mu_A$ is the transition dipole moment between $32d$ and $34p$ with the relevant $j$ and $m_j$ and $\mu_A$ is the transition dipole moment between $32d$ and the relevant $n = 30$ manifold state. To obtain a peak width that fits the measurement we used a distance of $R = 1.2 \mu$m. Secondly we adjusted the height of the total spectrum.

Despite all the simplifications in our model, the theoretical spectrum matches the measurement remarkably well in the lower panel of figure. The distance of 1.2 $\mu$m seems rather small; a more realistic number for the average inter-particle distance would probably lie between 5 and 10 $\mu$m. Due to the $1/R^3$ dependence of the interaction, the smaller distances will make a much stronger contribution. However, probably other broadening effects in the experiment play the largest role, like stray fields etc. (see section 3.4); the average particle distance is therefore strongly underestimated. The height of the total spectrum was adjusted by factor of 0.002, which is sensible since it is not correct to simply add up all 5000 peaks.

The experiment presented in figure 2.3 is a repetition of a measurement from [21], but the calculation we performed is much simpler. Secondly, we do now understand the peak at 0.4 V/cm, which is the resonance $32d+32d \rightarrow 34p + 30g$; at the time of [21] (2004) the g-quantum defect was not accurately known yet, it was measured in 2006 [3].
2.4 Dipole-dipole interactions between Rydberg atoms

Dipole-dipole interactions are one of the most important interactions between atoms or molecules. The Van der Waals forces are dipole-dipole interactions of induced dipoles; two atoms or molecules induce a dipole moment to each other and these dipoles will attract. Here we will discuss resonant dipole-dipole interactions. Two dipolar atoms (or molecules) can, next to attracting or repelling each other, exchange state – and transfer energy – through dipole-dipole interaction. This resonant energy transfer is sometimes called Förster Resonance Energy Transfer (FRET) [47], which occurs for example between molecules in biological systems [95, 39, 28, 59]. Dipole-dipole interaction can also occur between magnetic dipoles [55], but here we limit ourselves to electric dipoles.

For two atomic dipoles, schematically depicted in figure 2.4, we can construct the Hamiltonian from all six Coulomb interactions between both electrons and both cores.

\[
H = H_A + H_B + V_{dd},
\]

\[
V_{dd} = \frac{1}{R} \left( \frac{1}{|\mathbf{R} + \mathbf{r}_B|} - \frac{1}{|\mathbf{R} - \mathbf{r}_A|} + \frac{1}{|\mathbf{R} + \mathbf{r}_B - \mathbf{r}_A|} \right) \approx \frac{\mathbf{r}_A \cdot \mathbf{r}_B - 3(\mathbf{r}_A \cdot \mathbf{\hat{R}})(\mathbf{r}_B \cdot \mathbf{\hat{R}})}{R^3},
\]

(2.39)

where the Coulomb interactions between the core and its electron are contained in \( H_A \) and \( H_B \) and the interaction between the atoms is contained in \( V_{dd} \), the dipole-dipole interaction potential.

![Figure 2.4: Two atomic dipoles A and B schematically depicted as a red core, charge +1, and a blue electron.](image)

We use perturbation theory (subsection 2.3.2) to calculate the properties of the two atom system. The full quantum method would be to use the dipole-dipole interaction \( V_{dd} \) as an operator, operating on the combined wavefunction of the two atoms and the matrix elements of the perturbation would be \( \langle \psi'_A, \psi'_B | V_{dd} | \psi_A, \psi_B \rangle \). This method is described in [81] and can take into account multi-body interactions. Here we describe a simpler, semi-classical method, providing a basic understanding of the dynamics. This simplification is well justified for the low densities we have. We use the operator \( \mathbf{r} \) to calculate the dipole
2.4 Dipole-dipole interactions between Rydberg atoms

matrix elements $\mu$ per atom

$$\mu_A = \langle \psi_A' | r | \psi_A \rangle,$$  \hspace{1cm} \text{(2.40a)}

$$\mu_B = \langle \psi_B' | r | \psi_B \rangle.$$  \hspace{1cm} \text{(2.40b)}

Then we calculate the dipole-dipole interaction with

$$V_{dd} = \frac{\mu_A \cdot \mu_B - 3(\mu_A \cdot \hat{R})(\mu_B \cdot \hat{R})}{R^3}.$$  \hspace{1cm} \text{(2.41)}

So $V_{dd}$ is now simply a number, instead of an operator.

The $\mu$'s in equation 2.41 could be either permanent dipoles or transition dipoles; from first- respectively second-order perturbation theory (subsection 2.3.2). In other words, a dipole is a permanent dipole if $\psi'$ is equal to $\psi$ in equation 2.40, for different initial and final wavefunctions we have transition dipoles. Large permanent dipoles are for example the $k$-states in hydrogen – or the equivalent manifold states in heavier alkali's.

In this thesis we will mainly deal with transition dipoles, which can be transitions between any two states with $\Delta \ell = \pm 1$. These transition dipoles can be interpreted as oscillating dipoles. The atom is in a coherent superposition of e.g. the s- and p-state and the sum of these wavefunctions will have a dipole moment, while the separate pure angular momentum states do not. Due to the energy- and therefore phase-difference between these wavefunctions, the dipole moment will change direction with a frequency corresponding to this energy difference. Two of these oscillating dipoles can just as well dipole-interact. For both the permanent dipole as well as the transition dipole, $\mu$ scales like $n^2$, which results in a scaling law for the dipole-dipole interaction: $V_{dd} \sim n^4/R^3$.

2.4.1 A suitable dipole-dipole transition

In this section we describe how we have chosen a suitable dipole-dipole transition to use in our experiments, which was critical for the success of this work. We wanted our dipoles to be strong enough to be able to work at large distances. At the same time, there are some practical limitations, which have to do with laser excitation and detection, but also we want to avoid other interfering processes. At the end of the section we will give some properties of the chosen states.

Our original idea to study dipole-dipole interactions in a position resolved manner was to make use of two states with a maximum permanent dipole, which would exchange state. Like the following

$$60(k = -59) + 61(k = -60) \leftrightarrow 61(k = -60) + 60(k = -59),$$  \hspace{1cm} \text{(2.42)}

as is described in [81]. We could call the $n = 60$ state the $|0\rangle$ qubit and the $n = 61$ state the $|0\rangle$-qubit. In rubidium there is no such thing as a $k = -60$ or $k = -59$ state, but we can use the lowest manifold state. We can produce this state by exciting with the laser to the d-state and subsequently ramping the electric field to just beneath the Inglis-Teller limit (see figure 2.2 and equation 2.35). To measure a process like this a position resolved detection system is needed. We have developed such a detection method (chapter 4), but
Theoretical background

Figure 2.5: All energy level crossings for the $41d + 49s \rightarrow 49p + 42p$ transition. In red we depicted the difference in binding energy for one atom $E_{41d} - E_{42p}$, in blue the difference for the other atom $E_{49p} - E_{49s}$. Indicated are the exact $j$ values of the transitions. The circles indicate the resonances.

it is unfortunately not as easy to operate as we hoped. Secondly, we could not confirm that we were really producing the lowest manifold state.

We have chosen a different approach, which involves four different states, such that it does not need the position resolved detection technique.

$$n_1 \ell_1 + n_2 \ell_2 \leftrightarrow n_3 \ell_3 + n_4 \ell_4.$$  \hfill (2.43)

Now we can probe the interaction by just using state-selective field ionization (SFI), which can distinguish the four states. A process like $n_1d + n_2d \rightarrow n_3p + n_4m$, comparable to the measurement described in subsection 2.3.4, is not practical, because there are so many resonances. Furthermore, the processes from equation 2.37 between two $nd$ atoms of the same kind occur, which severely disturbs the signal. For this reason, we search for interactions between $s$-, $p$- and $d$-states only.

One possible candidate is the transition $25s + 33s \leftrightarrow 24p + 34p$, as investigated in [7], with an interaction strength of $V_{dd} \sim 1.5$ kHz at 20 $\mu$m. However, we prefer a stronger interaction, enabling us to work at larger distances, which will be achieved if $n$ is increased ($V_{dd} \sim n^4/R^3$). Up to $n=50$ we can clearly distinguish subsequent $n$-states using SFI. We considered transitions between $n=40$ and 50 that had the smallest energy difference at zero field (the energy difference between the left- and right hand side of equation 2.43). Secondly we have to check if there existed an actual resonance for the transition and if the transition dipole moments were large enough.

In rubidium, the process $41d + 49s \leftrightarrow 42p + 49p$ seemed most suitable. Note that this type of transition can still be written in the form of two qubits, just as for the transition of equation 2.42, despite the fact that there are now four states involved. Atom A starts in the $41d$ state, or the $|1\rangle$ state and changes to the lower lying $42p$ state, the $|0\rangle$ state, while atom B starts in $49s$, $|0\rangle$, and is excited to $49p$, $|1\rangle$. The fact that $|0\rangle$ and $|1\rangle$ have a different meaning for each atom is not important, as long as the energy difference is equal. In figure 2.5 we have depicted the difference energies for the transitions of both atoms.
2.4 Dipole-dipole interactions between Rydberg atoms

Figure 2.6: The two-atom energy levels of the $|41\ell d, 49s\rangle$ and $|42\ell p, 49p\rangle$ system. The red line depicts the $|42p_{\frac{1}{2}}, 49p_{\frac{1}{2}}\rangle$ state, where we use the quantum numbers $n\ell j|m_j\rangle$. Green is $|42p_{\frac{1}{2}}, 49p_{\frac{1}{2}}\rangle$. The blue solid line represents the $|41d_{\frac{3}{2}}, 49s_{\frac{1}{2}}\rangle$ state and the blue dashed line is the $|41d_{\frac{1}{2}}, 49s_{\frac{1}{2}}\rangle$ state. The last one is not excited by the laser in our experiment and therefore does not play a role. The inset shows the avoided crossings due to the dipole-dipole coupling between the atoms, zoomed in around the two relevant resonances $F_1$ and $F_2$.

Around 0.4 V/cm there is a group of four resonances. The specific transition including the quantum number $j$ for these resonances is

$$41d_{3/2} + 49s_{1/2} \leftrightarrow 42p_{1/2} + 49p_{3/2}. \tag{2.44}$$

In figure 2.6 we zoomed in around these resonances and in this case we depicted the sum of the binding energies of the two atoms, either in the $|41d_{\frac{3}{2}}, 49s_{\frac{1}{2}}\rangle$ state or the $|42p_{\frac{1}{2}}, 49p_{\frac{1}{2}}\rangle$ state. Both states split up in two, for the different $|m_j\rangle$ values. However, the initial state that is excited by the laser is only the $|41d_{\frac{3}{2}}, 49s_{\frac{1}{2}}\rangle$ state (the quantum numbers here are $n\ell j|m_j\rangle$). This is because the laser is linearly polarized and we have $\Delta m_j = 0$ starting for both atoms from the $|5s_{\frac{1}{2}}\rangle$ state. Therefore we have in the experiment only two resonances that play a role; they occur at fields of $F_1 = 0.3807$ V/cm and $F_2 = 0.4112$ V/cm.

All involved states have a purely quadratic Stark shift. We fitted the energy levels from the Stark map with a parabola and obtained all polarizabilities $\alpha$ (see Eq. 2.27). The fact that energy levels shift in a field means that they are not pure $\ell$ states anymore, but they are mixed states. The amplitudes of the states that are mixed-in can be read off in the eigenstates when diagonalizing the Hamiltonian for the Stark map. At 0.4 V/cm the mixing is still very small, all involved states still have an population of 0.998 or higher in the original $\ell$ state. This mixing in of other states also leads to a permanent dipole moment, which is equal to the slope of the line in the Stark map. We have summarized...
Table 2.4: The polarizability $\alpha$ of each involved state and the permanent dipole moment $\mu_{\text{perm}}$ at 0.4 V/cm (both in MHz/(V/cm) as well as atomic units) for all relevant states of the process given in Eq. 2.44. These values $\mu_{\text{perm}}$ are in fact the local derivative of the binding energy, or the slope in figure 2.6, i.e. $\mu_{\text{perm}} = 2\alpha F$. The indices in the first column refer to $j$ and $|m_j|$. Both the polarizability as well as the permanent dipole moment at 0.4 V/cm in table 2.4. The resulting permanent dipole moments are not very large, at least, compared to the transition dipole moments, which we will give below. We assume we can ignore effects due to the permanent dipole moment.

<table>
<thead>
<tr>
<th>state</th>
<th>$\alpha$ (MHz/(V/cm)$^2$)</th>
<th>$\mu_{\text{perm}}$ (MHz/(V/cm))</th>
<th>$\mu_{\text{perm}}$ (a.u.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>41d$^{3/2,1/2}$</td>
<td>1.1</td>
<td>0.43</td>
<td>0.34</td>
</tr>
<tr>
<td>49s$^{1/2,1/2}$</td>
<td>44</td>
<td>17</td>
<td>14</td>
</tr>
<tr>
<td>42p$^{1/2,1/2}$</td>
<td>83</td>
<td>33</td>
<td>26</td>
</tr>
<tr>
<td>49p$^{3/2,1/2}$</td>
<td>309</td>
<td>124</td>
<td>97</td>
</tr>
<tr>
<td>49p$^{3/2,3/2}$</td>
<td>260</td>
<td>104</td>
<td>81</td>
</tr>
</tbody>
</table>

Table 2.5: The size and the polarization of the various transition dipole moments $\mu_t$ in the process 2.44 and the resonance where the transition occurs. Note that here the sign of $m_j$ does matter, so the indices refer now to $j$ and $m_j$. We have given only initial $m_j$ values of 1/2; the initial $m_j$ values of -1/2 give equivalent values for the transition dipole moments. The errors are an estimate of the accuracy, based on the difference between the analytical and the numerical calculation.

<table>
<thead>
<tr>
<th>transition</th>
<th>$\mu_t$ (a.u.)</th>
<th>polarization</th>
<th>resonance</th>
</tr>
</thead>
<tbody>
<tr>
<td>41d$^{3/2,1/2}$ ↔ 42p$^{1/2,1/2}$</td>
<td>1000(3)</td>
<td>linear</td>
<td>$F_1$ and $F_2$</td>
</tr>
<tr>
<td>41d$^{3/2,1/2}$ ↔ 42p$^{1/2,-1/2}$</td>
<td>707(2)</td>
<td>circular</td>
<td>$F_1$ and $F_2$</td>
</tr>
<tr>
<td>49s$^{1/2,1/2}$ ↔ 49p$^{3/2,1/2}$</td>
<td>1135(3)</td>
<td>linear</td>
<td>$F_1$</td>
</tr>
<tr>
<td>49s$^{1/2,1/2}$ ↔ 49p$^{3/2,-1/2}$</td>
<td>803(2)</td>
<td>circular</td>
<td>$F_2$</td>
</tr>
<tr>
<td>49s$^{1/2,1/2}$ ↔ 49p$^{3/2,3/2}$</td>
<td>1390(4)</td>
<td>circular</td>
<td></td>
</tr>
</tbody>
</table>

The transition dipole moments are calculated with the numerical and the analytical method described in subsection 2.2.4. The results are given in table 2.5. Both dipoles are of the order of 1000 a.u., equal to ~2500 D (debye). These numbers are enormous when compared to ground-state molecules or atoms; also, they are an order of magnitude larger than for the 25s$^3$+33s interaction, where we have dipole moments of only ~60 and ~200 a.u. Secondly, note that the error of the calculation, i.e. half the difference between the numerical and the analytical method, is only 0.3%. If we calculate the dipole-dipole interaction strength (Eq. 2.41) for both transitions linearly polarized and both dipoles parallel to the distance vector $R$ we get $V_{dd} = 2\pi 35$ kHz for a distance of 40 $\mu$m. This means that the energy is transferred from one atom to the other and back in 14.5 $\mu$s, in other words the Rabi oscillation period of each atom is 14.5 $\mu$s. The radiation that drives the Rabi oscillation is a photon which is emitted by the 41d atom and absorbed by the 49s atom. The photon energy can be read off from figure 2.5: 1.1 cm$^{-1}$, corresponding to a wavelength of $\lambda = 0.91$ cm and a frequency of 32.8 GHz. This wavelength is much larger than the typical separation between the atoms, tens of microns, so it is a near-field interaction.
The energy transfer period of 14.5 $\mu$s is shorter than the natural lifetime of the states as well as the black-body transition lifetime (see subsection 3.4.3). This leads to the conclusion that the interaction is strong enough for this particular transition, even at the enormous distance of 40 $\mu$m; which is a distance we should be able to control and measure.

2.5 Simulations

In order to understand what would happen in the experimental setup and to tune the experimental parameters for observable results, we have performed several simulations. We will start with a simple 2-body calculation in subsection 2.5.1. We will compare two situations, namely one, where both particles are fixed in space, and the other, where again both particles are fixed in space, but one on a random location within a cigar-shaped ellipsoid at some distance $d$ from the other particle. The latter, averaged over lots of realizations, should represent the real experiment we are doing. In some limiting cases, the calculation can be done analytically.

In subsection 2.5.2 we describe a many-body simulation with one atom in state A among many atoms in state B, where the atoms are either arranged in a random way, like a ”frozen” gas or in an ordered way, like a crystal. The calculations predict a localization effect for the randomly organized system. Unfortunately we have no experimental verification of the effect.

2.5.1 Dipoles distributed over long cylinders

In general a coupled two-level system can be described by the Hamiltonian

$$H = \begin{pmatrix} -\delta/2 & V \\ V & \delta/2 \end{pmatrix},$$

(2.45)

where $V$ is the interaction between the unperturbed states and $\delta$ is the unperturbed energy difference between the two states. The detuning $\delta$, for example, vary as a function of field. If we diagonalize $H$ we obtain for the eigenenergies an avoided crossing at $\delta=0$ with a width $2V$. The amplitude of both states evolve in time as follows

$$\begin{pmatrix} c_1(t) \\ c_2(t) \end{pmatrix} = \left( \begin{pmatrix} c_1(0) \\ c_2(0) \end{pmatrix} e^{-iE_t/\hbar} \right) \Psi,$$

(2.46)

where $\Psi$ is the matrix containing the eigenvectors of $H$ and $E$ is the vector containing the eigenvalues of $H$. Starting with 100% probability in the first state the probability of the second state becomes

$$P_2(t) = |c_2(t)|^2 = \frac{4V^2}{4V^2 + \delta^2} \sin^2 \left( \frac{1}{2} \sqrt{4V^2 + \delta^2} t \right)$$

(2.47)

and $P_1(t) = 1 - P_2(t)$. This oscillation of the probability is called a Rabi oscillation.
In our experiments the two states are the two two-atom states \(|41d, 49s\rangle\) and \(|42p, 49p\rangle\) or \(|ds\rangle\) and \(|pp\rangle\) for short. \(V\) is the dipole-dipole interaction. For two single atoms the dipole-dipole interaction is (see also Eq. 2.41)

\[
V = \gamma \frac{\mu_A \mu_B}{R^3},
\]

(2.48)

where \(\mu_A\) and \(\mu_B\) are the transition dipole strengths \(|\langle 42p| \mathbf{r} |41d\rangle|\) and \(|\langle 49p| \mathbf{r} |49s\rangle|\) and \(\gamma\) is a factor between -2 and 1, determined by the angles between the dipoles and the distance vector \(\mathbf{R}\) between the atoms. At resonance (\(\delta = 0\)) the \(|pp\rangle\) probability becomes

\[
P_{pp'} = \sin\left(\gamma \frac{\mu_A \mu_B}{R^3} t\right)^2 \approx \gamma^2 \frac{\mu_A \mu_B^2}{R^6} t^2,
\]

(2.49)

where the approximation is valid for small \(Vt\). So, for small \(Vt\), \(P_{pp'}\) scales with \(t^2/R^6\).

In the experiment multiple atoms are distributed over long cylinders. The way we calculate the \(|pp\rangle\) probability of the system is by having one atom in a cylinder, with its center on \((x, y, z) = (0, d, 0)\) and one atom on position \((0, 0, 0)\). Interactions within one cylinder could play a role, but here we do not take them into account. We use for the dipole-dipole interaction

\[
V = \gamma(x, y, z) \frac{\mu_A \mu_B}{(x^2 + (y + d)^2 + z^2)^{3/2}}.
\]

(2.50)

where \(\gamma(x, y, z)\) contains the angular dependence. The cylinder is oriented along the \(z\)-axis with line density \(\lambda\) and radius \(\rho\). We can integrate the short-time probability \(P_{pp'} = V^2 t^2\) to get the total probability \(P_{\text{cyl}}\) for the system. In reality the cylinders are about \(500 \mu m\) long and have a radius of about \(8 \mu m\). The distance \(d\) between the cylinders is typically varied from \(0\) to \(80 \mu m\). Therefore, using infinitely long cylinders is well justified, but the radius of the cylinders should be taken into account. In cylinder coordinates we get

\[
P_{\text{cyl}} = \frac{\lambda \mu_A^2 \mu_B^2}{\pi \rho^2} \int_{-\infty}^{\infty} \int_{-\pi}^{\pi} \frac{\gamma(x, y, z)^2}{(r^2 \sin^2 \theta + (d + r \cos \theta)^2 + z^2)^{3/2}} t^2 r \, dr \, d\theta \, dz
\]

\[
= \frac{\lambda \mu_A^2 \mu_B^2}{d^3} \frac{\gamma'(\eta)^2}{12} \frac{(\eta - 1)^2 K\left(\frac{4\eta}{(\eta+1)^3}\right) - (7\eta^2 + 1) E\left(\frac{4\eta}{(\eta+1)^3}\right)}{(\eta - 1)^3 (\eta + \eta^2)^2} t^2,
\]

(2.51)

where we have replaced \(\rho/d\) with \(\eta\) and \(\gamma'(\eta)\) is now the angular dependence factor. \(K(m)\) and \(E(m)\) are the complete elliptic integrals of the first and second kind. We do not know what \(\gamma'(\eta)\) is, but for \(\eta \to 0\) we can do the integration that does include the angular dependence. For dipoles oriented in the \(y\)-direction we have

\[
P_{\text{lin}} = \frac{\lambda \mu_A^2 \mu_B^2}{128} \frac{1 - 3d^2/(d^2 + z^2)^2}{(d^2 + z^2)^3} t^2 \, dz
\]

\[
= \frac{123\pi}{128} \frac{\lambda \mu_A^2 \mu_B^2}{d^5} \frac{1}{r^2} t^2,
\]

(2.52)

which means that \(|\gamma'(0)| \approx 1.6\) for dipoles in the \(y\)-direction. So for interaction between atoms on lines, the probability scales like \(r^2/d^5\) for small \(Vt\). So this probability drops
2.5 Simulations

not as strongly with the distance as for the case of two point particles, where we have $P \sim t^2/R^6$.

For a calculation of the $|pp\rangle$ probability for large $Vt$, the behavior becomes oscillatory and analytical integrations are no longer possible; we need to perform numerical simulations of the system. For this we used a three-state system, equivalent with the 49s+41d→49p+42p system, with two avoided crossings 4 MHz apart (see Fig. 2.6). In figures 2.7, 2.8 and 2.9 we compared the cylinder configuration to a simple two particle system.

![Figure 2.7](image)

**Figure 2.7:** a) The excited state probability as a function of detuning for one point particle and one particle distributed over a cylinder at 20 $\mu$m distance. The red line is a sum of two Lorentzians with width $2V$, but for the calculation of $V$ we used a distance of 25 $\mu$m. b) The same for two particles at 20 $\mu$m distance. The red Lorentzian here is plotted for 20 $\mu$m.

We randomly placed one particle in an elongated ellipsoid, Gaussian in 3D with a half length of 200 $\mu$m and a radius of 8 $\mu$m (both numbers are $1/\sqrt{e}$ half widths or $\sigma$) at some distance of a probe particle. For the interaction strength we used equation 2.50 with $\gamma(x,y,z)\mu_A\mu_B = 4000$ MHz$\cdot$µm$^3$. We evaluated the $|pp\rangle$ probability by diagonalizing the Hamiltonian for 10000 random cases.

For the first simulation we had the probe particle at 20 $\mu$m distance from the elongated ellipsoid and we determined the average $|pp\rangle$ probability after 10 $\mu$s for various detunings (e.g. various static field values). We multiplied the total probability by 3 because this gives a more realistic picture; this is justified because in the real experiment there are a few tens of atoms in each volume instead of one (however the probability does not scale linearly with the density). The result is shown in figure 2.7a. We also performed the same calculation for two particles at a fixed distance of 20 $\mu$m, shown in figure 2.7b. Here we observe an oscillating function underneath a Lorentzian envelope with a half width of $2V = 2 \times 4000/20^3$ MHz, as is expected from equation 2.47. In figure 2.7a the oscillations are almost averaged out, because of all the different occurring distances and therefore different interaction strengths. Surprisingly, the oscillations have not disappeared completely. Another difference is that the Lorentzian envelope is narrower, we plotted here one with a width of $2 \times 4000/25^3$ MHz. Apparently, due to the low filling of the cylinder, the effective distance is 25 $\mu$m. This also gives rise to the lower probability of $\sim$0.3 at resonance.
Theoretical background

Figure 2.8: a) The excited state probability as a function of distance for one point particle and one particle distributed over a thin, long cylinder. In the inset of a) a log-log-plot of the data together with a a plot of equations 2.51 in green and 2.52 in red, showing that the simulation data indeed match the $d^{-5}$ scaling for large $d$. b) The excited state probability as a function of distance for two particles.

The Lorentzian line shape can be deduced from equation 2.47. For varying $V$, the oscillations average out and a Lorentzian remains. For large $Vt$ – we call the transition saturated – $\sin^2$ averages to $\frac{1}{2}$ and we have

$$P_2 \approx \frac{1}{2} \frac{4V^2}{4V^2 + \delta^2}. \quad (2.53)$$

This probability as a function of detuning resembles a peak with a height of $\frac{1}{2}$ and a half width of $2V$. In the case of small $Vt$ the equation becomes $P_2 \approx V^2 t^2 \text{sinc}^2 \frac{\delta}{2} t$; this is roughly similar to the Lorentzian shape

$$P_2 \approx \frac{4}{4 + \delta^2 t^2} V^2 t^2. \quad (2.54)$$

This peak has a half width of $2/t$ and a height of $V^2 t^2$.

For figure 2.8a and 2.8b we kept the detuning at one of the resonances and varied the distance between the probe particle and the cylinder resp. the other particle. The $|pp\rangle$ probability is plotted after 10 $\mu$s. In figure 2.8a we multiplied the probability by 3. Again we see oscillations in figure 2.8b. In the center it is actually oscillating very rapidly, which is why we have left the center part out. In figure 2.8a the oscillations seem to have damped out completely. In the inset in figure 2.8a we made a comparison to the simplifications 2.52 and 2.51, which both fit quite well for larger distances.

In figure 2.9 we depicted the evolution of the $|pp\rangle$ probability at resonance for several distances. Again for a particle and a cylinder (a), where we multiplied the probability with 3 and for just two particles (b). Also here the oscillations are not visible for the case of the cylinder. For large distances or short times the quadratic behavior of the probability is visible. If we wouldn’t have multiplied with three, the simulated probabilities all go to 0.5 for infinite times as expected. In the inset of figure 2.9a we depicted a double logarithmic plot of the transfer rate $1/\tau$, defined as $P(\tau) = 0.05$, again together with equations 2.52 and 2.51, showing that the $d^{-3/2}$ scaling of this parameter indeed fits.
Despite the fact that we used a strongly simplified calculation we could still predict the behavior of our system quite well. Qualitatively, but also quantitatively the calculations are not far off from the experimental data presented in chapter 5. Moreover, these simple calculations, especially the ones in analytical form gives us an intuitive understanding of the system. It appears that, for particles distributed over two cylinders oscillations will not appear in the experimental data, because each pair will have a different interaction strength, and hence a different oscillation frequency. Therefore it is not directly possible to measure if the interactions are coherent or not. In chapter 5 results of more extensive simulations are depicted, performed by J. V. Hernández and F. Robicheaux. Here the angular dependence, as well as many body interactions are taken into account.

2.5.2 Frustrated diffusion

In this section we study the difference between a random organization of dipoles compared to a lattice-like organization in the propagation of an excitation by dipole-dipole interaction. We placed one atom in the excited state (e.g. the 41d state) in the center of a 100 µm cube and 124 lower state atoms (e.g. in the 49s state) around it. In one case the 124 lower state atoms are distributed randomly, while in the other case they are distributed in a lattice with unit cell of 20 µm. The excited state can diffuse now over the cube through dipole-dipole interaction.

The system consists of 125 different basis states, namely the excited atom being one of the 125 atoms. All states have the same unperturbed energy and two of such states are coupled through the dipole-dipole interaction between the two atoms that exchange the excitation. We diagonalize the Hamiltonian and calculate the evolution of the system in time, as like equation 2.46 (in this case we have amplitudes $c_1$, $c_2$, ..., $c_{125}$). In figure 2.10 we depicted the excited state probability versus position $z$ (the direction of the dipoles), averaged over times between $t = 40–110 \mu s$, to eliminate the oscillations. For the case of
Figure 2.10: The diffusion of an excited state in a 3D-volume with atoms in a lower state. We compare two configurations, which are illustrated on the right hand side, a random organization of atoms in blue and an ordered or lattice-like organization in red. The green filled circle represents the atom that is in the excited state at $t = 0$. In the main figure, the blue line depicts the excited state probability in the $z$-direction for the randomly organized situation, the red line is for the case of the ordered situation. Note that the vertical scale is logarithmic.

In the random gas the probability is averaged over 1000 realizations.

Figure 2.11: The excited state probability as a function of time for the particles arranged on a 20×40 µm rectangle as depicted in the inset. In figure a) one particle is missing and the excited state probability is only weakly transferred to the green particle on the right hand side. In figure b) the excitation is transferred to the right hand side, due to the more regular structure of the arrangement of the particles.

In the ordered lattice case the excited state diffuses virtually homogeneously over the whole cube, with a bit more probability in the center. In the randomly distributed case however, the excited state is strongly localized in the center. This is due to the fact that the energy transfer is dipole-blockaded: atoms at small distances are shifted in energy and can not resonantly transfer energy to further lying atoms.

This effect is more clearly illustrated in figures 2.11a and 2.11b. Here a similar simulation is performed with only a few atoms in a 20×40 µm rectangle. The excited state starts on the atom in the lower left corner (depicted in red). In the three-atom case the
energy is hardly transferred to the atom on the right hand side. This is because the two atoms on the left hand side have shifted energy levels due to the strong dipole-dipole interaction. This makes the interaction with the right-hand side particle no longer resonant and blockade occurs. In the four atom case, figure b, the excited state does reach the right hand side of the rectangle, because on the right hand side the energy levels are just as much shifted as on the left side, making the transfer resonant again. In a randomly distributed gas asymmetric situations like the three atom configuration in figure 2.11a occur very often, resulting in a localization of the excitation in the area where it started.

This blockade effect can actually be used when just one atom per volume is desired. If the volumes are small enough and the laser power is tuned such that it excites 0, 1 or 2 atoms per volume, e.g. like a Poisson distribution, then only in the 1 atom per volume case the dipole-dipole interaction signal between the volumes is significant. When there is more than one atom per volume, the interaction within a volume induces a level shift, making the interaction between volumes non-resonant. The term ”dipole blockade” is mainly used for the effect that with a narrow bandwidth laser the number of excited Rydberg atoms in a certain volume is limited. In this case the ground state atoms that surround one Rydberg atom can no longer be excited, because this Rydberg atom shifts the Rydberg levels of the other atoms due to the dipole-dipole interaction. This makes the laser excitation no longer resonant.

The fact that no diffusion occurs in a randomly ordered system is rather interesting and unexpected and is a similar effect as Anderson localization [5]. In [70] the claim is made that diffusion does occur, but it is not mentioned how far it extends. Secondly, the transition the authors use is essentially different – a three-state system like \( ns + ns \rightarrow np + (n-1)p \). In our experiments we have not been able to proof this localization effect.

2.6 Summary

We have presented two methods to calculate the Rydberg wavefunctions, a numerical and an analytical method. There are only small differences between the results of both methods, assuring us that both type of calculations are accurate enough for our purposes. The wavefunctions form the basis for the calculation of e.g. the Stark shift or the (transition) dipole moments. The structure of the Stark map is demonstrated in a measurement of dipole-dipole interaction resonances. We used the Stark map calculation and the calculation of the transition dipole moments to find a suitable resonant dipole-dipole transition for the experiments in this thesis. We have chosen the transition \( 41d + 49s \leftrightarrow 42p + 49p \) which is resonant at a field of only \( \sim 0.4 \) V/cm. The relevant transition dipole moments are of the order of \( \sim 1000 \) a.u., resulting in a Rabi oscillation period of 14.5 \( \mu s \) for a distance of 40 \( \mu m \). Simulations of dipole interacting atoms distributed over elongated volumes provide us insight in their behavior in a realistic experimental setup. It appears that for dipoles distributed over two thin parallel lines separated by the distance \( d \), the short-time transition probability scales like \( P \propto \frac{\vec{r}^2}{d^5} \). A simulation of excitation diffusion through dipole-dipole interaction shows a localization effect for randomly organized systems, due to the dipole blockade effect.
3 Experimental Setup

The experimental setup used in the experiments described in this thesis consists of a rubidium magneto-optical trap (MOT), two pulsed Nd:YAG pumped dye lasers, a vacuum system in which the experiments take place and a detection system. The MOT provides cold ground-state atoms (83(7) $\mu$K); the cooling is necessary to keep the atoms in place during the experiments. These atoms are excited to Rydberg states by two focused dye laser beams, with spectral linewidths of 5.4(4) and 0.51(3) GHz, creating two separate elongated volumes of Rydberg atoms, with diameters of 11.6(4) $\mu$m and 16.3(5) $\mu$m. After a controlled interaction time, the Rydberg atoms are detected by means of state-selective field ionization that can distinguish the principal quantum number of the state. The dipole-dipole interaction experiments demand an accuracy and stability for the electric fields ($<2$ mV/cm) and magnetic fields ($<0.15$ G), which requirements are just met. This experimental setup thereby provides fair control over all important parameters of the dipole-dipole interaction.
3.1 Introduction

The aim of the experimental setup is to have control over all parameters of the dipole-
dipole interaction: the position of the atoms, the interaction time, the size of the dipole
moments and the resonance condition. In this chapter we will describe how we have
achieved control over these parameters. An illustration of the experiment and the impor-
tant parameters is depicted in figure 3.1. The vacuum chamber with all its components is
shown in figure 3.2.

Figure 3.1: In the experiment, two different types of Rydberg atoms, drawn here as red and orange
filled circles, are created in separate volumes from a cloud of cold ground state atoms (blue circles).
The dipole-dipole interaction between the different Rydberg atoms typically takes place on a timescale
of \( \tau \approx 5 \mu s \) for a distance of \( d = 25 \mu m \). This requires that the average speed \( \bar{v} \) of the atoms is much
slower than several meters per second, and thus a temperature below 100 mK. Secondly, the width \( w \)
of the volumes must be smaller than the interaction distance \( d \). An electric field \( F \) brings the dipole-
dipole interaction into resonance, which must be accurate up to the level of millivolts per centimeter for
a typical resonance, because this corresponds to \( 1/\tau = 200 \) kHz; the magnetic field \( B \) is for the same
reason required to be below 0.15 G. The density \( \rho \) of the atom cloud must be at least such that the
average inter-particle distance \( a \) is much smaller than the interaction distance \( d \), to ensure a sufficient
number of interacting atoms.

The position of the atoms is controlled in two dimensions by the creation of a small fo-
cus in the laser beam that excites the atoms to Rydberg states. This ensures that the atoms
are created in a cylinder-like volume of a few micrometers diameter. This is small enough,
since Rydberg atoms can interact over tens of micrometers distance. The typical interac-
tion time in our experiment is several microseconds. During this time, room temperature
atoms would have flown off already. Therefore, we also need to control the speed of the
atoms. This is done by using laser-cooled atoms, here realized with a magneto-optical
trap (MOT). These atoms, the majority in the 5s ground state, form a \( \sim 1 \) mm cloud with a
temperature of \( \sim 100 \mu K \). The working principle of the MOT, as well as the experimental
Figure 3.2: a) A cut-open view of the stainless steel vacuum chamber. The main reaction center is inside the right-hand 6-way cross, where all the orange and red laser beams come together and where the circular field plates are visible. On the outside we have two bigger MOT coils and four smaller MOT-moving coils. Inside, toward the left, the flight tube, which can be set at a voltage to slow down the electrons from the ionized Rydberg atoms on their way to the MCP. A turbo pump underneath the larger 6-way cross takes care of the ultra-high vacuum ($\sim 3\times10^{-8}$ mbar). Large compensation coils are placed around the setup to realize zero magnetic field. b) The setup zoomed in at the reaction center. The two plates $P_1$ and $P_2$ have a hole to allow one pair of MOT beams to go through, as well as ionization products. The plates are separately supplied by a voltage, together with the pillars that hold up the plates. c) The inside of the dispenser block. Charged particles are bent off by the two plates at opposite voltage. The cover is grounded and the neutral atoms go through the slit on top, which is aligned with the MOT center.
implementation and a measurement of the number of atoms is described in section 3.2.

The excitation to Rydberg states – states with high principal quantum number \( n \) – is achieved with pulsed dye lasers. These lasers operate at \( \sim 594 \) nm, which gets the ground-state atoms up to Rydberg states with two photons, schematically depicted in figure 3.3. The pulse length of only 8 ns provides an sharp starting time for the interaction. The wavelength can be tuned to excite only one \( n\ell \) state (\( \ell \) is the angular momentum quantum number). Two separate lasers are focused in the MOT cloud to make two different states. The lasers operate at a repetition rate of 10 Hz, which is consequently the repetition rate of the experimental data acquisition. More about the Rydberg excitation is described in section 3.3.

We have control over the size of the dipole moments, and consequently the interaction strength, by choosing specific Rydberg states. In the case of an interaction like \( n_1k_1 + n_2k_2 \leftrightarrow n_2k_2 + n_1k_1 \), with \( k \) the parabolic quantum number, the interaction is always resonant – the atoms have just exchanged their states – and every \( n \) can be chosen. The dipole moment of an atom in such a state \( nk \) is \( \frac{1}{2}nk \), where \( k \) can have values between

**Figure 3.3:** The energy level scheme of rubidium for s, p and d states. The laser transitions at 780 nm and 795 nm are used for the MOT. The 594 nm laser couples the 5s state with either ns or nd-states through a two-photon transition via a virtual p-state.

**Figure 3.4:** A more detailed picture of the MOT transitions in Rb-85. The nuclear spin \( I=5/2 \) gives rise to a hyperfine splitting depicted by \( F \), with the hyperfine shift \( \Delta \nu \) in MHz.
3.1 Introduction

$n + 1$ and $n - 1$. These dipole moments are independent of the electric field (at least for a large range of electric fields), i.e. they are permanent dipoles. Another possibility is to study interactions like $n_1\ell_1 + n_2\ell_2 \leftrightarrow n_3\ell_3 + n_4\ell_4$, with $\ell$ the angular momentum quantum number. These states have a dipole moment that increases linearly with electric field, which means their energy changes quadratically with the field. These dipole moments are not very large, but the different field dependence of these states can provide for resonances in these types of interactions. In these interactions the transition dipole moments – between $n_1\ell_1$ and $n_3\ell_3$ and between $n_2\ell_2$ and $n_4\ell_4$ – are very strong. These dipole moments are of the order $n^2$, like the permanent dipoles in the previously mentioned case. Since the resonances are usually rather narrow, the electric field has to be very accurate. Obtaining this accuracy is described in section 3.4. The motivation to select a specific combination of Rydberg states is described in chapter 2.

![Figure 3.5: A picture of the Coulomb potential $-1/r$, as experienced by the electron in a Rydberg atom, included are iso-potential lines and the classical trajectory of an electron. Figure a) is without external field, b) is with constant external electric field, where the electron can escape the atom.](image)

A few $\mu$s after the lasers have excited the atoms to the states $n_1$ and $n_2$ – the initial states in the dipole-dipole interaction process – we want to determine the fraction in the final states $n_3$ and $n_4$. This is achieved by ionizing the Rydberg atoms with an electric field [35]. Since the outer electron is very loosely bound, only a modest electric field is needed (e.g. $\sim 100$ V/cm for $n = 40$). A sketch of a two-dimensional potential for the electron in an atom is shown in figure 3.5a). This potential combined with an external homogeneous electric field is shown in figure 3.5b). Depending on the state of the atom (or energy of the electron), the electron escapes the atom at a different field. The electron flies against the direction of the field, toward a micro-channel plate (MCP), which detects the single electrons. When a field is applied that increases with time, one can deduce the original state of the atom from the arrival time of the electron. More details on the detection method are described in section 3.5. With an extended field ionization scheme together with time-of-flight data, the original position of the atoms can also be deduced. This is described in chapter 4.
3.2 The Magneto-Optical Trap

If an atom absorbs a photon, it does not only absorb its energy, but also its momentum. In most cases this is negligible, since the momentum of a room-temperature rubidium atom (~300 m/s) is a factor of ~50,000 more than the momentum of a visible photon. However, with methods developed in the 1980’s under the name ”laser cooling”, rewarded with the Nobel prize in 1997, the photon momentum can be used to slow down atoms to about 0.3 m/s and trap them [77, 67]. With this low velocity we can regard the atoms as ”frozen”, in other words we have control over the position of the atoms in the sense that they don’t move on the relevant time and distance scales (see Fig. 3.1).

![Diagram of the MOT setup](image)

Figure 3.6: A schematic representation of the MOT setup, including the direction of the current I through the coils, the resulting direction of the magnetic field B and the polarizations of the laser beams.

In the experimental setup, the atoms are obtained from a SAES Getters rubidium dispenser (Fig. 3.2). This is a ~1 cm long, ~1 mm thick tube, containing a mixture of rubidium chromate (Rb₂CrO₄) and a reducing agent (Zr 84% - Al 16%) [83]. They are mainly manufactured for the photo-tube industry, but are widely used in the laser-cooling community. When running a current of 4.5 - 7.5 A through the dispenser, it heats up to a few hundred degrees Celsius and atomic rubidium comes out of the slit at the front. However, we observed that charged particles come out as well, which is a problem for the field-sensitive Rydberg atoms. To solve this problem we have placed two plates in front of the dispenser, at voltages of +6 V and -6 V followed by a grounded plate with a ~1 mm slit. The charged particles are bent off in the field, while the neutral atoms go straight through the slit of the plate in front. The whole dispenser assembly can be seen in figure 3.2c.

The light used to cool the atoms stems from a 780 nm laser diode. The laser is 12 MHz red detuned from the $5^2S_{1/2} (F=3) \rightarrow 5^2P_{3/2} (F=4)$ resonance in $^{85}$Rb. This transition is de-
3.2 The Magneto-Optical Trap

Picted in figure 3.4. The red detuning ensures that an atom moving toward the laser beam is resonant with the light, due to the Doppler effect. After multiple processes of stimulated absorption and spontaneous emission of a photon, the net effect is a large absorbed momentum in the direction of the laser beam. This is because all the absorptions are in the same direction and add up, while all the emissions are in all directions, isotropically distributed, so they average out. There is a very small probability that an atom decays into the F=2 ground state, and then it can not longer be excited by the 780 nm laser. However, the small probability becomes a high probability after tens of thousands of cycles, so the atom will be lost for the cooling process. To solve this problem we add a 795 nm diode laser beam in the MOT center; the so-called repumper laser. This laser couples the 5^2S_{1/2} (F=2) state with the 5^2P_{1/2} (F=3) state, from here it has some probability to decay back into the 5^2S_{1/2} (F=3) state, from where it will continue to be cooled further. More details the diode laser system can be found in section 3.2.1.

To collect all the atoms in the same place (∼1 mm) a magnetic field is added, which is zero in the center and increasing in all directions (figure 3.6). Such a field is obtained with two coils in the anti-Helmholtz configuration, i.e. two coils with opposing currents. The coils consist of 125(5) windings with a radius of 5.5(0.3) cm at a distance ranging from 5.6 to 11.2 cm from the center. With a current of 9 A this should give a field gradient of 12 G/cm on the z-axis and 6 G/cm on the x and y axes. We will qualitatively describe how this field, combined with six laser beams of diameter ∼1 cm pointing toward the center, creates a trap for the Rubidium atoms.

The positive m states of the 5P_{3/2} (F=4) state shift down in energy in a negative B-field – due to the Zeeman effect – and σ^+ polarized light couples to these states, because of the selection rule Δm=+1. At the same field, negative m states are shifted upwards and σ^- light couples to them. So an atom at +z in figure 3.6 (negative field) will be more resonant to the σ^+ polarized laser beam, because of the red detuning of the laser. The beam at the right hand side in the picture will therefore push it toward the center.

The Zeeman shift depends on the magnetic field as \( \Delta E = m_F g_F \mu_B B \), with \( m_F \) the magnetic quantum number, \( g_F \) the Landé g-factor, \( \mu_B \) the Bohr magneton and B the magnetic field. Most of the cooling occurs between 5^2S_{1/2} (F=3,m_F=±3) and 5^2P_{3/2} (F=4,m_F=±4), which have a shift of respectively ±\( \mu_B B \) and ±2\( \mu_B B \). At the edge of a beam, 0.5 cm away from the center on the z-axis, the shift of the resonance frequency becomes ±8.4 MHz. This number is comparable to the laser detuning as well as to the Doppler shift for an atom velocity \( v=10 \text{ m/s} \); the Doppler shift is given by \( v/\lambda \), resulting in 12.8 MHz.

The MOT cloud is located at the point where the field is zero. This point can be moved in the z-direction, by changing the current in one of the MOT coils. To move the zero-point in the x-y plane two sets of smaller Helmholtz coils (10 windings) around the x and y-axis arms of the 6 way cross can be used (figure 3.2a). These coils just add a constant field in the x or y direction, which effectively moves the zero-point.

The number of atoms in the MOT cloud is typically 5.5(2.2)\( 10^{10} \text{ cm}^{-3} \), as we determined by fluorescence imaging described in section 3.2.2. The temperature of the MOT cloud is determined only in the final stage of writing this thesis [25]. Here, the temperature was obtained from real-time imaging of the expansion of the MOT cloud, after switching off the laser beams, with a result of 83(7) \( \mu \text{K} \). In all experiments described in this thesis the temperature was assumed to be below 300 \( \mu \text{K} \), which is typical for a ru-
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Beryllium MOT [100]. These temperatures of 300 μK resp. 83 μK correspond to an average speed of 0.27 μm/μs resp. 0.14 μm/μs. Both speeds are low enough to regard the particles as "frozen", when studying dipole-dipole interactions at distances of 20 - 50 μm in 10 - 20 μs.

3.2.1 Diode laser system for the MOT

![Diode laser setup diagram]

**Figure 3.7:** The diode laser setup. The light from the laser diode (LD) travels via the grating (gra) onto a mirror, which is attached to the grating, in order to keep the alignment of the exiting beam. The beam proceeds through a achromatic prism pair (APP), to change the beam profile from elliptic to circular. The following isolator prevents reflected laser light to enter the laser diode, which would give unwanted feedback. The combination of a λ/2 waveplate plus polarizing beam splitter cube (PBS) is used several times for splitting the beam and control the power in both ends. Here a weak beam is split off for the frequency locking setup, described in the main text. The Rubidium cell (Rb) transmission is monitored by a photodiode (PD). The complete setup in the blue rectangle is identical for both the 780 nm laser as the 795 nm laser. The 795 nm beam is then simply directed on the MOT. The 780 nm beam passes the 80 MHz acousto-optical modulator (AOM), where the +1st order is selected with a diaphragm (dia). The z-beam is split off first, and then both arms pass a telescope, which enlarges the beam size. One beam is split in two again for the x and y directions. After passing a λ/4 waveplate the beams enter the vacuum chamber. On the other side of the chamber the beams are reflected back onto the MOT cloud, passing another λ/4 twice.
A scheme of the cw-laser setup is depicted in figure 3.7. For both the cooling laser at 780 nm and the repumper laser at 795 nm we use grating stabilized diode lasers DL 100 from Toptica in Littrow configuration [106, 66, 80]. The light from the laser diode with a linewidth of $\sim 100$ MHz is collimated by a lens and then strikes a reflection grating, where the first order is focused back into the diode. This grating, together with the rear facet of the diode forms now the laser cavity, with a longer length than the original cavity – the diode itself. This results in a narrower linewidth of about 1 MHz. The angle of the grating determines the exact laser frequency and is automatically adjusted with a piezo actuator. Course adjustment over a few nm is performed with a screw on the grating.

![Figure 3.8](image)

**Figure 3.8:** In panel a) we depicted the signal of the photodiode in the laser-lock setup, while the laser frequency is scanned. Two Doppler-broadened absorption dips can be observed from the D2 line of the two most occurring rubidium isotopes. The small peaks in the absorption signal – the Lamb peaks – occur due to the counter propagating pump beam in the rubidium cell. In these much narrower peaks the hyperfine levels and crossovers are resolved. In panel b) the signal from the PDD is depicted, which is in fact the derivative of the photodiode signal. The dotted lines depict the hyperfine levels of the upper state as well as the crossovers (see also Fig. 3.4). The hyperfine resonances are denoted with the quantum number $F$ and the crossovers are denoted with letters. These crossovers are located exactly between the two involved resonances. Note that the lowest hyperfine state is not populated, since this transition is not allowed. We lock the laser on the Rb-85 2-4 crossover, depicted with the solid line denoted by "e".

A small amount of the laser light is deflected into a Doppler-free FM spectroscopy setup [13, 14]. Here part of the light, the probe beam, goes through a heated Rb vapor cell and then strikes a fast Thorlabs photodiode (1 ns rise time). The other fraction of the light, the pump beam, goes through the Rb cell in the opposite direction, overlapping with the probe beam. This creates narrow peaks on top of the Doppler-broadened absorption spectrum from the probe beam, known as "Lamb dips" (see Fig. 3.8a). These Doppler-free peaks are so narrow that the hyperfine splitting can be resolved. The peaks arise from
atoms with zero longitudinal velocity that absorb laser light from both beams at the same
time, therefore less absorption is visible from the probe beam. Apart from the resonances
itself, also crossover peaks are visible, located exactly between two resonances. This is
when atoms with some velocity are resonant with one transition of the pump beam and
with another transition of the probe beam.

A so-called Pound-Drever-Hall detector (PDD) superimposes a 20 MHz signal on the
current of the photodiode, creating a modulation of the laser frequency, which is converted
into amplitude modulation of the photodiode signal in the vicinity of a resonance. This
signal is in fact the derivative of the absorption spectrum, with a zero crossing at the
position of the resonance (Fig. 3.8b). With a home-built lock-in amplifier system, this
signal from the PDD is fed back into the piezo actuator, and sets the laser is back onto
the resonance.

The cooling laser is locked on the F=2, F=4 crossover peak, located at -93 MHz from
the F=4 resonance (see figures 3.4 and 3.8). The main beam goes through an Isomet
80 MHz Acousto-Optical Modulator (AOM). This is a crystal through which an acoustic
wave travels transversely to the light. The acoustic wave creates Bragg diffraction, where
several beams exit the crystal under a different angle, as well as at a different frequency.
These frequencies differ 80 MHz times the order number. We use the +1\textsuperscript{st} order, so that
the actual detuning of the laser becomes \(-93 + 80 = -13\) MHz. This is about twice the
natural linewidth. We lock the repumper laser on the F=3 resonance in the same manner,
and direct the beam onto the MOT center without any further manipulation. Both lasers
remain in lock for several hours, and therefore the MOT remains stable for the same
amount of time. A few hours of stability is required for running extensive automated
measurement series like the data presented in chapters 5,6 and 7, where each data point is
averaged over a few hundred 10 Hz dye-laser shots.

After passing through the AOM, the laser beam is split into three parts, one for each
orthogonal axis. To increase the maximum trapping velocity, the beams are expanded in
a telescope. The telescope consists of two positive lenses, increasing the beam size with
the ratio of the focal distances of the two lenses. These are a factor 8 resp. 6 obtaining
diameters of 1.3(1) and 1.0(1) cm (see Fig. 3.7). The z-beam is expanded less, because
this beam is limited by the 14 mm holes in the field plates P1 and P2 (Fig. 3.2). The
telescopes are set up such that the beams are slightly converging, to compensate for the
fact that at each surface of the the vacuum chamber windows 4\% of the power is lost.
This way the retro-reflected beams strike the MOT center with the same intensity as the
incoming beams. The intensity of the x and y-beams are about 5 mW/cm\(^2\) and for the
z-beam it is about 7 mW/cm\(^2\). These intensities are a few times larger than the saturation
intensity of the 5\(^2\)S\(_{1/2}\)–5\(^2\)P\(_{3/2}\) transition, which is \(I_s=1.64\) mW/cm\(^2\).

At all three incoming beams, as well as at the three end mirrors for the retro-reflection,
we placed quarter waveplates to change the polarization from linear to circular. At the in-
coming beams the plates have to be set at the correct angles to make either right handed or
left handed circular polarization, see Fig. 3.6. At the retro-reflection mirrors, the rotation
angle of the waveplates can be set arbitrary. This is because the light goes through the
waveplate twice in opposite direction, which ensures the polarization remains the same
with respect to the direction of the light.
3.2 The Magneto-Optical Trap

3.2.2 Measurement of the MOT density

We would like to have an inter-particle distance that is at least the same order of magnitude to the separation of the Rydberg volumes in the experiment, such that the distance between two atoms in the separate volumes is approximately equal to the distance between the volumes. If we take 10 $\mu$m for the inter-particle distance, we obtain a density of approximately $2 \times 10^8$ cm$^{-3}$.

The number of atoms in the MOT and the size of the cloud is obtained by measuring the fluorescence coming from the cloud of atoms. The cloud is imaged on the Prosilica EC750 CMOS camera, with an f=50 mm lens at a distance of about 25 cm from the MOT. A typical picture of the MOT cloud is depicted in figure 3.9.

![Figure 3.9: A picture of the fluorescing MOT with 1.8(4)$\times 10^7$ atoms.](image)

The number of atoms is determined through the relation [71]

$$N = \frac{F}{\eta \hbar \omega \rho \Gamma}, \quad (3.1)$$

where $F$ is the fluorescence signal, $\eta$ the detection efficiency, $\hbar \omega$ the photon energy, $\rho$ is the fraction of atoms in the 5p state and $\Gamma = 5.98$ MHz the natural linewidth of the 5p state.

We calibrated the camera by illuminating the CMOS chip directly with a small and weak, 780 nm laser beam. The chip has 480$\times$752 pixels and each pixel has an output value between 0 and 255 that represents the amount of light. We compared the integrated pixel signal divided by the camera’s shutter time with the power of the laser beam, measured with the Newport 818 power meter, for several laser intensities. We obtained 51(5) aJ per unit of integrated pixel signal. This energy corresponds approximately to 200 780–nm–photons.

The detection efficiency $\eta$ is dominated by the geometrical efficiency. This is the fraction of all spontaneously emitted light that hits the camera. In front of the camera lens is a 12.5(1) mm pinhole, which is at 22.0(0.5) cm from the MOT. Since the spontaneous emission is isotropic, the efficiency is the fraction of the pinhole area over the area of the
total imaginary sphere with radius 22 cm. Including the loss of 4% at each surface of the vacuum chamber window we obtain an efficiency of $\eta = 1.86(0.12) \cdot 10^{-4}$.

The excited state fraction is not straightforward to obtain. We use

$$\rho = \frac{1}{2} \frac{C^2 I/I_s}{1 + C^2 I/I_s + (2\delta/\Gamma)^2},$$

(3.2)

Here $I_s = 1.64 \text{ mW/cm}^2$ is the saturation intensity for the Rb $5S_{1/2} \to 5P_{3/2}$ transition, $I = 30(3) \text{ mW/cm}^2$ is the total intensity of all six laser beams together and $\delta = 13(1) \text{ MHz}$ is the detuning of the laser. $C$ is the average Clebsch-Gordan coefficient, taking into account all transitions between the various Zeeman sublevels, laser polarizations, etcetera. This number is rather challenging to calculate, however, it is likely that $C^2$ has a value between 0.4 and 1. We obtain an excited state fraction of $\rho = 0.20(5)$.

The number of atoms we obtain from the total signal in picture 3.9 is $1.8(4) \cdot 10^7$. The size of the cloud is obtained by fitting a 2D Gaussian profile. The pixel size corresponds to $19.4(2) \mu\text{m}$ in reality, calibrated from the 14 mm holes in the plates $P_1$ and $P_2$, visible in the complete $752 \times 480$ picture. We obtained $0.54(6) \text{ mm}$ for the $1/\sqrt{\epsilon}$ diameter, which results in a center density of $5.5(2.2) \cdot 10^{10} \text{ cm}^{-3}$. This density is sufficient for the experiment. It corresponds to an inter-particle distance of approximately $1.5 \mu\text{m}$.

### 3.3 Rydberg excitation

For the excitation to the Rydberg states we use pulsed dye lasers. The benefits of these lasers are that they are relatively easy to operate, the pulse energy (2–3 mJ) is more than enough for our purposes and the pulse length of 8 ns is much shorter than the typical interaction time that we study. Furthermore, these type of lasers have been widely used for decades already, and we happened to own two of these lasers already. We use a wavelength of 594 nm, such that two of these photons can excite from the $5s$ ground state to a chosen Rydberg $s$- or $d$-state (see Fig. 3.3). This wavelength is also easy to work with, since the needed laser dye, Sulforhodamine-B, is one of the most efficient, common and long-lived. Another option for rubidium used in other laboratories is a continuous or pulsed laser at 480 nm, which would excite to Rydberg states from the $5p$ state, the excited state present in the MOT cloud. One of the most important benefits of a continuous 480 nm laser is the much narrower linewidth. In the following sections we will describe some properties of the two-photon process, followed by a section about the working principles of the dye lasers and the optics setup used. After that, two typical wavelength scans are described, which are used to calibrate the laser, followed by a section about the determination of the size of the Rydberg volumes.

#### 3.3.1 Two-photon excitation

There are some obvious differences between a two-photon process and a one-photon process. The Rabi frequency of a two-photon process follows from the dressed-state approach [23] and is given in a simplified form by

$$\Omega = \frac{\Omega_1 \Omega_2}{2\delta},$$

(3.3)
with $\Omega_1$ the Rabi frequency of one photon from the ground state to the intermediate state and $\Omega_2$ the Rabi frequency of the other photon from the intermediate state to the Rydberg state and $\delta$ is the detuning from the intermediate state (we assume that the detuning to the final state is negligible). In reality there are a lot of intermediate states, and the Rabi frequency is the sum over all such terms. In the case of the two-photon excitation of the rubidium nd- or ns-series the $5p$ state is assumed to play the most important role. We need quite a large intensity (typically $\sim 100 \text{ MW/cm}^2$), because the $5p$–nd/ns transition dipole moment is rather small, but mostly because the detuning from the $5p$ state is very large. This high intensity can easily be obtained from a pulsed laser.

The excitation probability scales with the intensity squared, opposed to a single photon process, where it scales linearly with the intensity. This follows from the fact that the excitation probability is proportional to $\Omega^2$ for low power and/or short excitation times and $\Omega_1$ and $\Omega_2$ are proportional to the electric field amplitude. An interesting property for a two-photon process in a focused beam is that the density of Rydberg atoms along the length of the beam decreases outside the focus, whereas in a single photon process the line density of Rydberg atoms is constant over the length of the beam. For this reason we can regard the Rydberg volume simply as a cylinder or cigar-shaped ellipsoid and we can align the focus on the MOT cloud by simply looking for the region with the largest amount of Rydberg atoms. Another difference is that the width of the Rydberg volume is $\sqrt{2}$ smaller than the laser waist for a two-photon process. A drawback from using a two photon process is that also a three photon process is not unlikely to occur, which will ionize the atoms. The resulting electrons and ions could disturb the experiment. We use low intensities, such that less than 1% of our field-ionization signal is from a three-photon ionization process.

3.3.2 Nd:YAG pumped pulsed dye lasers

We have two sets of dye lasers, since we want to have two different Rydberg states in separate volumes (Fig. 3.1). Both dye lasers are pumped with Q-switched Nd:YAG lasers. In these lasers neodymium-doped yttrium aluminum garnet (Nd:YAG) is the active medium. The Nd ions in the crystal are excited by a flash lamp during approximately 200 $\mu$s. The excited ions immediately drop down to a long-lived state, the upper level of the lasing transition. The most probable transition from this state is at 1064 nm to the lower level of the lasing transition, from which it decays to the ground state. At the point of maximum population inversion an electro-optic Q-switch is triggered. This Q-switch consists of a Pockels cell and a polarizer positioned at one of the cavity mirrors. When a high-voltage is applied to the Pockels cell, the polarization of the light is changed, and therefore the output through the polarizer. When the Q-switch is triggered, the cavity is operational, and the 1064 nm light is fed back into the active medium resulting in lasing. The output pulse is about 8 ns long and the peak optical power is tens of megawatts [89, 87].

The laser light is frequency doubled in a nonlinear crystal KD’P (potassium dideuterium phosphate). The light and the crystal should be "phase matched", which means that the index of refraction for the fundamental and the second harmonic should be the same. The resulting 532 nm (green) light is used as a pump for the dye laser.

The Quanta-Ray PDL-3 Pulsed Dye Laser (in short: PDL) [88] and the Lambda
Physik ScanMate Dye Laser (SCM) have the same principle of operation, which we shortly discuss below. In the dye lasers, the green pump beam is split into two parts and both beams are focused with a cylindrical lens onto a dye cuvette. The first beam, the so-called oscillator beam, will start the lasing; the second beam will amplify it. The laser dye, Sulforhodamine-B dissolved in methanol, absorbs the pump light and emits light in the 580–620 nm range. This orange light is amplified in the laser cavity, passing through the gain medium – the laser dye – several times. The laser cavity consists of a grating at one end and an output mirror at the other end. The grating is hit under grazing incidence after being expanded (Littrow-scheme), where a higher order reflection is fed back into the gain medium. The angle of the grating determines the exact wavelength of the laser light, which can be set at 1 pm precision by a stepper motor. A problem that arises in high gain lasers like these is amplified spontaneous emission (ASE). This ASE gives a broadband background contribution to the laser pulse. Both the SCM and PDL laser have different methods to minimize the contribution of ASE, which we will not discuss here. The output beam of the oscillator cavity is now again guided through a dye cuvette, and amplified by the second beam of green laser pump light. In both lasers an output pulse energy of 2–3 mJ is obtained. The SCM laser has an option of an intra-cavity etalon, which reduces the linewidth of the laser light. An etalon (or a Fabry-Pérot interferometer) consists of two parallel reflecting surfaces and the transmission depends on the wavelength and the angle of incidence. At a certain angle, only a narrow distribution around the wanted wavelength is transmitted. The angle of the etalon is automatically synchronized with the grating angle by means of a stepper motor. The resulting linewidth of the lasers is determined in section 3.3.3.

We want to excite the atoms to Rydberg states in a small region in space (∼10 µm), in order to study the spatial dependence of the interaction. The smallest focus size is obtained with the largest beam size and the strongest lens. For gaussian beams the waist, defined as the $1/\sqrt{e}$ diameter of the intensity profile, is given by

$$w_0 = \frac{2\lambda}{\pi F d},$$

with $\lambda$ the wavelength of the light, $F$ the focal length of the lens and $d$ the diameter of the beam at the lens. In practice the waist is larger, because the beam is usually not perfectly Gaussian and, to a much lesser extent, because of lens errors. The lens we use is an achromatic doublet of $F = 200$ mm, which limits the effects of spherical aberration. Because we use the lens outside the vacuum chamber for practical reasons, we are forced to use a long focal length $F$. For the diameter of the beam we use 8 mm. A much larger beam is impractical, moreover, lens errors become more important, since the diameter of the lens is only 30 mm. The beam profile of both lasers is not nicely Gaussian. It is strongly elongated and has some brighter and darker areas on the outside. Fortunately, the fluence of the laser pulses is much larger than necessary, so after enlarging the beam size by a factor 8 in a telescope, we select only a small fraction of the center of the beam with an iris diaphragm, set to 8 mm (see figure 3.10).

Both beams are joined on a 50/50 beam splitter cube, where one of the exiting beams is directed onto achromatic lens and focused on the MOT cloud, the other exiting beam goes through a similar lens and is focused onto a Philips CMOS webcam (from which
3.3 Rydberg excitation

Figure 3.10: A schematic drawing of dye-lasers setup. The ScanMate laser beam (SCM) is first expanded by a factor 8 in a telescope, where after a center section is cut out by an iris diaphragm (dia) set to 8 mm. The second telescope lens is moved laterally by a stepper motor (SM). The power in the beam is controlled by a polarizing beam splitter cube (PBS) together with a half wave plate. The PDL laser beam is split off in a PBS first to control the power and then enlarged by a similar telescope. Both beams are joined in a 50/50 beam splitter cube (50/50), where both output beams are focused with similar lenses on the MOT cloud and through a gray filter on a webcam (cam). The triangles in the figure represent prisms, which are used as mirrors.

the original camera lens is removed), to image both waists. Two prisms in the PDL beam path, functioning as mirrors, can be precisely adjusted to overlap both beams just after the 50/50 cube as well as at the webcam. The second lens of the telescope in the ScanMate laser beam is attached to a stepper motor, which can move the lens laterally. Since this lens has the same focal length as the final focusing lens, a displacement of this lens translates to the exact same displacement of the focus in the MOT as well as on the webcam. The exact width of the beams as well as the separation between the beams is calibrated by a two-photon mixing process using one photon from each beam described in section 3.3.4.

3.3.3 Wavelength scan

To calibrate the wavelength and the linewidth of the laser we use the known spectra of the atoms as a reference. The angle of the grating in the laser can be automatically tuned with a stepper-motor, thereby scanning the wavelength. These scans are performed on a daily basis, because the laser frequency depends on the exact atmospheric pressure as well as the humidity, furthermore the laser itself might be drifting. During these scans the laser focus is positioned approximately 1 cm behind the MOT cloud, such that a large atom number is illuminated at low fluence. In this way we can assume an unsaturated excitation and a negligible AC-Stark shift or broadening, while keeping a large signal. Within a few microseconds the atoms are ionized by an electrical field pulse and detected by a micro-channel plate. The total signal of the particles is counted and this number is depicted as a function of wavelength in figure 3.11.
Figure 3.11: Result of two wavelength scans. a) performed with the PDL laser b) performed with the ScanMate laser with intra-cavity etalon.

In figure 3.11 we observe a regular peak structure, connected to the binding energies of the concerning Rydberg states. Starting from the 5s ground state, only s- and d-states can be excited with two photons, because each photon has an angular momentum of 1. We observe that the excitation probability of the d-state is about five times larger than of the s-state. The binding energy of the states given by

\[ E_n = -\frac{\text{Ry}}{n'^2}, \]

with \( \text{Ry} = 109736.605 \text{ cm}^{-1} \) the Rydberg constant for rubidium \cite{35}, and \( n' = n - \delta_\ell \), with \( n \) the principal quantum number and \( \delta_\ell \) the quantum defect depending the angular momentum quantum number \( \ell \) of the state (see also chapter 2). The resonance wavelength is given by \( \lambda = 2 \times 10^7 / (E_n - E_g) \) with \( E_g \) the energy of the ground state, equal to the ionization potential of rubidium \( E_g = -33690.798(2) \text{ cm}^{-1} \) \cite{35}. The factor \( 10^7 \) arises from the conversion from cm to nm; the factor 2 from the 2-photon process. The wavelength has to be corrected for the index of refraction in air, since the laser operates in the air, while the atoms are in vacuum. We use \( \lambda_{\text{air}} = \lambda / \nu \). For this index we used \( \nu = 1.000276 \), where the last digit is uncertain, due to the dependence on atmospheric pressure and temperature \cite{1}.

The wavelength scan is fitted with a series of Gaussian peaks centered around the expected values of \( \lambda_{\text{air}} \) with equal widths, heights and vertical offset. We assume the calibration error of the wavelength is constant; on this scale a refractive index error would give a constant error. The resonance peaks come closer together for higher \( n \), which enables us to label the resonances with the right quantum numbers. If the scan consists of at least three succeeding \( nd \) peaks the labeling is already reliable. From the width of the peaks we can deduce the linewidth of the laser. The fitted widths of the peaks (2\( \sigma \) or the 1/\( \sqrt{e} \) full width) in the PDL scan is 6.4(5) pm, corresponding to a linewidth of 0.181(14) cm\(^{-1}\) or 5.4(4) GHz. The linewidth specified in the manual \cite{88} is 0.07 cm\(^{-1}\); much less than what we measure. This could be due to alignment errors of the laser. The SCM scan is performed with the intra-cavity etalon in place and fitted with double Gaussian peaks, a narrow peak on top of broad peak, which we think reflects the frequency
profile of the laser. Here we get 9.2(1.6) pm for the broad peaks and 0.60(4) pm for the narrow peak, corresponding to 0.017(1) cm$^{-1}$ or 0.51(3) GHz. This linewidth is actually smaller than the value specified in the manual [56], which is 0.024 cm$^{-1}$ for the laser with intra-cavity etalon. Both linewidths are suitable for the 41d-49s experiments, which are described in chapters 5, 6 and 7, where we use the PDL for the 41d excitation and the SCM for the 49s excitation.

### 3.3.4 Size of the Rydberg volumes

The focused laser beam excites the MOT atoms to Rydberg states, creating an elongated volume of Rydberg atoms. The length is determined by the size of the MOT cloud, because outside the MOT cloud the rubidium density is orders of magnitude lower; this size is $\sim$500 $\mu$m. For the diameter of the Rydberg volumes $1/\sqrt{2}w_n$ is expected for unsaturated excitation, (with $w_n$ the laser beam waist), because of the two-photon process. However, if the excitation is saturated, the diameter is slightly larger. The exact diameter is important for the spatially resolved measurements (chapter 5).

![Figure 3.12: The number of atoms versus laser pulse energy $E$. Fig. a) depicts the number of 41d atoms excited with the PDL laser, Fig. b) depicts the number of 49s atoms excited with the SCM laser. The red lines are fits through the data, with the fit parameter $E_{\text{sat}}$ indicated.](image)

To determine the saturation of the 41d and 49s excitation we measured the number of Rydberg atoms versus laser pulse energy (see Fig. 3.12). The density profile of the excited atoms is modeled as

$$\rho(r) = \frac{1}{2} \left(1 - e^{-F(r)/F_{\text{sat}}^2}\right), \quad (3.6)$$

with $F(r)$ the fluence profile of the laser and $F_{\text{sat}}$ the saturation fluence. For a Gaussian laser beam $\rho(r)$ becomes

$$\rho(r) = \frac{1}{2} \left(1 - e^{-\left(\frac{sr^2}{w^2}\right)}\right), \quad (3.7)$$

with $w$ the waist of the laser beam, and $s$ the saturation parameter $s = E/E_{\text{sat}}$. $E$ is the pulse energy of the laser and $E_{\text{sat}}$ the saturation energy $E_{\text{sat}} = \frac{1}{2}\pi w^2 F_{\text{sat}}$. The $1/\sqrt{e}$ diameter of
the density profile is

\[
d = w \sqrt{2 \log(s) - \log \left( \frac{1}{2} - \log \left( \sqrt{e} - 1 + e^{-s^2} \right) \right)}.
\] (3.8)

The measurements (Fig. 3.12) are fitted to the integral of the density profile \(\rho(r)\). For 41d we get \(E_{\text{sat}} = 2.6(0.1) \mu J\) and for 49s \(E_{\text{sat}} = 3.1(0.2) \mu J\) (see Fig. 3.12). At first sight one would expect a lower value for 41d, but the linewidth of the 41d laser is about twice as large, leading to a similar value (the SCM laser was used without the intra-cavity etalon). In the experiments described in chapters 5, 6 and 7 we use \(E_{\text{tot}} = 5.0(1) \mu J\) for 41d and 3.0(1) \mu J for 49s. Assuming Gaussian beams the diameter of the 49s beam becomes 0.85 \(\times w_{49}\) and the 41d beam 1.19 \(\times w_{41}\) (see Eq. 3.8).

The laser waists are determined by a so-called two-photon overlap measurement. For this we detune the 49s laser by 20 GHz to the blue and we make sure that the laser pulses overlap in time, such that absorbing one photon of each laser leads to excitation to the 44d state. By moving the 49s beam over the 41d beam one obtains the convolution of the two laser beams in the 44d signal. A measurement is depicted in Fig. 5.3 and fitted to a Gaussian profile with a 1/\(\sqrt{e}\) full width of 22.8(6) \µm.

![Figure 3.13: The width of the photon overlap versus laser pulse energy \(\sqrt{E_1 E_2}\).](image)

The two-photon overlap measurement is in fact a convolution of the laser beams, or

\[
\int \frac{1}{2} (1 - e^{-\mathcal{F}_1(x,y)\mathcal{F}_2(x-x_0,y)/\mathcal{F}_{\text{sat}}}) dxdy,
\] (3.9)

which is known analytically if the fluence profiles \(\mathcal{F}_1\) and \(\mathcal{F}_2\) of both lasers are Gaussian. In Fig. 3.13 the width of the photon overlap peak is plotted versus the laser pulse energy and fitted with the width of the convolution Eq. 3.9. The agreement with the measured data in Fig. 3.13 show the validity of the model. The fit-parameters are \(E_{\text{sat}} = 2.55(10) \mu J\) and the unsaturated 1/\(\sqrt{e}\) diameter: 19.4(4) \µm. Assuming identical laser beams the waists are 1/\(\sqrt{2}\) times this number or 13.7(4) \µm. Including the saturation effect, this gives Rydberg volume diameters of 11.6(4) \µm for 49s and 16.3(5) \µm for 41d. Relevant for the experiments are not the widths of the individual Rydberg volumes, but the width of the convolution. The upper limit of this convolution is 23 \µm, without assumptions on
3.4 The role of electric and magnetic fields

3.4 The role of electric and magnetic fields

A static electric field brings the dipole-dipole interaction into resonance, at least in the case of an interaction between angular momentum states like $n_1 \ell_1 + n_2 \ell_2 \leftrightarrow n_3 \ell_3 + n_4 \ell_4$. In the case of an interaction between permanent dipoles, i.e. between atoms in parabolic states, a small field might be needed to align the dipoles or be able to excite to these states with a laser. The parabolic states have a dipole moment of $\frac{3}{2}nk$, which means their energy changes by $\frac{3}{2}nk$ when the field is changed by 1 atomic unit. If, for example, the interaction is studied between two subsequent $n$ states in the highest $k$ state ($k = n - 1$), the difference in their dipole moments is $3n$. For $n = 50$ this means that their energy difference has shifted by $\approx 190$ kHz for a field change of only 1 mV/cm. For the $41d + 49s$ interaction this number is $\approx 130$ kHz. These numbers are of the same order of magnitude of the dipole-dipole interaction strength itself, and thus with an inhomogeneous field, the resonances are significantly broadened.

Here we describe how we calculated and optimized the dimensions of the electric field plates and how we reduced the temporal noise of the field. In section 3.4.2 we describe how the magnetic field influences dipole-dipole interactions and in section 3.4.3 we describe how electromagnetic radiation of the background (black-body radiation) reduces the lifetime of the Rydberg atoms, together with their natural lifetime.

3.4.1 Electric field

We aim for a design of the electric field plates, such that the field is nicely homogeneous and the atoms that are spread out over the MOT cloud experience the same electric field, with a maximum difference of 2 mV/cm, because this would give a broadening of about 200 kHz, approximately the same value as the interaction strength (Fig. 3.1). The MOT sets a few limits on the design of the field plates. We want the MOT beams to be at least 1 cm in diameter, so this is the minimum distance between the plates. Next we need to make a hole in the plates for the beam in the $z$-direction. We choose to have this beam under a slight angle so that it just misses the MCP and the electrons can go straight to the detector. So this hole will have to be slightly larger than 1 cm.
The electric field $\mathbf{F}$ between two field plates is calculated numerically in two dimensions. We apply Gauss’ law in vacuum $\nabla \cdot \mathbf{F} = 0$, and with $\mathbf{F} = -\nabla V$ we get Laplace’s equation

$$\frac{\partial^2 V}{\partial x^2} + \frac{\partial^2 V}{\partial z^2} = 0,$$

with $V$ the electric potential or voltage $^{[45]}$. The calculation is done with the finite element method using the program FreeFEM. This method solves partial differential equations very efficiently by using a triangular mesh.

The optimization of the field homogeneity was done in an iterative process. We optimized on the smallest curvature in both directions, giving the best homogeneity around the center region. In figure 3.14a the result of the calculation is plotted for an optimal configuration: two plates with outside diameter 5.5 cm, a hole of 1.4 cm diameter and a distance of 2.5 cm. This configuration is also the one we built in our setup (see Fig.
3.4 The role of electric and magnetic fields

3.2b). The calculation is done in cylindrical coordinates with a 0 V border at 8 cm from the center. This border distance has also been decreased to 4 and 6 cm, because in reality there might be grounded objects closer to the plates. The numerical result is fitted with a polynomial of ten orders in two dimensions. The black isopotential lines in the figure are drawn from this fit. To get an estimate of the influence of the dispenser block, we also performed a calculation in Cartesian coordinates with a strip added on the side at 0 V. The result is shown in figure 3.14b.

With 1 V on one of the plates, the field in the center is 0.371(5) V/cm, pointing in the z-direction, in other words the effective plate distance is 2.69(3) cm. The error arises from the uncertainty of the real distance between the plates in the vacuum chamber, which is 0.3 mm; the field does not change much with the different distances of the 0 V border in the calculation. The curvature in the z-direction is 10 mV/cm², but with the smaller border distance it becomes 20 mV/cm². In the calculation with dispenser a field of 14.5 mV/cm arises in the x-direction (note that this increases the total field by only 0.1%); the curvature is not increased. Furthermore, we can verify that within the MOT size of 0.5 mm diameter, the electric field is constant within 2 mV/cm, giving a broadening of less than 200 kHz.

Both plates P1 and P2 (Fig. 3.2b) are connected to separate power supplies. The power supplies used for chapters 5, 6 and 7 have a reduced noise level, below 3 mV, which translates to ~1 mV/cm or 130 kHz broadening. The Agilent 33250A arbitrary pulseform generator has a noise level that is low enough already. This power supply is connected to plate P2 and provides the constant or oscillatory fields to tune the dipole-dipole interaction into resonance. Plate P1 is connected to a home-built fast high-voltage pulse generator (SFI-box) to ionize the atoms for detection (see section 3.5). This power supply used to pick up a lot of noise from the environment. This problem was solved by integrating a line filter in the apparatus and shielding the apparatus, by building a metal box around it, serving as a Faraday cage.

3.4.2 Magnetic field

The Zeeman shift of the Rydberg states is of the same order of magnitude of the Zeeman shift of the ground state or 5p state, because it does not depend on the principal quantum number n. The Zeeman shift is given as \( \Delta E = m_j g_j \mu_B B \) (see section 3.2). Since we use low-\( m \) states the shift is of the order of \( \mu_B B \). The magnetic field in the MOT cloud with 0.5 mm size varies about half a Gauss and therefore the Zeeman shift gives a broadening to the E-field dipole-dipole interaction resonances of the order of 1 MHz, which is significant. For the measurements described in chapters 6 and 7 the MOT magnetic field is switched off just before the laser excitation to the Rydberg states. The MOT coils are switched off with a fast home-built IGBT switch. This ensures a low B-field environment during the interaction process of the Rydberg atoms. The exact procedure is further described in section 6.3, as well as the difference in broadening with and without the MOT magnetic field.

To compensate for the earth magnetic field of about 0.5 G we placed two large sets of compensation coils around the vacuum chamber as depicted in figure 3.2. The currents through the coils have been optimized for the lowest field in both directions, measured with a Stefan Mayer Instruments Fluxmaster. The remaining fields are below 0.14 G
throughout the chamber, resulting in a broadening of ~200 kHz. The low field is also important for the ionized electrons to fly straight to the detector.

### 3.4.3 Black Body Radiation and Natural Lifetime

A remaining field to disturb the sensitive Rydberg atoms is the black body radiation field. At room temperature this radiation is mainly in the GHz–THz range. These frequencies correspond to the energy spacings between the Rydberg levels, and therefore the radiation can induce transitions to energetically nearby states. An approximation of the black-body rate (the rate at which the concerned Rydberg state is depopulated) is given by [35]

\[
\frac{1}{\tau_{bb}} = \frac{4\alpha^3 k_B T}{3n^2},
\]

(3.11)

with \(\alpha\) the fine structure constant. The equation is accurate for large principal quantum number \(n\) (\(n > 15\)). Interestingly this rate depends on the principal quantum number only. For \(n\) between 40 and 50 we obtain rates between 8 and 12 kHz at 300 K.

Most relevant is the black body transition rate between 49s and 49p, because in the experiments described in chapters 5, 6 and 7 this transition is used as a measure for the dipole-dipole interaction rate. The stimulated emission and absorption rate for black body radiation from the initial state \(|n\ell\rangle\) to the final state \(|n'\ell'\rangle\) is given by [35]

\[
\frac{1}{\tau_{n'n',n\ell}} = \frac{4\alpha^3 k_B T}{3} \frac{\ell_{\text{max}}^2}{2\ell + 1} \omega_{n'n',n\ell}^2 |\langle n'\ell' \mid r \mid n\ell \rangle|^2,
\]

(3.12)

where \(\ell_{\text{max}}\) is the larger of \(\ell\) and \(\ell'\). Note that for \(\langle n'\ell' \mid r \mid n\ell \rangle\) only the radial part of the wavefunctions is needed. We have \(|\langle 49p \mid r \mid 49s \rangle| \approx 2420(30)\) and \(\omega_{49p,49s} = 2\pi 32.4(4)\) GHz, resulting in a rate of ~ 2.9(2) kHz.

The atoms can also spontaneously decay to the ground state or another lower lying state. The spontaneous decay rate depends on \(n^*\) and \(\ell\). It is given by [35]

\[
\tau = \tau_0 n^{*\alpha},
\]

(3.13)

where \(\tau_0\) is for rubidium 1.43, 2.76, 2.09 or 0.76 ns and \(\alpha\) is 2.94, 3.02, 2.85 or 2.95 respectively for the s, p, d or f states. For the relevant states in the described experiments we obtain \(\tau_{49s} = 9.1\) kHz, \(\tau_{49p} = 3.4\) kHz, \(\tau_{41d} = 13.3\) kHz and \(\tau_{42p} = 5.5\) kHz. So for these states, the black body rate and the natural lifetime are of the same order of magnitude, but most importantly both are much smaller than the typical interaction strength of 200 kHz.

### 3.5 Rydberg detection

The Rydberg atoms are detected with the method of State-selective Field Ionization (SFI) [35], which enables us to distinguish different Rydberg states. In hydrogen the outer electron is bound to the nucleus by a Coulomb potential, \(-1/r\) in atomic units. If an electric field \(F\) is applied in the \(z\)-direction the potential seen by the electron is

\[
V = -\frac{1}{r} + Fz.
\]

(3.14)
The potential has a saddle point on the \( z \)-axis at \( z = -1/\sqrt{F} \) with a value of \( V = -2 \sqrt{F} \). This means that if an electron has an energy equal or larger than \(-2 \sqrt{F}\) it can escape from the atom and the atom is ionized. If we simply use the binding energy at zero field, we have \(-1/(2n^2) = -2 \sqrt{F}\) and we obtain

\[
F = \frac{1}{16n^4}.
\] (3.15)

However, when the field is increased the Stark effect will play a role, leading to a different ionization field. The minimum and maximum Stark shift is approximately given by \( \Delta E = \pm \frac{3}{2} n^2 F \) and the ionization field becomes \( F = 1/(9n^4) \) or \( F = (11 - 4 \sqrt{7})/(9n^4) \).

In non-hydrogenic atoms all states are coupled leading to avoided crossings between the states. Depending on the speed at which the field is increased the atom ionizes at a different field value. In an experiment performed with sodium atoms in the d-state \([48]\) two peaks are observed in the field-ionization signal. Part of the atoms ionize at \( F = 1/(9n^4) \), because the d-state strongly couples to a state with a minimum Stark shift. The other part ionizes around the classically expected value of \( F = 1/(16n^4) \). The first part follows a (mainly) diabatic path, because this state with the minimum Stark shift ignores all the couplings and when the field is increased, the atom stays in the same quantum state. The last part follows an adiabatic path, because the atoms remains at approximately the same energy and it only wiggles a bit through the Stark map (see Fig. 2.2). In case of rubidium, however, a two-peak structure in an SFI measurement is not visible, probably because the couplings between the states is stronger than in sodium. The states ionize at a value somewhere between \( 1/(16n^4) \) and \( 1/(9n^4) \).

In our experiments we create a field ramp by applying a voltage ramp on the field plate P1 (see Fig. 3.2), provided by a home-built fast high-voltage pulse generator (SFI-box). The maximum voltage can be set up to 2 kV (resulting in a field of \( \sim 750 \) V/cm) and the rise time is usually set around 10 \( \mu \)s. This field ramp is not exactly linear in time, but has a slight curvature. In some cases we used an Agilent 33250A arbitrary pulseform generator, amplified by a home-built fast high-voltage amplifier (1 MHz bandwidth, 400 V output), which gives a cleaner, linear ramp, but has a lower maximum voltage. The field pulse is triggered just after the Rydberg atoms are created or after a desired interaction time. The released electrons fly toward a micro-channel plate detector and are detected (see Fig. 3.2).

The Hamamatsu micro-channel plate detector (MCP) consists of two slabs with a regular array of small channels, that function as electron multipliers under a strong electric field (we apply 2.3 kV). If an electron hits one channel, it starts a cascade of electrons that propagates through the channel, which amplifies the original signal by a factor about \( 10^3 \). After the second channel, \( \sim 10^6 \) electrons hit an anode and this current peak is recorded by an HP Infinium oscilloscope. Some reflections appear in the electronics, such that each

\*In the Stark map Fig. 2.2 none of the lines actually cross each other. The adiabatic path is when one would exactly follow one of the lines depicted. These lines only wiggle a bit, but go approximately horizontal and stay at the same energy as at zero field (especially at high fields and higher \( n \)). The diabatic path would be visible if looked at the figure from a distance and one recognizes the fan structure. A diabatic path would follow these straight lines. Which path the atom follows depends on the speed at which the field is increased, according to Landau \([57]\) and Zener \([107]\).
Figure 3.15: An example of a non-averaged scope trace. Indicated with red circles are the electron hits as recognized by the described procedure.

electron that hits the detector results in a damped oscillation. We set the oscilloscope to the 30 MHz bandwidth limited setting (a bandwidth filter) and the signal appears as a single peak. Most MCP’s have an electron detection efficiency of about 50%.

To get a reliable number of detected electrons we save all single scope traces and count the single peaks in the signal afterward. This method is favorable over simply averaging the signal if the signal to noise ratio is very low. An example of a scope trace is given in figure 3.15. The analysis consists of finding local minima, sticking out below -1 mV. The minima are then selected on basis of the slope (one data point has to be 10% lower than the preceding point) and the height (10% below the nearby maxima). In the figure the fourth and the last hit might be incorrectly tagged as single electron hits. The analysis procedure could be further improved by filtering low frequencies out and defining better restrictions for the slopes and heights of the peaks. The arrival times of the electrons are saved and counted for the relevant time windows, that belong to the specific Rydberg states, calibrated in an SFI measurement.

In figure 3.16 measurements are depicted of the field ionization of d-states for various values of \( n \). For this measurement we used the home-built fast high-voltage pulse generator (SFI-box) connected to the field plate P1, triggered just after the laser excitation. The ramp is recorded via the other field plate P2, which is weakly capacitively connected to P1. The transfer function is first calibrated at a lower voltage, and we assume that the frequency dependence is negligible for these ramps. The average ramp speed is 36 \((\text{V/cm})/\mu\text{s}\). The measurements are averaged over 200 laser shots and the oscilloscope time trace is transformed to a signal versus field picture. In this transformation, the flight time of the electrons over 28 cm to the detector is also taken into account. With \( V \) the applied voltage on the plate P1, the potential energy of the electrons that start in the middle between the plates is \( \frac{1}{2}eV \), with \( e \) the electron charge. We can ignore the short flight between the plates and immediately transform the potential energy into kinetic energy \( \frac{1}{2}m(L/t)^2 \) with \( m \) the electron mass, \( L \) the flight path (28 cm) and the flight time \( t \) becomes 0.668 \( \mu\text{s} \sqrt{V} \). The ionization time, the time for the electron to find the saddle
3.6 Summary

We have built a setup with which we have control – up to a certain level – over dipole-dipole interactions between Rydberg atoms. With tightly focused laser beams we create narrow cigar-shaped volumes of Rydberg atoms, 11.6(4) µm resp. 16.3(5) µm in diameter. The atoms remain in position during the µs experiment, because they are ultra-cold (83(7) µK, \(\bar{v} = 0.14 \mu m/\mu s\)), due to the fact that we use magneto-optically trapped atoms. Consequently, we have obtained control over the position of the atoms in two dimensions for a duration of tens of microseconds. The exact state of the Rydberg atoms is controlled by the frequency of the laser light, which is accurate to 5.4(4) GHz resp. 0.51(3) GHz, clearly resolving the \(n_d\) and \(n_s\) states around \(n = 50\). For the atoms in both volumes to undergo resonant dipole-dipole interaction with each other, the resonance condition should be accurately fulfilled. Since the dipole-dipole interaction itself has a strength of the order of 100’s of kHz, the broadening due to background fields should be of the same order of magnitude or less. This is obtained by applying an accurately homogeneous electric field (curvatures below 20 mV/cm²) with a low noise level (~1 mV/cm) and by having a low magnetic field (below 0.14 G). The respective broadenings are 260, 130 and 200 kHz, all three comparable to the typical interaction strength. The Rydberg atoms are probed by ionizing them in a field ramp and detecting the released electrons. Around \(n = 50\) we are capable to resolve the principle quantum number of the Rydberg states, due to the different field values at which they ionize. This enables us to probe the dipole-dipole

![Figure 3.16: Signal of the Rydberg atoms in arbitrary units versus ionization field. In fact the depicted data are time traces transformed to field, using the field ramp.](image)

point in the potential, is negligible. As a result, the average field values of ionization (depicted by the black dots) vary here from \(1/(12.4n^4)\) for 50d to \(1/(12.8n^4)\) for 45d. But most importantly, it shows that we can accurately distinguish the Rydberg states of subsequent \(n\) in the \(n = 50\) range. In chapter 4 a novel method of SFI is described, in order to determine the position of the atoms simultaneously with the state. In [9] the state selective field ionization is studied for different linear ramp speeds.
interaction.
We present a technique for state-selective position detection of cold Rydberg atoms. Ground state Rb atoms in a magneto-optical trap are excited to a Rydberg state and are subsequently ionized with a tailored electric field pulse. This pulse selectively ionizes only atoms in for instance the 54d state and not in the 53d state. The released electrons are detected after a slow flight towards a micro channel plate. From the time of flight of the electrons the position of the atoms is deduced. The state selectivity is about 20:1 when comparing 54d with 53d and the one-dimensional position resolution ranges from 6 to 40 \( \mu \text{m} \) over a range of 300 \( \mu \text{m} \). This state selectivity and position resolution are sufficient to allow for the observation of coherent quantum excitation transport.

4.1 Introduction

In this chapter we present experimental results that are a first step towards exploration of the coherent evolution of systems of Rydberg atoms. We show that the position-dependent readout of $|0\rangle$ and $|1\rangle$ states is indeed possible. The possibility to create and measure Rydberg atoms in a position and state resolved way is demonstrated here by exciting a narrow region in a cold cloud of rubidium atoms to the 54d state. The one-dimensional position sensitive detection is done with a time-of-flight technique. Time of flight is our choice in preference to a position-sensitive multi-channel plate detector because of simplicity and compatibility with existing equipment.

The time-of-flight approach makes it difficult to combine these measurements with other time-resolved techniques. State resolution is therefore based on the threshold character of field ionization, rather than timing as in conventional ramped state selective field ionization (SFI) [35]. The combination of techniques leads to conflicting optimizations. In the remainder of this paper these problems will be quantified and solved.

4.2 State and position determination

We first focus on the state determination by SFI [35]. The electric field $E$ in which a Rydberg atom ionizes is roughly given by the classical ionization threshold, which is at an energy of $U = -2 \sqrt{E}$, in atomic units, and corresponds to a field of $E = 1/16n^4$. Due to Stark shifts, however, the energy of the state changes with the electric field. Therefore the ionization field is different. In non-hydrogenic atoms many Stark states couple with each other, forming a level scheme with many avoided crossings. The best state selectivity is achieved when the crossings are traversed either all adiabatically or all diabatically. In the latter case the field has to be ramped up faster than technically convenient, so we choose to ramp the field very slowly. In the adiabatic case the energy stays approximately the same and the ionization field will just be around $1/16n^4$.

For the time-of-flight method we use the same electric field that ionizes the atom. This electric field pushes the electron through one of the ionizing electrodes into a field-free flight tube. The time it takes for the electron to fly through the tube is a measure for its kinetic energy at the beginning of the tube, and therefore a measure for the potential energy at its starting position. At the position of the atom cloud the electric potential decreases linearly with position (see Fig. 4.1). Consequently the electric potential, and thus the arrival time of the electron on the detector is a measure for the original position of the Rydberg atom. To magnify the arrival time differences beyond the instrumental time resolution we use a long flight tube and take care that the electrons are slow during flight. This is achieved by setting a potential on the flight tube that is almost the same as the potential at the starting position. The arrival time of the electron as a function of the initial position $x$ is approximately

$$t(x) = L \sqrt{\frac{m_e}{2e(Ex + V_0)}},$$

(4.1)

with $L$ the length of the flight tube $e$ the charge and $m_e$ the mass of the electron, $E$ the electric field and $V_0$ the potential at $x=0$ minus the potential on the flight tube.
For the time-of-flight technique the time of ionization has to be well defined. This is in contradiction with the slow field ramp needed for the state selective field ionization. Therefore we use a more advanced scheme for the combination of the two techniques: a slow field ramp brings the field just below the ionization limit of the $|1\rangle$ state, followed by a fast pulse to go above the limit of the $|1\rangle$ state. In addition, this pulse has to stay below the ionization limit of the $|0\rangle$ state, in order to detect the atoms in state $|1\rangle$ exclusively.

**Figure 4.1:** Schematic representation of the components that provide the electric field together with equipotential lines, which are calculated numerically. The applied voltages are: -115 V on the first plate (P1), +10 V on the second plate (P2), -48 V on the flight tube and +90 V on the mesh in front of the MCP. Below is a graph of the electric potential on axis. Also depicted are the position of the MOT (black dot) and the dye laser beam.

### 4.3 Experimental Setup

To make sure the atoms don’t move on the relevant timescale of the experiment we use a magneto-optical trap (MOT) of $^{85}$Rb atoms as our cold atom source. A typical rubidium MOT has a temperature below 300 µK [100], which corresponds to an average velocity of 0.3 µm/µs. The atoms are loaded from a dispenser into the MOT, created at the intersection of three orthogonal pairs of counterpropagating $\sigma^+ - \sigma^-$ laser beams at the center of a magnetic quadrupole field. The background pressure is $3 \times 10^{-8}$ mbar. Charged particles from the dispenser are removed by deflection in an electric field of 50 V/cm, which is shielded from its environment. The laser frequency is tuned about 13 MHz below the $5S_{1/2} (F=3) \rightarrow 5P_{3/2} (F=4)$ resonance. A repumping laser beam tuned to the $5S_{1/2} (F=2) \rightarrow 5P_{1/2} (F=3)$ resonance is added at the MOT center. The cooling laser and the repumping laser are both Toptica DL100 diode lasers at resp. 780 nm and 795 nm and are locked to the rubidium resonance with Doppler-free FM spectroscopy. During all measurements
the cooling laser is blocked, so that all atoms are pumped to the $5S_{1/2}$ (F=3) ground state by the repumping laser. To create atoms in a Rydberg state we excite cold atoms with an 8 ns, $\sim 2 \mu J$ laser pulse at 594 nm by a two-photon process from the $5s$ state to $53d$ or $54d$ (resp. $|0\rangle$ and $|1\rangle$). This light is provided by a Lambda Physik dye laser pumped with a Spectra Physics frequency-doubled, Q-switched Nd:YAG laser with a repetition rate of 10 Hz. The linewidth of the dye laser pulse is $0.15 \text{ cm}^{-1}$, which is well below the energy spacing between $53d$ an $54d$ ($1.55 \text{ cm}^{-1}$). A small contamination of the nearest $s$-state, which lies $0.32 \text{ cm}^{-1}$ higher, can not be excluded but is of no further relevance. The dye laser beam is focused in the MOT cloud by a lens placed on a micrometer translation stage. The beam waist is determined to be approximately $23(1) \mu m$, measured with the knife-edge technique. The polarization of the light is parallel to the electric field that will be applied after the laser pulse. The Rydberg atoms are field ionized by applying voltage pulses on two field plates P1 and P2 spaced by 2.5 cm (see Fig. 4.1). The circular stainless steel plates have a diameter of 5.5 cm and have a 1.4 cm hole in the middle for the transmission of one of the pairs of counterpropagating MOT laser beams. The released electrons go through the positively charged field plate P2 into a 40 cm long stainless steel tube, which is at negative potential and serves to slow down the electrons. After a 45 cm flight the electrons are detected on a Hamamatsu Micro Channel Plate (MCP). A copper mesh is placed in front of the MCP. The mesh is 95% open and set at $+90 \text{ Volt}$.

Additional coils outside the vacuum chamber compensate for background magnetic field. This is important for the electrons to fly straight towards the detector. The axis of the magnetic quadrupole field for the MOT is in the same direction as the electric field between the field plates.

For the simultaneous position and state detection we use the pulse scheme depicted in the lower half of Fig. 4.2. Shortly after the dye laser pulse we apply a slow voltage ramp on the field plate P1 which goes from 0 V to -115 V in 1.2 $\mu$s. It then remains -115 V for 1.3 $\mu$s, while a 10 V pulse is applied on the other plate P2 with a duration of 30 ns and a rise time of 10 ns. After that the voltage on the plate P1 is increased to -230 V in 1.2 $\mu$s, to ionize all atoms that remained unionized. With the fast pulse we aim to ionize the upper state atoms exclusively at a well defined time. The slow ramp is provided by an Agilent 33240A arbitrary waveform generator amplified by a home-built amplifier system. The 10 V pulse is provided by an HP 8114A high power pulse generator.

For the calibration of the conversion from time – of – flight to position we shift the Rydberg – production volume along the flight path by moving the lens that focuses the dye laser beam in the MOT cloud and record the signal of the MCP for every lens position. The voltage on the flight tube is carefully tuned, such that the electrons created at the edge of the MOT nearest to the plate P2 are still clearly detected on the MCP. This usually means that they have a flight time of $\approx 1 \mu s$, or equivalently, an energy of $\approx 0.5 \text{ eV}$. At lower velocities the signal density drops.

Before describing our experimental results, we consider the effect of the timing of our experiment on the achievable position resolution. First the thermal spreading of atoms during the slow ramp of the electric field is approximately $0.9 \mu m$, assuming a MOT temperature of $300 \mu K$. A second concern is that atoms may move during the ramp due
4.4 Results

In the upper panel of Fig. 4.2 the MCP signal during the voltage pulses is depicted for two different dye laser wavelengths. The upper trace is the signal for $\lambda=594.166$ nm, which excites to the $54d$ state or the $|1\rangle$ state and the trace below this is the signal for $\lambda=594.193$ nm, leading to excitation of the $53d$ state or the $|0\rangle$ state.

In the upper trace of Fig. 4.2 three different peaks can be distinguished, labelled a, b and c. Apparently atoms, originally in the $54d$ state, can be in different states at the time of ionization. This is because the atoms have traversed the avoided crossings in the Stark map differently during the slow field ramp, ending up in a distribution over blue and red states in the Stark manifold. The fast pulse sequentially ionizes some of these states within a very short time window (during the 10 ns rise time of the pulse). As the potential at the time of electron release is different, electrons acquire different velocities depending on whether they originate from red or blue states, which results in peak a (the

**Figure 4.2:** In the lower panel the applied voltage pulses on the plates P1 and P2 are shown. The laser pulse is shown as L. In the upper panel the MCP signal for the $|0\rangle$ and $|1\rangle$ state (resp. 53d and 54d) are depicted. The first two peaks a and b are both resulting from the fast pulse on P2 at t=0. The peaks c originate from the atoms that are left over. The voltage on the flight tube is -48 V and the position of the dye laser beam is 280 $\mu$m on the scale of Fig. 4.3.

to electrostatic forces on their dipole moments [97]. These forces can arise from the field gradient associated with small inhomogeneities in the applied electric field. In our setup the gradient is approximately 20 V/cm$^2$ at a field of 50 V/cm, which gives a displacement of less than 0.06 $\mu$m over the duration of the slow field ramp. Both numbers are negligible compared to the required position resolution of 20 $\mu$m.

4.4 Results

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Simultaneous position and state measurement of Rydberg atoms

Figure 4.3: Measured time-of-flight spectra for different laser focus positions $x_L$. The peaks correspond, apart from a minus, to the peak $b_1$ as defined in Fig. 4.2. The time $t=0$ is when the 30 ns pulse is applied to plate P2.

fast electrons released first) and peak b (the slowest electrons). Peak c results from atoms in a red state that remain unionized by the fast pulse and are ionized by the subsequent ramp. This slow ramp is used for estimating the ionization efficiency during the earlier pulses.

For the time-of-flight measurements we will focus on peak b, because this peak is most sensitive to the position of the Rydberg-production volume. Peak $b_1$ consists of 30% of the total signal of state $|1\rangle$. When the atoms are prepared in state $|0\rangle$, the same measurement should ideally give no signal ($b_0$). From our measurements we find an upper bound that is 1% or 2% of the total signal. The ratio of the genuine and spurious $|1\rangle$ signal is therefore about 20. In future experiments we will attempt to improve this ratio further. The spurious signal could result from atoms that are actually excited to the $|1\rangle$ state, due to the finite laser linewidth, or from atoms in the $|0\rangle$ state that ionize in a smaller field.

Fig. 4.3 shows time-of-flight spectra for the state $|1\rangle$. The peaks correspond to the peak $b_1$ as defined in Fig. 4.2 for several positions of the dye laser focus. It can be seen that for faster electrons the peaks are narrower, but also start to overlap more. The peaks in the time-of-flight spectrum appeared to be narrowest when the duration of the fast pulse applied on plate P2 was 30 ns.

We transformed the time-of-flight spectra to the position domain using Eq. 4.1 with $L=40$ cm. As the potential is significantly more complicated (see Fig. 4.1) than assumed for the simple model of Eq. 4.1, we use an effective field $E$ and $V_0$ as fit parameters. Also we add a short offset time of 13 ns, based on numerical calculations, for the parts of the flight that are outside the flight tube. Our fit is based on more traces than depicted in Fig. 4.3, since the measurements were taken for lens positions 20 $\mu$m apart. The peaks in the position domain can be excellently fitted with gaussian profiles, as shown in Fig. 4.4. Optimal agreement of the center positions of the fitted gaussians with the known 20
micron between lens positions is obtained for $E=50.2$ V/cm and $V_0=-2.2$ V.

It can be seen that in the position domain the peaks get narrower for larger $x$ (i.e. "downhill" on the potential in Fig. 4.1), while the corresponding peaks in the time domain (i.e. later times) get wider. For large $x$ the broadening due to the finite width of the Rydberg-production volume $w$ (due to a finite laser beam waist) is dominant. For small $x$ the time broadening effect $\tau$ is dominant. This time width $\tau$ and not the Rydberg-production volume $w$ is the limit for the spatial resolution of our time-of-flight system. Therefore we decouple $\tau$ from the widths $\Delta x_{\text{TOF}}$ of the gaussians by fitting the two broadening effects to the following expression

$$\Delta x_{\text{TOF}} = \sqrt{w^2 + \left(\frac{dx(t)}{dt}\right)^2 \tau^2},$$

with $x(t)$ the inverse function of Eq. 4.1. In the inset of Fig. 4.4 $\Delta x_{\text{TOF}}$ is plotted against the center position $x_{\text{TOF}}$ together with the fitted dependence. The fitted parameters are $\tau=20(1)$ ns and $w=21(1)$ $\mu$m, which is comparable to $1/\sqrt{2}$ times the measured laser waist (the $1/\sqrt{2}$ factor originates from the two-photon character excitation).

In Fig. 4.5 we plot the center positions of the gaussians $x_{\text{TOF}}$ against the read-off position of the lens that focuses the dye laser beam into the MOT cloud ($x_L$), together with the line $x_{\text{TOF}} = x_L$. The data points show an excellent agreement with the straight line, which shows that the simple model Eq. 4.1 is well suited to convert flight times into position. The small horizontal error bar is an estimate of the read-off error ($2 \mu$m). The vertical error bars provide the spatial resolution of our system and are obtained by transforming the time width $\tau$ to position. The spatial resolution ranges from 6 to 40 $\mu$m.
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Figure 4.5: The center positions of the fitted gaussians of Fig. 4.4 $x_{TOF}$ plotted against the read-off position $x_L$. The solid line is $x_{TOF} = x_L$. The small horizontal error bar is an estimate of the read-off error (2 $\mu$m). The vertical error bars are obtained by transforming the fitted time width $\tau$ to position and give the spatial resolution of our system.

over a distance of 300 $\mu$m (1/e diameter), and it is better than 20 $\mu$m over a range of 150 $\mu$m.

4.5 Conclusions

We have demonstrated a technique for state-selective position detection of cold Rydberg atoms. The state selectivity is about 20:1 when comparing 54d with 53d and might be improved by using a narrower spectral linewidth of the Rydberg-exciting laser or by optimizing the electric field pulse sequence. The position resolution ranges from 6 to 40 $\mu$m over a distance of 300 $\mu$m. These n-state and position resolution are sufficient to allow for the observation of long range hopping transport by coherent dipole-dipole interactions in cold Rydberg systems [81].
5 Spatially resolved observation of dipole-dipole interaction

We have observed resonant energy transfer between cold Rydberg atoms in spatially separated cylinders. Resonant dipole-dipole coupling excites the 49s atoms in one cylinder to the 49p state while the 41d atoms in the second cylinder are transferred down to the 42p state. We have measured the production of the 49p state as a function of separation of the cylinders (0 - 80 µm) and the interaction time (0 - 25 µs). In addition we measured the width of the electric field resonances. A full many-body quantum calculation reproduces the main features of the experiments.

5.1 Introduction

In the experiment presented in this chapter, we control the spatial separation between Rydberg atoms in two dimensions and study the dynamics of resonant excitation transfer by dipole-dipole interactions. Distinct Rydberg states (49s and 41d) are created from a magneto-optical trap (MOT) of ground-state Rb atoms by pulsed two-photon excitation using two independent focused laser beams, separated by tens of microns (see Fig. 5.1). Dipole-dipole coupling causes a transition of a Rydberg atom (49s) in one volume to a higher state (49p) and a simultaneous transition of an atom (41d) in the second volume to a lower state (42p). This transition occurs over separations between the volumes of Rydberg atoms of up to 50 µm, over 10 µs time scales.

![Figure 5.1: Two pulsed laser beams excite cold atoms to Rydberg states. The 41d atoms in one volume interact with the 49s atoms in the other volume. The resonant dipole-dipole interaction giving rise to a change of states 41d + 49s → 42p + 49p is in the near field limit (R~40 µm ≪ λ=0.9 cm). We resolve the interaction in time and space by varying the beam separation.](image)

Dipole-dipole interactions in Rydberg atoms can be induced by tuning two transitions into resonance using a static electric field. A transition in rubidium which has particularly large transition dipole moments and a resonance at a low electric field (∼0.4 V/cm) is

\[ 41d_{3/2} + 49s_{1/2} \leftrightarrow 42p_{1/2} + 49p_{3/2}. \]  

The resonant transition frequency is 33 GHz. The strength of the dipole-dipole interaction is given by

\[ V = \left( \frac{\mu_1 \cdot \mu_2 - 3(\mu_1 \cdot \hat{R})(\mu_2 \cdot \hat{R})}{R^3} \right), \]

where \( \mathbf{R} \) is the distance vector between the interacting particles, \( \mu_1 \) (\( \mu_2 \)) is the dipole moment of the 41d\(_{3/2}\)\(\rightarrow\)42p\(_{1/2}\) (49s\(_{1/2}\)\(\rightarrow\)49p\(_{3/2}\)) transition. Both dipole moments are of the order of 1000\(a_0\)e. The transition probability \( P \propto \sin^2(Vt) \) shows quantum beats with a period of \( \sim 14 \) µs for \( R=40 \) µm (if \( \mu_1, \mu_2 \parallel \mathbf{R} \)). For short times (\( Vt \ll 1 \)), we can make the approximation \( P \propto \frac{t^2}{R^6} \). For dipoles on parallel lines at distance \( d \), the short-time probability averages to \( P \propto \frac{t^2}{d^5} \).

5.2 Experimental setup

A standard \(^{85}\text{Rb} \) MOT is used, which has a temperature of 300 µK or less. This temperature corresponds to an average speed of 0.3 µm/µs, so the atoms hardly move on the
5.2 Experimental setup

The cold ground state atoms (5s) are excited to a Rydberg state by an 8 ns laser pulse of 594 nm in a two-photon process. There are two separate lasers, one for each of the two Rydberg states, focused next to each other in the MOT cloud. The one that excites to the 49s state (from now on called the 49s beam or 49s laser as opposed to the 41d beam or laser) has a linewidth of 0.09(1) cm\(^{-1}\) and the 41d laser has a linewidth of 0.21(2) cm\(^{-1}\). The foci are imaged on a CCD camera, with a pixel size of 5.6 \(\mu m\). The 49s beam can be laterally moved by a motorized lens.

An electric field surrounding the MOT cloud is created by applying a differential voltage on two 5.5 cm diameter circular plates, 2.5 cm apart, perforated by a 14 mm hole to allow MOT beams and ionization products to pass. This electric field, parallel to the laser beam separation and polarization, brings the dipole-dipole transfer (Eq. 5.1) into resonance. After a variable interaction time an electric field pulse (from 0 to 150 V/cm in 5 \(\mu s\)) ionizes the atoms and the released electrons are detected by a micro-channel plate (MCP). This gives different electron arrival times for the 49p and 49s atoms; the signal of the 41d and the 42p states are not distinguished. We use the 49p signal as a measure of the dipole-dipole interaction. Every electron is timed separately (the detection efficiency for MCP’s is typically 40-70\%) and all data are averaged over 200 laser shots.

The initial number of measured 49s\(_{1/2}\) atoms per shot is 12.5(2.5) and the total number of atoms in the 41d volume is 70(10). Of these atoms 15(3) are in the 43p state and we presume the same number in the 42p state. We get 24(5) atoms in the 41d\(_{3/2}\) state and 16(3) atoms in the relevant 41d\(_{1/2}\) state, based on the ratio that is given by our laser polarization. All mentioned atom numbers do not take the finite detection efficiency of the MCP into account. During the experiment, the number of Rydberg atoms slightly decreases due to spontaneous decay (rates around 8 kHz). Secondly, black body radiation induces transitions to nearby lying Rydberg states with rates around 10 kHz [35]. Reference measurements are done with an isolated 41d beam, resulting in 0.5 49s atom and 0.5 49p atom, which are subtracted from the data.

The Rydberg volumes are cigar-like ellipsoids. Their length is determined by the size of the MOT cloud (~0.5 mm). For unsaturated excitation we expect that the diameter of the Rydberg volume is 1/\(\sqrt{2}\) times the laser beam waist, because of the two-photon process. However, laser fluence dependent measurements show that both excitations are slightly saturated in our experiment, resulting in slightly larger diameters. The laser waists are determined with a two-photon overlap measurement. For this we detune the 49s laser by 20 GHz to the blue and we make sure that the laser pulses overlap in time, so that absorbing one photon of each laser leads to excitation to the 44d state. By moving the 49s beam over the 41d beam one obtains the convolution of the two laser beams in the 44d signal. A measurement is depicted in Fig. 5.3 and fitted to a Gaussian profile with a 1/\(\sqrt{e}\) full width of 22.8(0.6) \(\mu m\). From another measurement, not saturated, we obtain a laser waist of 13.7(0.4) \(\mu m\) for each laser waist, assuming identical beams. Taking the measured saturation with increasing power into account, the Rydberg volume diameters in our experiment are 11.6(0.4) \(\mu m\) for 49s and 16.3(0.5) \(\mu m\) for 41d. Without assuming identical beams, the convolution of the two saturated Rydberg volumes (relevant for the experiment) has a full width of less than 23(1) \(\mu m\). Due to the finite temperature, the diameter of the cylinders increases at most 2 \(\mu m\) in 25 \(\mu s\). See also section 3.3.4.
5.3 Simulations

For the interpretation of the data, we simulated the populations in each state by performing a fully quantum calculation with a limited number of atoms fixed in space. The matrix elements were computed using standard angular momentum algebra and numerical integration for the radial matrix elements. We used the energy levels of Ref. [60] to determine the radial functions needed for the matrix elements. No adjustments were made to get better agreement with the measurements. We randomly placed 25 atoms in each cigar-shaped ellipsoid (Gaussian in 3D) and performed a series of calculations starting with one randomly picked 49s atom. We added neighboring atoms in the calculation until the time dependent probability for the 49s → 49p transition converged. We obtained good results with two 49s atoms and the two nearest 41d atoms, which are depicted in the figures; full convergence was obtained with one additional 41d atom. This demonstrates that many-body effects are important. Adding more electronic energy levels per atom or adding atoms initially in the 42p or 43p state slightly slowed down the transition times (∼20%).

5.4 Results

![Figure 5.2: The 49p fraction after 10 μs as a function of electric field for different beam separations; 21, 31, 41 and 51 μm. Solid lines are Lorentzian fits. Red dashed (20 μm), green dash-dotted (30 μm), blue dotted (40 μm) and purple dashed (50 μm) lines are simulations. Resonances are denoted as F₁ (0.38 V/cm) and F₂ (0.41 V/cm). On the top axis a relative energy scale is given.](image)
As a first experiment we monitored the $49p$ fraction $N_{49p}/(N_{49s} + N_{49p})$, resulting from the resonant dipole-dipole interaction, after 10 $\mu$s as a function of the applied static electric field. The measurements are performed with beam separations between 21 and 51 $\mu$m and are depicted in Fig. 5.2. Two resonances can be seen, due to a small difference in the Stark shift of the $49p_{3/2}$ $|m_j|$ states. At the field $F_1(F_2)$, $|m_j| = \frac{3}{2}$ is resonant. The $|m_j|$ splitting of the $41d_{3/2}$ state is not visible, because only $|m_j| = \frac{1}{2}$ is excited by the laser. The field values have a systematic error of 2%, due to the uncertainty in the effective plate distance.

The calculated resonances fit within this error, confirming that the creation of $49p$ in the $49s$ volume requires the tuning into resonance with the transition in the adjacent $42p$ volume. For the top axis a conversion of 127 MHz/(V/cm) is used, based on the calculated difference of the total polarizabilities of the initial and final states.

It is clearly visible in Fig. 5.2 that the resonance peaks become higher and broader as the distance between the foci is reduced (FWHM are resp. 18(2), 20(1), 29(1) and 29(1) mV/cm). This is consistent with the notion that the dipole-dipole interaction (Eq. (5.2)) gets stronger with shorter distance. These features are also reproduced in the simulations. However, the widths of the calculated resonance peaks are much narrower than observed in the experiment. This is mainly due to the magnetic field of the MOT ($\sim 1.4$ MHz). It will further be discussed in connection with the time dependent measurement, as well as the origin of the background of 0.1. The increasing background at shorter distance, visible in simulation and measurement, is due to an increase of overlap between the Rydberg volumes, as well as presumably many-body effects.

![Figure 5.3: The 49p fraction after 10 $\mu$s as a function of the position of the 49s beam at the resonance $F_1$ (●) and $F_2$ (□). Blue solid ($F_1$) and green dashed ($F_2$) lines are simulations. The measured beam overlap (▲) is fitted to a Gaussian (red dotted line). Above the graph CCD pictures of the laser foci are depicted corresponding to the horizontal axis tick marks. The log-log plot inset depicts the wings of the data together with a fit to $d^{-5}$.](image)

To investigate the distance dependance of the interaction, we tuned the field to each of
Spatially resolved observation of dipole-dipole interaction

The resonances and measured the transferred fraction after 10 $\mu$s as a function of the separation (Fig. 5.3). The most important result in this figure is that a range of distances exists, where the overlap of the lasers vanishes, while the interaction is still clearly present. This demonstrates dipole-dipole energy transfer between Rydberg atoms in separate volumes.

Fig. 5.3 also shows simulations of the experiment. The effective range of interaction is well reconstructed, but slightly overestimated in the calculation. The inset shows that the wings of the data follow a $d^{-5}$ behavior, as expected. A striking discrepancy with the simulation is that the experimental signal decreases where the beams overlap. This is because non-resonant processes take place in the high density 41d beam, reducing the number of 49s atoms from 12.5(2.5) to 10(2); we particularly observe 4(1) atoms in the 47p state, but possibly Penning ionization occurs as well [101]. Besides, there are 5(1) 44d atoms due to the two-laser excitation.* Within the accuracy of the experiment there is no significant difference between the resonance $F_1$ (●) and the resonance $F_2$ (□). The asymmetry of the measured curve might be due to a deviation from a Gaussian of the laser beam profiles or a decrease in the density for positive positions (~10%).

Next we will discuss the main result in this chapter, the temporal evolution of the interaction. In Fig. 5.4 the evolution of the 49p fraction is measured for various beam separations with the field tuned to the resonance $F_1$. The figure clearly shows that the rate of transfer slows down for larger distance, but even at a 50 $\mu$m separation an apprecia-
ble transfer is observed. The rates slow down because the interaction strength decreases strongly with distance (Eq. (5.2)). Quantum beat oscillations, which are expected on basis of the coherent coupling between atoms, are not observed. A spread in distances of the interacting atoms, and hence a spread in interaction strength, causes dephasing of these oscillations. The transition rate $1/\tau$ in the inset is based on the time $\tau$ where the measured 49p fraction has reached 0.17. It is plotted versus distance, slightly corrected for the finite cylinder width. The data points nicely fit the scaling law $d^{-5/2}$. The minor deviation might be related to the different asymptotes of the curves.

Fig. 5.4 also shows results of the simulations. The curves reproduce the experimental data in a qualitative way, but the calculated growth rate is slightly higher and the final production is somewhat different. An important factor to note is that temporal fluctuations in the electric field, that are fast with respect to the dipole-dipole interaction rate, will slow down the transfer rate. These fluctuations diabatically detune an interacting atom pair in and out of resonance, thereby reducing the fraction of time during which the interaction actually takes place. Indeed, in early runs of the experiment, where the high-frequency noise level of the field generated by the field plates was 10 rather than 2 mV/cm, the observed transfer times were a factor 2.8(5) longer. Moving ions in the MOT cloud might also contribute to electrical field noise. Furthermore, the initially present 42p and 43p atoms also slow down the transfer rate, as indicated by the simulations.

The final 49p fraction is not 50%, which one would expect on basis of binary interaction. This reduced transfer is due to time-independent broadening effects, mainly the magnetic field (see also Fig. 5.2). However, since the transferred fraction is also below 50% in the simulations, it is probable that also many-body effects play a role. The main process can be accompanied by interactions between the initial 49s and the reaction product 49p, as well as between 41d and 42p. These processes are always resonant and occur within a single cylinder. Spurious processes within the 49s beam can be observed when the 41d laser is turned off, depicted as $\triangle$ in Fig. 5.4. This is mainly due to black body radiation; the transition from 49s to 49p has a rate of 6.7 kHz [35], but possibly also non-resonant two-body processes play a role.

**5.5 Conclusions**

Up till now distance dependence of dipole-dipole interactions in Rydberg atoms has only been measured indirectly by varying the density. Here, we have taken the next step by having interaction between atoms in different volumes at a well defined separation and orientation. This is remarkable since an interaction is measured between two mesoscopic gaseous systems at a macroscopic distance. We map the interaction by varying the electric field, mutual separation and interaction time and our data fit the here presented simulations as well as a straightforward scaling law. The distance restriction is not strong enough to show the coherence of the underlying processes. Furthermore, due to the fact that only interactions at large distances play a role, this experiment is more sensitive to many-body interactions and external fields. When this approach is combined with position-sensitive measurements (chapter 4) it allows for many variations with more complexity and/or reduced dimensionality and hence coherent evolution.
Spatially resolved observation of dipole-dipole interaction
Radio-frequency driven dipole-dipole interactions in spatially separated volumes

Radio-frequency (RF) fields in the MHz range are used to induce resonant energy transfer between cold Rydberg atoms in spatially separated volumes. After laser preparation of the Rydberg atoms, dipole-dipole coupling excites the 49s atoms in one cylinder to the 49p state while the 41d atoms in the second cylinder are transferred down to the 42p state. The energy exchanged between the atoms in this process is 33 GHz. An external RF-field brings this energy transfer into resonance. The strength of the interaction has been investigated as a function of amplitude (0–1 V/cm) and frequency (1–30 MHz) of the RF-field and as a function of a static field offset. Multi-photon transitions up to fifth order as well as selection rules prohibiting the process at certain fields have been observed. The width of the resonances has been reduced compared to earlier results by switching off external magnetic fields of the magneto-optical trap, making sub-MHz spectroscopy possible. All features are well reproduced by theoretical calculations taking the strong AC-Stark shift due to the RF-field into account.

6.1 Introduction

In chapter 5 we investigated the dipole – coupling between Rydberg atoms in two separated volumes as a function of static electric field, minimum particle distance and interaction time. In the present chapter we report how a radio-frequency field can provide additional control over the interaction strength. Radio – frequency and microwave fields have previously been used to investigate the properties of both isolated and interacting Rydberg atoms e.g. in [17, 34, 108, 16, 73]. The techniques presented in this article will allow precision spectroscopy on 2 – atom states and might be useful in quantum computation schemes to minimize electric field inhomogeneities which influence the dephasing of qubits implemented as Rydberg atoms [17].

In the experiment presented in this chapter we create two spatially separated volumes of Rydberg atoms by exciting cold ground-state atoms from a magneto-optical trap. This is achieved by focusing two dye lasers tuned to different Rydberg states into the trap (see Fig. 6.1). This procedure creates a few tens of Rydberg atoms in each laser focus. After letting the atoms interact for a fixed period of time, they are field-ionized, providing information on the states present. From this one can determine the number of atoms in the initial and final states, and hence the fraction of atoms having interacted. The interaction is influenced by external fields during the interaction time.

![Figure 6.1: Schematic of the experiment. Fields are represented by the labelled arrows. The two upright areas symbolize the laser beams creating the Rydberg atoms. Only two atoms are shown for simplicity while in reality there are some tens of atoms in total. The Rydberg volumes are typically 25 µm apart at a width of 15 µm. The change of states of the Rydberg atoms is also depicted.](image)

Fig. 6.1 shows a schematic picture of the experiment. The photon associated with the dipole-dipole interaction is symbolized by the arrow between the two atoms. The other arrows represent external RF- and static electric fields. During the interaction time the interaction is tuned into resonance by these externally applied fields. An additional external field due to black body radiation is always present in the experiment.
Fig. 6.2 shows an illustration depicting the order of magnitude of the typical energy density of all time-dependent fields. Note that the energy density has been multiplied by $\omega$ giving units of J/m$^3$ as this leads to constant intervals $d\omega$ in the logarithmic plot. Also note that the scale of the y-axis covers more than 20 orders of magnitude. This shows that while there is a contribution of the black body-radiation in the background, the contribution of the dipole-dipole and RF-fields is much larger in the relevant frequency ranges.

![Figure 6.2: Energy density spectrum of the three different radiation fields, to which the atoms are exposed. The blue line (long dashes) represents the always and everywhere present black body radiation, depicted for 300 K. Resonances of the vacuum chamber and the metallic field plates are not taken into account for this illustration. The red line (solid) represents the photon that goes from one atom to the other, due to the dipole-dipole interaction at 25 $\mu$m. The line shape is lorentzian based on experimental parameters. The green line (dashed) represents the oscillating field that is externally applied.](image)

The process in our experiment can be interpreted as amplitude modulation of the dipole – field by the external RF-field. Surprisingly, the amplitude of the carrier wave (dipole-dipole radiation) is much smaller than the amplitude of the modulating RF-field.

### 6.2 Radiofrequency Assisted Dipole-Dipole Interactions

The initial states chosen in this experiment are 41d and 49s, created in separate volumes. They exchange a photon in a resonant dipole-dipole interaction forming the states 42p and 49p in one of the processes

\[
\begin{align*}
41d_{\frac{1}{2}, \frac{1}{2}} + 49s_{\frac{1}{2}, \frac{1}{2}} &\rightarrow 42p_{\frac{1}{2}, \frac{1}{2}} + 49p_{\frac{3}{2}, \frac{1}{2}}, \\
41d_{\frac{3}{2}, \frac{1}{2}} + 49s_{\frac{1}{2}, \frac{1}{2}} &\rightarrow 42p_{\frac{3}{2}, \frac{1}{2}} + 49p_{\frac{3}{2}, \frac{1}{2}},
\end{align*}
\]
Radio-frequency driven dipole-dipole interactions in spatially separated volumes

where the notation denotes $n_{j,m_j}$. Both the $41d_2$ and the $49p_2$ states split in an electric field into two substates $|m_j| = 1/2$ and $|m_j| = 3/2$. Note that the initial $41d_2$-state is not excited by the laser, because of its polarization, so it does not play a role in our experiment and was ignored in Eq. 6.1. The approach of using four different states makes it possible to control the distance between the atoms, by having separate volumes for the two different initial states. A position-resolved detection technique is then not necessary; state selective detection of the final $49p$-state only will probe the dipole-dipole interaction.

The binding energies and polarizabilities of the states from Eq. 6.1 are obtained from a calculated Stark map, using the Numerov method [109] and making use of the Rubidium-85 quantum defects given in [60, 38, 3]. In Fig. 6.3 the sum of the binding energies of the two atoms is plotted versus the electric field. Both the initial state ($41d + 49s$) as well as the final state ($42p + 49p$) undergo a purely quadratic Stark shift, which is much stronger for the final pp-state than for the initial sd-state. This is caused by the strong Stark shift of the $49p$-state.

![Figure 6.3: Schematic representation of two-atom energy versus electric field. An external RF-field of 10 MHz is symbolized by the arrows. At a purely static field of 0.38 V/cm and 0.41 V/cm the dipole-dipole transition is resonant. At 0.3 V/cm it is brought into resonance by one RF-photon of 10 MHz, while at 0.15 V/cm two such photons bring the system into resonance. The inset shows the avoided crossings due to dipole-dipole interaction at a separation of 25 μm.](image)

For the dipole-dipole transfer to become resonant the energy difference between the initial and final two-atom state has to be zero. This energy difference can be written as

$$W = W_0 - \frac{1}{2}\alpha F^2,$$

where $W_0$ is the zero-field energy difference, $W_0 = 25.15(13)$ MHz, $F$ the total electric field and $\alpha$ the difference polarizability, i.e. the difference in polarizability between the initial and the final two-atom state. The nonlinear term in the field is due to the purely quadratic Stark shift of the energy levels. The energy difference at zero field ($W_0$) is
already very small, which is one of the reasons we have chosen this particular process. The difference polarizabilities are \( \alpha_a = 347.04(4) \text{ MHz/(V/cm)}^2 \) for process (6.1a) and \( \alpha_b = 297.40(4) \text{ MHz/(V/cm)}^2 \) for process (6.1b); this makes the process resonant at the static fields \( F_a = 0.3807(15) \text{ V/cm} \) and \( F_b = 0.4113(16) \text{ V/cm} \) respectively.

For non-zero electric fields the quantum number \( \ell \) does not describe a proper eigen-state of the system. We label those states as having the angular momentum quantum number \( \ell \) that connect adiabatically to the zero-field states with that quantum number. However at an electric field of 0.4 \text{ V/cm} the mixing in of other \( \ell \)-states is still negligible.

The strength of the dipole-dipole interaction, and therefore half the size of the avoided crossing is given in atomic units by

\[
V = \frac{\mu_1 \cdot \mu_2 - 3(\hat{\mu}_1 \cdot \hat{R})(\mu_2 \cdot \hat{R})}{R^3},
\]

(6.3)

where \( \mathbf{R} \) is the distance vector between the interacting particles, \( \mu_1 \) is the dipole moment of the \( 41d \rightarrow 42p \) transition and \( \mu_2 \) the dipole moment of the \( 49s \rightarrow 49p \) transition. The dipole moments are rather large, of the order of 1000 \text{ a}_0 \text{ e}, which is another advantage of this particular process eqn. 6.1. The exact value and polarization of each dipole \( \mu \) depends on the \( \Delta m_j \) of the transition. The dipole is either linearly polarized (\( \mu = \mu_z \)) or circularly polarized (\( \mu = \mu_x \pm i \mu_y \)). For the field strength of the oscillating dipoles we typically obtain 4 \( \mu \text{ V/cm} \) at a separation of 25 \( \mu \text{m} \). We can consider the dipole-dipole interaction process as the exchange of a photon from one atom to the other. This photon has an energy of 32.8 GHz. The inset in Fig. 6.3 shows the avoided crossing due to the dipole-dipole interaction for this separation. The frequency of the quantum beat oscillation of the system (or the Rabi oscillation of the single atoms) is \( \omega_{QB} = 2V \approx 2\pi \times 200 \text{ kHz} \).

In the presence of an external RF-field the sd-state can couple to the pp-state even far away from the static field resonance, as symbolized by the arrows in Fig. 6.3. This is the case if the energy of the RF-photon equals the energy difference \( W \) between the states, so

\[
N \omega = W_0 - \frac{1}{2} \alpha F_{\text{eff}}^2,
\]

(6.7)
where $N$ is the number of photons involved in the transition and $\omega$ is the frequency of the external RF-photon. Note that in the case of a purely linear Stark shift the behavior is essentially different. In that case the time-average of the RF-field becomes zero and only the static field $F_S$ is relevant. The term $\frac{1}{2}\alpha F_{RF}^2$ in Eq. 6.5 is also called the AC-Stark shift.

### 6.3 Improved Experimental Setup

The setup used is similar as previously described in chapters 3, 4 and 5. A Magnetooptical Trap (MOT) is used to create a cold cloud of $^{85}\text{Rb}$ atoms. Rubidium atoms from a dispenser 3 cm away are trapped and cooled by three orthogonal pairs of counterpropagating laser beams in a quadrupole magnetic field. The cold atom cloud contains typically several $10^7$ atoms at a temperature well below 300 $\mu$K. Charged particles emitted by the dispenser are deflected by statically charged plates in front of the dispenser. The background pressure is below $3 \times 10^{-8}$ mbar. These conditions ensure that the atoms in the MOT do not move on the relevant timescales of the experiment.

Rydberg atoms are created by 8 ns laser pulses at 594 nm in a two-photon process from the cold ground-state Rb atoms. There are two separate sets of Nd:YAG-pumped dye lasers, one which excites atoms to the 49s state and one which excites to the 41d state. The laser creating the 49s atoms has a linewidth of 0.017(11) cm$^{-1}$ and a pulse energy of 3.0(1) $\mu$J; the laser creating 41d atoms has a linewidth of 0.21(2) cm$^{-1}$ and a pulse energy of 5.0(1) $\mu$J. The repetition rate of both lasers is 10 Hz. Each can be focused into a different region of the MOT to create two spatially separated, cigar-shaped volumes of Rydberg atoms within the magneto-optical trap. The focus of the laser creating the 49s atoms can be moved laterally within the trap by use of a stepper motor. The beam separation can accurately be measured in a two-photon mixing process using one photon from each beam. For this measurement we detune the laser creating the 49s atoms by 20 GHz to the blue and make sure that the laser pulses overlap in time. This leads to the excitation of the 44d state as described in chapter 5. As reported there the diameter of the Rydberg volumes is 11.6(0.4) $\mu$m for 49s- and 16.3(0.5) $\mu$m for 41d-atoms.

**Figure 6.4:** Schematic of the timing of one experimental cycle. First the magnetic field is switched off, a laser pulse excites Rydberg states which then are field ionized after 20 $\mu$s interaction time. Finally the magnetic field is switched on again.

The MOT is situated between two circular stainless steel plates with a diameter of
6.3 Improved Experimental Setup

5.5 cm which are spaced 2.50(5) cm apart. Both plates are perforated by a 14 mm hole in the center to allow ionization products to pass through, as well as to enable optical access to the MOT. These plates serve to generate the static- and radio-frequency fields used to manipulate the dipole-dipole interaction as well as for the field ionization.

The manipulating fields are generated by an Agilent 33250A Arbitrary Waveform Generator attached to one of the plates. The amplitude of these external fields is in the range of 0–1 V/cm and the frequency of the external RF-field is in the range of 1–30 MHz. The polarization of the RF-field is always parallel to the separation between the Rydberg volumes as they are separated along the symmetry axis of the plates.

The other plate is connected to a home-built fast high-voltage pulse generator to provide a negative field ramp that ionizes the Rydberg atoms after the interaction time. The ramp rises from 0 to 150 V/cm in 5 μs. Electrons are detected on a Hamamatsu Multi-channel Plate (MCP). Since each state ionizes at a different electric field the signal arises at a different time for different states [35]. This allows to distinguish between 49s, 49p and 41d atoms. Electrons from 41d atoms arrive at the same time as electrons from 42p, so this method does not allow a distinction between these two states. We use the final state fraction \( N_{49p}/(N_{49p} + N_{49s}) \) as a measure of the dipole-dipole interaction.

It is important that both power supplies connected to the plates have a low noise level during the interaction time. This was realized by a metal shielding box around the home built pulse generator and an implemented line filter. The Agilent 33250A had a low noise level already. The total level of electric field noise is below 2 mV/cm. We did not observe any influence of the RF-field on the SFI process as the amplitude of the RF-field is negligible compared with the magnitude of the SFI pulse, and as the frequency of the RF-field is too low to induce transitions to neighboring Rydberg states.

We have greatly improved our setup by adding the ability to switch the coils generating the magnetic quadrupole field for the MOT off during the dipole-dipole interaction time. Previously the magnetic field varied by \( \approx 0.5 \) G over the length of the Rydberg volumes. This resulted in a broadening of the resonances by \( \approx 1.5 \) MHz, due to Zeeman shifts of the involved states. For all results reported here the coils were switched off for 6 ms in each 100 ms cycle using a fast home-built IGBT switch. The current in the coils decays with a decay time of 25 μs. Measurements are performed 2.2 ms after the coils were switched. The magnetic field has been measured to be below 0.14 G at this time. The remaining field is assumed to be due to background stray fields. The timing sequence of the experiment is shown in Fig. 6.4.

Fig. 6.5 shows the 49p-fraction \( N_{49p}/(N_{49p} + N_{49s}) \) as a function of static electric field for various separations of the dye laser beams creating the Rydberg atoms. The widths of the resonances have been improved significantly compared to the results presented in chapter 5 and are given in table 6.1. Resonance widths are extracted from the data by fitting two independent Lorentz profiles to the measurements. The improvement is clearly due to the switching of the magnetic field. Our results are now in good agreement with the results of simulations (chapter 5) which are also given in table 6.1. The remaining difference might be due to remaining variations in the electric field, or due to ions in the MOT-cloud. Note that a width of 5 mV/cm corresponds to only 661 kHz for resonance \( F_a \) and 612 kHz for resonance \( F_b \). Both the simulations as well as the measurements yield widths bigger than the theoretical predictions of 200 kHz. This discrepancy can be
Radio-frequency driven dipole-dipole interactions in spatially separated volumes

Figure 6.5: Static field scans at 20 (blue circles) 30 (red squares) and 40 (green triangles) µm separation between the 41d and the 49s volumes. Solid lines are fits with two independent Lorentz profiles. Dashed lines are theoretical computations (chapter 5). The inset shows the energy level diagram for two atoms at 25 µm. The energy scale is relative to the energy of the 41d + 49s state at zero field.

Table 6.1: Width (FWHM) of the resonances $F_a$ and $F_b$ for varying beam separations. Experimental and theoretical values are given. Theoretical data is taken from chapter 5.

Diabatic Switching of the Dipole-Dipole Interaction

In a first experiment we investigated the switching of the dipole-dipole interaction by repeatedly changing the electric field diabatically from the resonance $F_a$ to a value $F_{off} = 0$ V/cm. The results are shown in Fig. 6.6. This figure depicts the 49p-fraction as a function of interaction time. The main curve in Fig. 6.6 (red solid lines, triangles) shows the 49p-fraction for an electric field that is switched several times from 0 V/cm to $F_a$ and
6.5 Observation of RF-assisted dipole-dipole transitions

Fig. 6.6: 49p-fraction as a function of interaction time for different fields $F = F_a$ (green circles), $F = 0$ (blue squares) and $F$ alternating between between $F_a$ and 0 in the manner depicted on top of the figure (red triangles). Lines depict a smooth spline fitted to the data. The switching is done at a very fast rate of 76 V/cm/µs. This is much faster than the frequency of the quantum beat oscillations times the width of the resonance ($2 \times 10^{-4}$ V/cm/µs) ensuring that the system follows the field diabatically. Clearly the 49p-fraction rises during the times where $F = F_a$ and stagnates when $F = F_{\text{off}}$. This shows how the dipole-dipole interaction can be switched on and off by rapidly changing the electric field. Reference measurements are performed for $F = F_a$ at all times (green dashed curves, circles) and for $F = 0$ at all times (blue dashed-dotted curves, squares). In the first case the 49p-fraction rises in the manner already measured in chapter 5. When the field is off-resonant at all times there is a small background contribution due to black body radiation (see figure 6.2).

6.5 Observation of RF-assisted dipole-dipole transitions

Fig. 6.7 shows the 49p-fraction as a function of the frequency of an external sinusoidal RF-field with constant amplitude and offset, here $F_S = 260$ mV/cm and $F_{RF} = 80$ mV/cm. In this case amplitude and offset are chosen such that the field never reaches the resonance fields $F_a$ or $F_b$. The figure shows a series of double peaks; each sub-peak corresponds to one of the two resonances from the different $|m_j|$ substates, and each set corresponds to a different number of photons (N) needed to make the transition resonant, as noted in the figure. Up to 5-photon transitions are clearly visible in the figure. The frequency of each N-photon transition is $1/N$-th of the one-photon transition frequency. The position of the peaks agrees very well with theoretical calculations based on eqn. 6.7 indicated by vertical lines (dashed: $F_a$, dotted: $F_b$) calculated for an effective field of $F_{\text{eff}} = 266.1$ mV/cm.

Fig. 6.8 again shows the 49p-fraction (normalized to the highest measured signal) as a function of frequency of the RF-field. The lower panel shows a frequency scan for
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Figure 6.7: Observed frequency dependence of RF-assisted dipole transitions at $F_S = 0.26$ V/cm. Multi-photon peaks with $n=1–5$ involved photons are clearly distinguishable, the number of involved photons is denoted in the figure. The double peak structure corresponds to the two resonances $F_a$ and $F_b$. The solid line is a smooth spline fitted to the data.

$F_S = 0$ V/cm and $F_{RF} = 331$ mV/cm, hence no static field is present in this measurement. The upper panel uses a small static field of $F_S = 60$ mV/cm and $F_{RF} = 320$ mV/cm so both measurements have the same effective field $F_{eff} = 234$ mV/cm. All peaks with odd photon number are clearly suppressed in the lower panel, but easily distinguishable in the upper panel. This can be understood on the basis of selection rules: The dipole-dipole interaction Eq. 6.1 requires that one unit of angular momentum is transferred from one atom to the other, $49s \rightarrow 49p$ requires $\Delta \ell = +1$ and $41d \rightarrow 42p$ requires $\Delta \ell = -1$. To achieve this, an odd number of photons must be involved in the process. Since there is already one photon transmitting the dipole-dipole interaction the number of external RF-photons must be even if the static field is zero. Already at static fields of only 60 mV/cm this selection rule becomes invalid as a static-field photon can account for the missing angular momentum without transferring any energy.

In Fig. 6.8 the importance of the AC-Stark shift can clearly be observed. E.g. for resonance a, the energy difference between the sd- and the pp-state is 24.53 MHz at 60 mV/cm. Without AC-Stark shift the 1, 2 and 3-photon resonances are expected at 24.53, 12.26 and 8.17 MHz. Including the AC-Stark shift

$$\Delta \omega = \frac{1}{4} \alpha F_{RF}^2,$$

we get significantly different numbers, i.e. 15.64, 7.82 and 5.31 MHz. These resonant positions clearly correspond to the observed peaks. The theoretical predictions including this AC-Stark shift are depicted in the figure as dash-dotted lines (resonance a) and dotted lines (resonance b).

In Fig. 6.9 the 1-photon resonance is used to perform spectroscopy on the 2-atom states, measuring the energy difference between initial and final states for several values
6.5 Observation of RF-assisted dipole-dipole transitions

Figure 6.8: Measurements at small static field $F_S$ (upper panel) and at $F_S = 0$ (lower panel) showing selection rules prohibiting odd photon number transitions (see text). The solid lines are a smooth spline fitted to the data.

of static field offset. The figure shows the 49p-fraction as a function of both RF- and static field. The position of the peaks corresponds to the energy levels in Fig. 6.3 as depicted by the dashed lines. These dashed lines are the same as the difference between the energy levels depicted in Fig. 6.3, but now with the x- and y axis interchanged. In the upper panel the amplitude of the radio-frequency field is $F_{RF} = 80$ mV/cm. In the lower panel $F_{RF} = 160$ mV/cm was used since the signal dropped too much for the lower amplitude at these small static field values. This leads to an appreciable contribution of the AC-Stark shift in the lower panel. The dashed lines show the theoretical resonance position taking AC-Stark shift into account on basis of Eq. 6.7. The +1 and −1 photon resonance position as a function of static field allows us to determine the total difference polarizability and difference energy between initial and final state. For the states investigated in this chapter this yields $W_0 = 25.07(10)$ MHz, $\alpha_a = 347.2(2.1)$ MHz/(V/cm)$^2$ and $\alpha_b = 300.5(1.9)$ MHz/(V/cm)$^2$. Note that for these results the field strength was calibrated using static field scans such as shown in Fig. 6.5. For accurate spectroscopy independent measurements of the electric field strength would be needed.
Figure 6.9: +1 and −1 photon resonances at various effective field values. In the lower panel (small static fields) the RF-amplitude had to be increased leading to a larger AC-Stark shift. The fitted resonance positions are marked by black dots. Some 2-photon resonances are also visible and marked in the figure.

6.6 Conclusion

The results presented in this chapter have been improved significantly compared to chapter 5 by switching off the magnetic field coils during measurements. The widths of the resonance peaks are now in good agreement with the simulations presented in chapter 5. This improvement makes sub-MHz spectroscopy possible. We have shown that the dipole-dipole interaction can be manipulated by rapidly switching the electric field. Furthermore we have shown multi-photon resonances in RF-assisted dipole-dipole interaction with up to 5 involved photons where the interacting dipoles are localized in separate volumes. For large amplitudes of the RF-field significant AC-Stark shifts can be observed due to the quadratic field dependence of the involved states. The positions of the resonance peaks agree well with the theory presented here. The methods explored in this chapter allow to perform spectroscopy on Stark states of two Rydberg atoms and we have determined the energy difference and difference in polarizability with good precision.

In this chapter we have shown several versatile methods of manipulating the interactions between Rydberg atoms in separated volumes by external fields. These methods should prove to be valuable for the manipulation of dipole-dipole interaction between neutral atoms.
We have observed Stückelberg oscillations in the dipole-dipole interaction between Rydberg atoms with an externally applied radio-frequency field. The oscillating RF field brings the interaction between cold Rydberg atoms in two separated volumes in resonance. We observe multi-photon transitions when varying the amplitude of the RF-field and the static electric field offset. The angular momentum states we use show a quadratic Stark shift, which leads to an essentially different behavior than linearly shifting states. Both cases are studied theoretically using the Floquet approach and are compared. The amplitude of the sidebands, related to the interaction strength, is given by the Bessel function in the linearly shifting case and by the generalized Bessel function in the quadratically shifting case. The oscillatory behavior of both functions correspond to Stückelberg oscillations, an interference effect described by the semi-classical Landau-Zener-Stückelberg model. The measurements prove coherent dipole-dipole interaction during at least 0.6 $\mu$s.

This chapter is based on the paper: C. S. E. van Ditzhuijzen, Atreju Tauschinsky, H. B. van Linden van den Heuvell, Observation of Stückenberg oscillations in dipole-dipole interactions, submitted for publication (2009), preprint arXiv:0907.4275
7.1 Introduction

In a previous experiment we prepared two different types of Rydberg atoms in separate volumes and we observed energy transfer from one sample L to the other sample R (chapter 5). In other words bit L changed from 0 to 1, while bit R changed from 1 to 0. One of the motivations to study dipole-dipole transfer from spatially separated volumes is to investigate the feasibility of Rydberg atoms for quantum information applications. To that end the phase of the involved states and not just the population should be a relevant parameter. In this paper we investigate Stückenberg oscillations in dipole-dipole interactions. Such oscillations are a sign of the phase memory of the bits, hence of the quantum character of the information that is carried by the bits. The oscillations can be observed for quantum states that are subjected to a time-varying field and that are degenerate at some points during this time-dependence of the field.

First we will give a theoretical description of polar and polarizable systems in a RF field, so we compare the situation for states with a linear versus a quadratic Stark shift. We use the Floquet approach, which is applicable for periodically varying fields. In this framework a two-level system plus oscillating field is replaced by an infinite number of sidebands on the two states. For a quadratic Stark shift the result is essentially different and more complicated than for a linear Stark shift. Next we compare the Floquet description with the classical limit. In addition we interpret the oscillations of the population of the sidebands as Stückenberg oscillations.

In the subsequent section we describe the details of the experiment and present the results of the measurements. Related experiments have been performed with transitions within single Rydberg atoms, in most cases for states with a linear Stark shift [16, 58, 102], but also quadratic shifts have been studied [108]. Collisions of Rydberg atoms in an oscillating field have been studied in an atomic beam setup [79, 50], as well as in a cold cloud [17, 6]. In our experiment we control the interaction strength, by maintaining a fixed distance between the Rydberg-atom volumes. Because of the small unperturbed energy mismatch between initial and final states, we use a rather low frequency for the oscillating field; it lies in the radio frequency range rather than in the microwave range, giving the opportunity to check the coherence during a longer period of time.

7.2 A coupled two-level system in an oscillating field

Inspired by the forthcoming experiment, we recall the two-level system [35], coupled by an interaction $V$ in the presence of a field $F(t)$

$$F(t) = F_S + F_{RF} \cos \omega t,$$  

(7.1)

with an oscillating and a static part. We write the full Hamiltonian as

$$H = H_0 + H_S + H_{RF} + V,$$  

(7.2)

where $H_0$ is the Hamiltonian of the two-level system without interaction, $H_S$ the interaction with the static field and $H_{RF}$ the interaction with the oscillating field. We want to know the population in both states, so we need to know the full wavefunction $\psi(r, t)$,
by solving the Schrödinger equation. We solve the problem by starting from the known static solution from $H_0 + H_{F_S}$, as described below, and then we add the other two terms as perturbation. In the next sub-section 7.2.1 "Numerical solutions in the time domain" we work out the problem by first adding the interaction term $V$ to the Hamiltonian, and then the oscillating field $H_{RF}$. This provides us with two coupled equations which can be solved numerically in the time domain. In the subsequent sub-sections we work in the reverse order. In section 7.2.2 "Floquet approach" we add the oscillating field $H_{RF}$ first and work out the problem in the frequency domain, which basically provides us with an infinite number of steady states in the form of sidebands. The classical limit of the sideband population, section 7.2.3, gives some physical insight in its behavior. Then, in section 7.2.4 "Coupling between dressed and undressed state" we add the interaction term $V$ to the Hamiltonian. The oscillations in the coupling as a function of RF amplitude can be understood in terms of Stückelberg oscillations, explained in section 7.2.5.

We will start treating the perturbation of the Hamiltonian $H = H_0 + H_{F_S}$ in a static field. With just the first two terms of the Hamiltonian

$$H = H_0 + H_{F_S},$$

we get a solution

$$\psi(\mathbf{r}, t) = a \psi_1(\mathbf{r}, t) + b \psi_2(\mathbf{r}, t),$$

(7.3)

where $\psi_1$ and $\psi_2$ are the eigenstates of the system and $a$ and $b$ are their normalized time-independent amplitudes,

$$\psi_1(\mathbf{r}, t) = \psi_1(\mathbf{r}) e^{-iW_1 t},$$

(7.4a)

$$\psi_2(\mathbf{r}, t) = \psi_2(\mathbf{r}) e^{-iW_2 t},$$

(7.4b)

where $W_1$ and $W_2$ are the eigenenergies of the two stationary states.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure7.1.png}
\caption{Energy level diagrams of state $|1\rangle$ and $|2\rangle$ as a function of electric field. State $|1\rangle$ has no Stark shift, state $|2\rangle$ has a linear Stark shift in Fig. a) and a quadratic Stark shift in Fig. b). Due to the coupling between the states, the crossing becomes an avoided crossing, depicted in the inset. When an oscillating field is applied, depicted in purple, this translates as an oscillating energy for state $|2\rangle$, depicted on the right hand side in red. The orange dotted lines depict the average energy during the oscillation and the corresponding field; in a) this field is simply $F_S$, the average field, but in b) it is different and we define it as the effective field $F_{eff}$, given in Eq. 7.25.}
\end{figure}

We will discuss two cases, one for states with a linear Stark shift and one for states with a quadratic Stark shift. The first case is depicted in Fig. 7.1a. State $|1\rangle$ always has
energy 0 and state |2⟩ has energy $W_0$ without field and a linear energy shift as a function of electric field $F$ (all equations in this chapter are in atomic units):

\begin{align}
W_1 &= 0, \\
W_2 &= W_0 - kF. 
\end{align}

Here $k$ is the permanent dipole moment of state |2⟩. This system is in fact equivalent to two states with difference energy $W_0$ and a difference in dipole moment of $k$, which makes this two-level system applicable to many realistic systems. The two levels cross at the field

$$F_{\text{lin}} = \frac{W_0}{k}. \quad (7.6)$$

The second case is sketched in Fig. 7.1b. Again, state |1⟩ has energy 0 and state |2⟩ has energy $W_0$ at zero field but it has an energy shift that decreases quadratically as a function of electric field $F$:

\begin{align}
W_1 &= 0, \\
W_2 &= W_0 - \frac{1}{2} \alpha F^2. 
\end{align}

Here $\alpha$ is the polarizability of state |2⟩. Similar to the linear case this system is equivalent to two states with difference energy $W_0$ and a difference in polarizability of $\alpha$. The two levels cross at

$$F_{\text{quad}} = \sqrt{\frac{2W_0}{\alpha}}. \quad (7.8)$$

### 7.2.1 Numerical solutions in the time domain

The most direct method to calculate the evolution of the wavefunction in an oscillating field is to numerically solve the two coupled differential equations for both states. For this we first add the coupling $V$ and then the time dependent field perturbation $H_{F\delta F}$. When we add the interaction $V$, the degeneracy at the crossing is lifted and an avoided crossing arises, which has a width $\Omega_0$. This $\Omega_0$ can also be interpreted as a quantum beat oscillation frequency: the population of state $\psi_1$ and $\psi_2$ oscillate against each other with this frequency after an appropriate initial condition. The avoided crossing is depicted in the inset of Fig. 7.1. The width is given by

$$\Omega_0 = 2\langle \psi_2(r)|V|\psi_1(r)\rangle. \quad (7.9)$$

When we add a time-dependent field it is convenient to separate the time and spatial dependence of the wavefunction (Eq. 7.3)

$$\psi(r, t) = T_1(t) \psi_1(r) + T_2(t) \psi_2(r). \quad (7.10)$$

$\psi_1(r)$ and $\psi_2(r)$ are the two original states without interaction. This separation of time- and spatial dependence is only allowed if both spatial states remain approximately the
same despite the applied fields. We will see in section 7.3.1 that this is well validated. Using this wavefunction in the time-dependent Schrödinger equation we obtain

\[ i \dot{T}_1(t) = W_1(t) + \frac{\Omega_0}{2} T_2(t), \quad (7.11a) \]
\[ i \dot{T}_2(t) = W_2(t) + \frac{\Omega_0}{2} T_1(t). \quad (7.11b) \]

Now the energies \( W_1 \) and \( W_2 \) depend on time. For the linear case we have

\[ W_2(t) = W_0 - k (F_S + F_{RF} \cos \omega t), \quad (7.12) \]

and for the quadratic case

\[ W_2(t) = W_0 - \frac{1}{2} \alpha \left( F_S^2 + 2 F_S F_{RF} \cos \omega t + F_{RF}^2 \cos^2 \omega t \right) \quad (7.13) \]

and in both cases \( W_1(t) = 0 \).

We have numerically solved the two coupled equations in Eq. 7.11. However, the results are less informative than the approach described in the next section, the Floquet approach. First of all, because numerical calculations are in general less intuitive than analytic solutions. Furthermore, although equivalent, an answer in terms of a coherent superposition of stationary states is more informative than the time-dependence of a wavefunction. However, this numerical approach can be used for all time-dependent fields, whereas the Floquet approach only works for periodic functions.

### 7.2.2 Floquet approach

In the Floquet approach we add the perturbations in the opposite order as in the numerical calculation; so we first perturb the states \( \psi_1 \) and \( \psi_2 \) with the oscillating field and subsequently add the interaction \( V \) between the two states [35, 108]. As in the previous section we separate the time and spatial dependence, but this time we keep two separate wavefunctions

\[ \psi_1(\mathbf{r}, t) = T_1(t) \psi_1(\mathbf{r}), \quad (7.14a) \]
\[ \psi_2(\mathbf{r}, t) = T_2(t) \psi_2(\mathbf{r}). \quad (7.14b) \]

Using these in the time-dependent Schrödinger equation we obtain

\[ i \dot{T}_1(t) \psi_1(\mathbf{r}) = W_1(t) T_1(t) \psi_1(\mathbf{r}), \quad (7.15a) \]
\[ i \dot{T}_2(t) \psi_2(\mathbf{r}) = W_2(t) T_2(t) \psi_2(\mathbf{r}). \quad (7.15b) \]

We can factor out the spatial dependent part and integrate Eqs. 7.15

\[ T_1(t) = 1, \quad (7.16a) \]
\[ T_2(t) = e^{-i \int W_2(t) dt}, \quad (7.16b) \]
Figure 7.2: The sidebands of state $|2\rangle$ for the linear case in an oscillating field of 8 MHz for various RF amplitudes. The darkness of a line represents the population of the regarding sideband, where the original state, or the $n=0$ sideband, is colored blue. The red lines depict the borders of the classically allowed region. We used $k = 2\pi \times 60$ MHz/(V/cm) and $W_0 = 2\pi \times 25$ MHz.

This expression shows that the wavefunction consists of the original spatial wavefunction, but a modified time dependence in the form of an infinite number of sidebands. This is the steady-state solution of the problem, consisting of a number of states with different amplitudes, given by the Bessel function $J_n\left(\frac{kF_{RF}}{\omega}\right)$, all with a different energy $W_{2,n} = W_0 - kF_S - n\omega$. 

In Fig. 7.2 the sidebands of $\psi_2$ are plotted for various RF amplitudes. The original energy of $\psi_2$, or in other words the $n=0$ sideband, is depicted in blue. The color strength depicts the population of the sideband $J_n(kF_{RF}/\omega)^2$. It is clearly visible that more sidebands are populated when a larger RF amplitude is applied. The red lines indicate the
Figure 7.3: The sidebands of state \( |2\rangle \) for the quadratic case in an oscillating field of 8 MHz for different RF amplitudes. The population of the sideband is depicted as the darkness of the line. The \( n = 0 \) sideband is depicted in blue. The red lines depict the position of the asymptotes of the distribution of occurring energies. We used the values corresponding to the experimental values of the left resonance, as mentioned at Eq. 7.54.

classical limits, the maximum and minimum field values that occur in the oscillation. This is equivalent with the order and absolute argument of the Bessel function being equal.

The case of the quadratic Stark shift is quite different and more complicated. Combining Eq. 7.16b and 7.13 we get

\[
T_2(t) = e^{-i(W_0 - \frac{1}{2}a(F_S^2 + \frac{1}{2}F_R^2))t} e^{i a F_S F_R \sin \omega t} e^{i a R \sin 2\omega t}. \tag{7.20}
\]

Here we have two exponents to expand in terms of Bessel functions

\[
e^{ix \sin \omega t} e^{iy \sin 2\omega t} = \sum_{m' = -\infty}^{\infty} J_{m'}(x) e^{i m' \omega t} \sum_{m = -\infty}^{\infty} J_m(y) e^{i 2m \omega t},
\]

\[
= \sum_{m = -\infty}^{\infty} e^{im \omega t} \left( \sum_{m = -\infty}^{\infty} J_{n - 2m}(x) J_m(y) \right), \tag{7.21}
\]

where \( n = m' + 2m \) is used on the second line. The amplitude of the sideband, so the inner summation in the above expression, is known as the generalized Bessel function \( J_n(x, y) \). It was first used by Reiss [78] and investigated by e.g. Dattoli et al. [24]. It is defined here as

\[
J_n(x, y) = \sum_{m = -\infty}^{\infty} J_{n - 2m}(x) J_m(y). \tag{7.22}
\]
In practice the summation can be taken up to \( |m| = y + 3y^{1/3} + 3 \) or \( |m| = \frac{1}{2}(n + x + 3x^{1/3} + 3) \), because beyond these points the Bessel function is negligible. Instead of evaluating the sum, it is also possible to calculate the generalized Bessel function on basis of the recurrence relation \([63]\) in analogy with the well-known approach to calculate the regular Bessel function on basis of its recurrence relation.

The wavefunction becomes

\[
\psi_2(r, t) = \psi_2(r) e^{i(\omega t - \frac{1}{2} \alpha (F_R^2 + \frac{1}{2} F_S^2))} \sum_{n=-\infty}^{\infty} e^{i\omega n} J_n \left( \frac{\alpha F_R F_S}{\omega}, \frac{\alpha F_R^2}{8\omega} \right). \tag{7.23}
\]

A similar description of sidebands on quadratically shifted states is given in ref. \([108]\).

If we compare this wavefunction with the linear case it is the generalized Bessel function instead of the regular Bessel function that determines the sideband amplitude. The result depends on both the static field \( F_S \) as well as the RF amplitude \( F_{RF} \), whereas for the linear case it depends on the RF amplitude only. The energy of the sidebands is also essentially different

\[
W_{2,n} = W_0 - \frac{1}{2} \alpha \left( F_S^2 + \frac{1}{2} F_{RF}^2 \right) - n\omega. \tag{7.24}
\]

Where in the linear case the energy of a particular sideband shifts with the static field \( F_S \) only, in the quadratic case there is an additional term that depends on the RF amplitude \( \frac{1}{4} \alpha F_{RF}^2 \). This term corresponds to the AC-Stark shift of the state. It is convenient to define an effective field \( F_{eff} \), such that the energy shift of such a static field corresponds to the energy shift of a static plus an RF field:

\[
F_{eff}^2 = F_S^2 + \frac{1}{2} F_{RF}^2. \tag{7.25}
\]

The different behavior is visible in Fig. 7.3, where the sidebands are plotted versus static field for several RF amplitudes. Again, the color strength depicts the population of the sideband: \( J_n \left( \alpha F_{RF} F_S / \omega, \alpha F_{RF}^2 / (8\omega) \right)^2 \). As in the linear case more sidebands are populated with a larger RF field, but a clear difference is that the population of a sideband is now not only a function of the RF amplitude, but also of the DC field, visible as the changing color strength within one sideband. Secondly, the different positions of the sidebands in each subfigure illustrates that the sideband energy also depends on \( F_{RF} \) (note that the original state, or the \( n = 0 \) sideband, in blue, is shifted downwards for larger \( F_{RF} \)).

The next step would be to add the coupling between state \(|1\rangle\) and state \(|2\rangle\) (section 7.2.4), but first we will look closer into the population of the sidebands for different frequencies. This provides us with insight in the range of populated sidebands and other details on the behavior of the (generalized) Bessel function.
7.2 A coupled two-level system in an oscillating field

Figure 7.4: Depicted in black is the sideband population of state $|2\rangle$ as a function of the sideband energy (Eqs. 7.19 and 7.24) for different frequencies. $\omega$ is $2\pi$ times the mentioned frequency. We used again $k = 2\pi \times 60$ MHz/(V/cm) and $W_0 = 2\pi \times 25$ MHz and field values of $F_S = 0.2$ V/cm and $F_{RF} = 0.3$ V/cm. In red is depicted the distribution of occurring energies, or in other words the classical limit, calculated with the method of Eq. 7.26.

7.2.3 Classical limit of the sideband population distribution

To illustrate the sideband population and to obtain some physical insight in its behavior, we depicted the sideband population for a fixed field amplitude and offset as a function of the sideband energy (Eqs. 7.19 and 7.24) and various frequencies in figures 7.4 and 7.5. In other words, we plot the squared (generalized) Bessel function $J_n(x)^2$ respectively $J_n(x, y)^2$ for fixed $x$ and $y$ (per subfigure) as a function of $n$. The pictures can also be seen as a vertical cut through a subfigure of Fig. 7.2 or 7.3 with the appropriate parameters $F_S$, $F_{RF}$ and $\omega$. For large frequencies, i.e. small $x$ and $y$, the function peaks at $n = 0$ and drops off on both sides, so there are not many populated sidebands. For smaller frequencies (or large $x, y$), more and more sidebands arise. Around $n = 0$ the population oscillates and near $|n| = |x|$ the Bessel function reaches a maximum, after which it drops exponentially to zero. Interestingly, the sideband populations extend further in energy beyond $|n| > |x|$ for the higher frequencies as for the lower frequencies. For the number of populated sidebands beyond $|n| = |x|$ the opposite is true. These extensions can be understood as a quantum mechanical effect, whereas the energy levels are in a classically forbidden region. The generalized Bessel function also has these features. In addition, it has a more asymmetric structure, which we will discuss further below.

For low frequencies the (generalized) Bessel function approaches the classical limit, meaning it approaches the density distribution of the occurring energies. To calculate such a density distribution it is useful to know how a distribution, $f(x)$, changes under a

---

1The Bessel function $J_n(x)$ is roughly proportional to $Ai(2^{1/3} \frac{n^2 x}{n^2})$ for $n \geq x$. The Airy function $Ai(z)$ drops of as $\exp(-2^{1/3}z^{2/3})$ for large $z$. Combining this, we obtain that the larger-order Bessel function is smaller than some error $\epsilon$, with $\epsilon = \exp(-Q)$ if $n > x + Q^{2/3} x^{1/3} + \frac{1}{2}Q$. This drop-off limits the required summation range.
change of variable, from $x$ to $y$. If $y = g(x)$ the resulting density function becomes

$$P(y) = \left| \frac{1}{g'(g^{-1}(y))} \right| f(g^{-1}(y)),$$

(7.26)

with $g'$ the derivative and $g^{-1}$ the inverse function. For non-monotonic functions $g(x)$ we have

$$P(y) = \sum_{k}^{n} \left| \frac{1}{g'(g^{-1}_{k}(y))} \right| f(g^{-1}_{k}(y)),$$

(7.27)

where $n$ is the number of solutions in $x$ for $g(x) = y$ and $g^{-1}_{k}(y)$ are these solutions.

**Figure 7.6:** The same type of plot as in Fig. 7.5, but now at an even lower frequency and zoomed in around the middle asymptote. The blue curve depicts the moving average of the sideband population depicted in black.

We could choose $g$ to be the energy as a function of time, given by Eq. 7.12 and 7.13. It is, however, instructive and easier to first calculate the distribution of the occurring fields. So we use $g(t) = F(t) = F_S + F_{RF} \cos \omega t$. We start with $f$ as a uniform normalized distribution. It is sufficient to have instances during half a cycle of the cosine, so the
distribution becomes
\[ f(t) = \begin{cases} \frac{\omega}{\pi} & \text{if } 0 \leq \omega t \leq \pi \\ 0 & \text{elsewhere} \end{cases} \] (7.28)

Applying these functions \( f \) and \( g \) to eqn. 7.26 we obtain for the probability density distribution of the field
\[ P(F) = \begin{cases} \frac{1}{\pi \sqrt{F^2 - (F - FR)^2}} & \text{if } F_S - FR \leq F \leq F_S + FR \\ 0 & \text{elsewhere.} \end{cases} \] (7.29)

This function diverges at \( F = F_S \pm FR \). These asymptotes correspond to the turning points of the field. Here the function \( F(t) \) is flat; the derivative is zero.

The function \( F(t) \) is sketched in the energy level diagrams, Fig. 7.1, as a purple line. How the field oscillation translates to an oscillation in energy (Eq. 7.12 and 7.13) is depicted on the right hand side as the red line. In the linear case, the function simply remains a cosine, but in the quadratic case the function is distorted. The latter one has a third turning point when the field goes through zero, i.e. when \( FR > F_S \). The probability density distribution of the energy is for the linear case in essence the same as the distribution of the field, but for the quadratic case it is different. For \( FR < F_S \) it looks similar to the linear case, but it is somewhat asymmetric, for \( FR > F_S \) the density distribution consists of two parts that are added up (Eq. 7.27 is needed). Both are again an asymmetric version of the linear case; one runs from \( W_0 - \frac{1}{2}a(F_S + FR)^2 \) to \( W_0 \), and the other from \( W_0 - \frac{1}{2}a(F_S - FR)^2 \) to \( W_0 \), so this distribution has three asymptotes. Both functions are plotted together with the strength of the sidebands in Figs. 7.4 and 7.5. Here they are multiplied by the frequency to match the average height of the sidebands. The asymptotes of the distribution functions are also depicted in Figs. 7.2 and 7.3 as red lines.

We observe in Figs. 7.4 and 7.5 that for the lower frequencies the sideband population indeed converges nicely to the distribution functions. The height of the peaks oscillate around the red line. In most cases values occur between 0 and twice the red line. In the case of the generalized Bessel function, on the right hand part of the plot some much higher peaks occur. However, the moving average still approaches the red line. This is depicted in Fig. 7.6, where we zoomed in around the asymptote \( W_0 - \frac{1}{2}a(F_S - FR)^2 \). The moving average of the sideband population is depicted in blue.

### 7.2.4 Coupling between dressed and undressed state

Now that we know the wavefunctions in an oscillating field, we can add the coupling \( V \) between \( \psi_1 \) and \( \psi_2 \). Every sideband of \( \psi_2 \) crosses with \( \psi_1 \). After introducing the coupling \( V \), every crossing becomes an avoided crossing. When we use the wavefunctions given by Eq. 7.14 we get
\[
\langle \psi_1(\mathbf{r}, t) | V | \psi_2(\mathbf{r}, t) \rangle = \langle \psi_1(\mathbf{r}) | V | \psi_2(\mathbf{r}) \rangle \ T_1^* T_2. \tag{7.30}
\]

The \( n \)th sideband of the linear case (see Eq. 7.18) becomes
\[
\langle \psi_1(\mathbf{r}, t) | V | \psi_2(\mathbf{r}, t) \rangle_n = \langle \psi_1(\mathbf{r}) | V | \psi_2(\mathbf{r}) \rangle \ e^{-i(W_0 - kF_S - \omega)\tau} J_n \left( \frac{kFR}{\omega} \right). \tag{7.31}
\]
Figure 7.7: Calculated interaction strength as a function of static and oscillating field for the linear case. The vertical lines depict the $n \omega$ resonances, where $n$ varies from $-6$ (the one most right) to $6$ (the one most left) and $\omega$ is fixed to $2\pi \times 4$ MHz. The darker the color the stronger the interaction. Destructive interference between the two states is depicted by the curved lines in red and green, according to St"uckelberg theory described in subsection 7.2.5. The blue lines give the boundaries of the classically allowed region; in other words these lines depict field combinations where the crossing (Eq. 7.6) occurs at an extremum of the RF oscillation.

The resonance condition is seen in the exponent

$$n \omega = W_0 - kF_S.$$  

(7.32)

The coupling strength at this resonance (the half width of the $n^{\text{th}}$ avoided crossing) is given by the time independent part of Eq. 7.31

$$\frac{\Omega_n}{2} = \langle \psi_1(r) | V | \psi_2(r) \rangle \frac{kF_{RF}}{\omega} J_n \left( \frac{kF_{RF}}{\omega} \right).$$

(7.33)

Here we used Eq. 7.9, which gives the coupling strength $\Omega_0$ without oscillating field. So the total coupling $\frac{1}{2} \Omega_0$ is distributed over all avoided crossings, according to the Bessel function. In a similar manner, we get the resonance condition for the quadratic case

$$n \omega = W_0 - \frac{1}{2} \alpha \left( F_S^2 + \frac{1}{2} F_{RF}^2 \right).$$

(7.34)
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Figure 7.8: Calculated interaction strength as a function of static and oscillating field for the quadratic case. The quarter circles in varying gray depict the $n\omega$ resonances. Here $n$ runs from $-25$ (in the upper right corner) to 3 (in the lower left corner); the frequency of the RF field is here $\omega = 2\pi \times 8$ MHz. The darker the color the stronger the interaction. Destructive St"uckelberg interference between the two states is depicted in red, green and orange. The blue lines give all field combinations where the crossing is reached during an extremum of the RF oscillation.

and the coupling strength is

$$\frac{\Omega_n}{2} = \frac{\Omega_0}{2} J_n \left( \frac{\alpha F_{RF} F_S}{\omega}, \frac{\alpha F^2_{RF}}{8\omega} \right). \quad (7.35)$$

In Fig. 7.7 we plot the coupling resonances as a function of RF amplitude, $F_{RF}$, and static field, $F_S$, for a fixed frequency $\omega$ for the linear case. A grayscale depicts the squared coupling strength or $J_n (kF_{RF}/\omega)^2$. The resonances are just vertical lines, since the resonance depends on the static field only. The blue lines indicate the boundaries of the classically allowed region, which correspond to the regions where the field $F(t)$ (Eq. 7.1) reaches the static field resonance $F_{lin}$ (Eq. 7.6). These classical boundaries correspond to the boundaries depicted as red lines in Fig. 7.2 and to the asymptotes depicted in red in Fig. 7.4. Above these lines the coupling strength oscillates as a function of $F_{RF}$, which we can interpret as St"uckelberg oscillations, described in the next section.

Fig. 7.8 shows the corresponding plot for the quadratic case. Here the resonance lines are curved, in fact, each line follows a line of constant effective field, defined in Eq. 7.25. The boundaries of the classically allowed region are again indicated with the two lower
blue lines. They correspond to the lines $F_S + F_{RF} = F_{\text{quad}}$ and $F_S - F_{RF} = F_{\text{quad}}$ (with $F_{\text{quad}}$ defined in Eq. 7.8). There is a third blue line, higher up in the figure, which indicates the boundary $F_S - F_{RF} = -F_{\text{quad}}$ above this line both the crossing at $-F_{\text{quad}}$ as well as the crossing at $+F_{\text{quad}}$ are involved. Note that the third boundary or asymptote from Figs. 7.3 and 7.5, the one with value $W_0$, is not relevant, because it is never involved in the crossing with state $|1\rangle$. Within the classically allowed region, i.e. above the lower two blue lines, we see the St¨uckelberg oscillations.

At this point there is enough information to start the measurements; it is known where to expect resonances and how strong they are. However, we will first explain why the oscillations in the interaction strength can be interpreted as St¨uckelberg oscillations.

### 7.2.5 St¨uckelberg oscillations

St¨uckelberg oscillations [91] are due to an interference effect between two quantum states, which occur when an avoided crossing is traversed at least twice. Imagine we start with all population in the state $|1\rangle$ and subsequently we sweep the field through the resonance and back (Fig. 7.1). When sweeping through, part of the wavefunction will follow the state $|1\rangle$ diabatically and "ignores" the avoided crossing, while the other part will follow the eigen-energy adiabatically and ends up in the state $|2\rangle$. When coming back again to the crossing, both parts of the wavefunction have built up a different phase, due to their energy difference and due to a difference in passing the crossing. This phase difference leads to interference, which can be observed when varying for example the duration or the amplitude of the field sweep. In this particular case we have a sinusoidal oscillation of the field with a fixed frequency and we vary both $F_S$ and $F_{RF}$.

To calculate the interference patterns we use the Landau-Zener-St¨uckelberg (LZS) model [57, 107, 91], as described in e.g. [51] and apply it to the situations sketched in Fig. 7.1. It is assumed that the avoided crossing (Eq. 7.9) is small compared to the range of occurring energies during an RF oscillation (Eq. 7.12 or 7.13). In that case we can separate the phase evolution due to energy differences and phase jumps due to avoided crossings. The Hamiltonian of the two-level system in a diabatic description is

$$H = \begin{pmatrix} W_a(t) & V \\ V^* & W_b(t) \end{pmatrix},$$

(7.36)

where $W_a$ and $W_b$ are the unperturbed eigen-energies of respectively state $|a\rangle$ and $|b\rangle$. In order to better exploit the symmetry of the problem, we choose a definition of energies that is slightly different than before: $W_b = -W_a = \frac{1}{2}W_2$, with $W_2$ defined in Eq. 7.12 or 7.13. The energies $W_a$ and $W_b$ during an RF oscillation are illustrated in Figs. 7.9 and 7.11 for a specific choice of $F_S$ and $F_{RF}$.

In the LZS model we describe the evolution of the wavefunction with transfer matrices. The transition at a level crossing is described by the transfer matrix [51]

$$M = \begin{pmatrix} \sqrt{1-\epsilon} & \sqrt{\epsilon} e^{-i\phi} \\ -\sqrt{\epsilon} e^{i\phi} & \sqrt{1-\epsilon} \end{pmatrix}.$$  

(7.37)

The matrix $M$ is applicable for crossings where state $|a\rangle$ crosses $|b\rangle$ from the lower-energy side. In the opposite case the transpose of $M$, $M^T$ should be used. The phase angle $\phi$ is
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the so-called Stokes phase

$$\phi = \frac{\pi}{4} + \arg \Gamma(1 - i\delta) + \delta(\ln(\delta) - 1)$$  \hspace{1cm} (7.38)

with values between 0 and \(\pi/4\). And \(\epsilon\) is the Landau-Zener adiabatic transfer probability

$$\epsilon = 1 - e^{-2\pi\delta}. \hspace{1cm} (7.39)$$

Both quantities depend on the adiabaticity parameter \(\delta\) which in its turn depends on the coupling strength \(V\) and the rate of change of the diabatic energies

$$\delta = \frac{V^2}{|d(W_b - W_a)/dt|}. \hspace{1cm} (7.40)$$

In the diabatic limit \(\delta\) is small and subsequently \(\epsilon\) as well, while \(\phi\) approaches the value \(\pi/4\). In the adiabatic limit \(\epsilon\) approaches 1 and \(\phi\) approaches 0.

After the crossing, the fraction in state \(|b\rangle\) develops a different phase compared to the fraction in state \(|a\rangle\), before coming back to the crossing. Both phase evolutions are described by the matrix

$$G_{ij} = \begin{pmatrix} e^{-i\Theta_{aij}} & 0 \\ 0 & e^{-i\Theta_{bij}} \end{pmatrix} \hspace{1cm} (7.41)$$

with \(\Theta_{aij} = \int_{t_1}^{t_2} \sqrt{W_a(t)^2 + V^2} dt \approx \int_{t_1}^{t_2} W_b(t) dt\) and equivalent for \(\Theta_{bij}\). The approximation is valid for small \(V\). Because \(W_b = -W_a\) also \(\Theta_{aij} = -\Theta_{bij}\). We use the shorter notation \(\Theta_{ij}\), which equals \(\Theta_{bij}\). Note that \(\Theta_{ij}\) is a function of \(F_S\) and \(F_{RF}\). The matrices \(M\) and \(G\) which describe the phase development at and between the crossings are indicated in figures 7.9 and 7.11. The colored surfaces indicate the phase of state \(|b\rangle\) as it develops between the crossings.

For the case of states with a linear Stark shift the situation is depicted in figure 7.9 and, assuming that we start with all probability in \(|a\rangle\), the amplitude of both states after \(N\) oscillations is given by

$$(G_{23}MG_{12}M^T)^N \begin{pmatrix} 1 \\ 0 \end{pmatrix} = S^N \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \hspace{1cm} (7.42)$$

where \(S\) describes the time evolution of the wavefunction over one RF oscillation. The matrix \(S\) is unimodular (\(\det S = 1\)). Therefore we can write \(S^N\) in terms of Chebychev polynomials \(U_n(\xi)\) \(^{[37]}\)

$$S^N = S \ U_{N-1}(\xi) - 1 \ U_{N-2}(\xi), \hspace{1cm} (7.43)$$

where

$$\xi = \text{Tr}(S). \hspace{1cm} (7.44)$$

The solution of the full wavefunction in the linearly Stark-shifting case after \(N\) oscillations can be expressed in terms of above defined quantities. In view of the size of the expression, we just give the population of state \(|b\rangle\)

$$P_b = 4(1-\epsilon)\epsilon^2 \sin^2(\Theta_{12} + \phi) U_{N-1}^2(\xi) \hspace{1cm} (7.45)$$
Figure 7.9: The energies of states $|a\rangle$ and $|b\rangle$ as a function of time during the RF-oscillation for the case of linear shifting Stark states. Indicated are the times where the two states cross, $t_1$, $t_2$ and $t_3$, and the matrices that describe the evolution of the wavefunction at the indicated time intervals: $M^T$, $G_{12}$, $M$ and $G_{23}$. The green and red colored areas indicate the phase evolution of the state $|b\rangle$, $\Theta_{ij}$, which is contained in the $G_{ij}$ matrix.

with the trace

$$\xi = \cos(\Theta_{12} + \phi) \cos(\Theta_{23} - \phi) - (1 - 2\epsilon) \sin(\Theta_{12} + \phi) \sin(\Theta_{23} - \phi).$$  (7.46)

In the near diabatic limit ($\epsilon \ll 1$) the population $P_b$ reduces to

$$P_b = 4\epsilon \sin^2(\Theta_{12} + \phi) \frac{\sin^2(N(\Theta_{12} + \Theta_{23}))}{\sin^2(\Theta_{12} + \Theta_{23})}. $$  (7.47)

In figure 7.10 we depicted the population of state $|b\rangle$ as a function of the static field $F_S$ and the RF amplitude $F_{RF}$ for $N=3$. Note the similarity with figure 7.7. For larger $N$ the vertically elongated areas of large population become narrower.

The quadratic case allows for a similar approach, but there is a twist. In cases where the states cross each other at only one field, either $+F_{\text{quad}}$ or $-F_{\text{quad}}$ (Eq. 7.8), the same equations are used as for the linear case. However, for large RF amplitudes it is possible that the two states cross at two different fields: $F = \pm F_{\text{quad}}$. The speed at which each crossing is crossed is usually different, so we have different values for the adiabaticity parameter $\delta$ which will be labelled $+\delta$ and $-\delta$. The same holds for parameters that depend on $\delta$: $\phi$, $\epsilon$ and $M$. The situation is illustrated in Fig. 7.11. After $N$ oscillations the wavefunction has evolved to

$$(G_{45} M_{12}^T G_{23} M_{12}^T)^N \begin{pmatrix} 1 \\ 0 \end{pmatrix}. $$  (7.48)
Figure 7.10: The population of state $|b\rangle$ as a function of the static field $F_S$ and the RF amplitude $F_{RF}$ (Eq. 7.47) for $N=3$, where red is a large population and blue is a population of 0.

The parameter of the Chebychev polynomial now becomes

$$\xi = (1 - \epsilon_+) \cos(2\Theta_{12} + \Theta_{23} + \Theta_{45}) + \epsilon_- \cos(2\phi_- + 2\Theta_{12} - \Theta_{23} + \Theta_{45})$$
$$+ 2 \sin(\Theta_{45} - \phi_-) \left[ \epsilon_+ \sin(2\Theta_{12} + \Theta_{23} + \phi_-) - 2 \sin(\Theta_{23} - \phi_-) \right]$$
$$\times \left( \epsilon_+ \epsilon_- \cos(2\Theta_{12} + \phi_- + \phi_+) + \sqrt{(1-\epsilon_+)\epsilon_-(1-\epsilon_-)\epsilon_+} \right),$$

(7.49)

where we have used $\Theta_{12} = \Theta_{34}$. After $N$ oscillations the $|b\rangle$ population is

$$P_b = 4 \left[ (1-2\epsilon_+) \sqrt{(1-\epsilon_+)} \epsilon_- \sin(\Theta_{23} - \phi_-) - \sqrt{(1-\epsilon_-)} \epsilon_+ \sin(2\Theta_{12} + \Theta_{23} + \phi_-) \right]^2 U^2_{N-1} (\xi).$$

(7.50)

For near-diabatic transitions $\epsilon_+, \epsilon_- \ll 1$ the transferred population reduces to

$$P_b = 4 \frac{\sin^2(N(2\Theta_{12} + \Theta_{23} + \Theta_{45}))}{\sin^2(2\Theta_{12} + \Theta_{23} + \Theta_{45})} \left[ \sqrt{\epsilon_+} \sin(2\Theta_{12} + \Theta_{23} + \phi_+) - \sqrt{\epsilon_-} \sin(\Theta_{23} - \phi_-) \right]^2.$$ 

(7.51)
Figure 7.11: A similar figure as figure 7.9, now for the case of quadratically shifting Stark states and large RF amplitudes, such that both the $+F_{\text{quad}}$ and the $-F_{\text{quad}}$ crossing occur. At $t_1$ and $t_4$ the field is at the $+F_{\text{quad}}$ crossing and at $t_2$ and $t_3$ the field is at the $-F_{\text{quad}}$ crossing. Furthermore, it is clearly visible that $\Theta_{12}$ and $\Theta_{34}$ are equal, they cover the section between $+F_{\text{quad}}$ and $-F_{\text{quad}}$.

This $|b\rangle$ population is plotted as a function of the static field $F_S$ and the RF amplitude $F_{RF}$ for $N = 3$ in figure 7.12. This figure shows a similar structure as figure 7.8.

In the expressions for the probability of state $|b\rangle$ (Eqs. 7.47 and 7.51) we recognize two factors. Firstly, the familiar $\sin^2(Nx)/\sin^2(x)$ factor which gives rise to the resonance condition $x = n\pi$; a very ubiquitous phenomenon. Working out this resonance condition we get exactly the same result as before: equations 7.32 and 7.34. Secondly, we identify what we can call the Stückelberg interference factor:

$$I_1 = 4\epsilon \sin^2(\Theta_{12} + \phi)$$

for the linear case or the quadratic case where only one crossing occurs and

$$I_2 = 4\left[\sqrt{\epsilon} \sin(2\Theta_{12} + \Theta_{23} + \phi) - \sqrt{\epsilon} \sin(\Theta_{23} - \phi)\right]^2$$

for two-crossings case. Note that these interference factors are independent of $N$ and they occur already for one single oscillation.

In figure 7.7 we have depicted the regions of destructive interference. The green lines are the destructive interference regions for the situation described above. The red lines give the destructive interference regions for the case of a different starting point of the RF oscillation: between $t_1$ and $t_2$ (Fig. 7.9). Note that in this case the phase difference that results in the interference (Eq. 7.52) involves $\Theta_{23}$ (the red area in Fig. 7.9) instead of $\Theta_{12}$ (the green area). This is because only the phase difference that has developed between the first two crossings is relevant; the last part gives just an overall phase factor to the wavefunction.

In figure 7.8 the double crossings case occurs in the upper left corner, above the upper blue line. Below this blue line (but above the lower two blue lines) the one-crossing case is valid. In this lower part we have equivalently plotted the destructive interference in red.
and green. For the two-crossings case the interference factor $I_2$ (Eq. 7.52) involves the first three colored area’s from figure 7.11, $\Theta_{12}$, $\Theta_{23}$ and $\Theta_{34}$ (which is equal to $\Theta_{12}$). So here the phase difference that has developed between $t_1$ and $t_4$ is relevant. The orange regions in figure 7.8 correspond to the regions where this factor $I_2 = 0$. Starting between $t_2$ and $t_3$, the interference factor $I_2$ would contain $\Theta_{45}$ instead of $\Theta_{23}$ and the relevant phase difference has developed between $t_3$ and $t_6$ ($t_6$ is the moment of the crossing after $t_5$); this is indicated as the red regions in figure 7.8. Other starting points are possible, but not indicated in the figure since no new physics is involved.

The semi-classical Landau-Zener-Stückelberg (LZS) model we described is an alternative for the Floquet model, described in sections 7.2.2 and 7.2.4. Both models give approximately the same results, as we compare figure 7.10 with figure 7.7 and figure 7.12 with figure 7.8. Secondly, the regions of destructive interference correspond nicely with the minima of the (generalized) Bessel function in figures 7.7 and 7.8. This shows that both theories work well in the description of our measurements. The Floquet approach is valid for an infinite number of RF periods. In the experiment we work with an 8 MHz oscillation during 20 $\mu$s, i.e. 160 periods, so we believe that the Floquet approach is very well validated. The LZS description is not fully quantum mechanical - the exact time evolution near the crossings is not taken into account - and it does not give an answer.
outside the classically allowed regions, i.e. below the lower blue lines in figures 7.7 and 7.8. However, the LZS description might give more physical insight in the problem, in the interpretation of the occurrence of maxima and minima in the interaction strength, while the Floquet approach is more a mathematical exercise.

7.3 Experiment

7.3.1 Spectroscopy

In the following part we will describe the performed experiments and compare the results with the above presented theory. For the two interacting states we have used two two-atom states, which are coupled through dipole-dipole interaction. We prepare atoms in the 41d state in a confined region in space, and some tens of microns away, we prepare atoms in the 49s state; in both cases by laser excitation. Around 0.4 V/cm the binding energy of one 41d atom plus the binding energy of one 49s atom equals the sum of binding energies of one 42p atom and one 49p atom. The two-atom energy level diagram is depicted in Fig. 7.13. Both the 41d and the 49p have Stark splitting of the two \(|m_j|\) states, giving four two-atom states. The 42p + 49p state (notation: \(n\ell \ell_m j_m j\)) is plotted in green and the 42p + 49p state is plotted in red. The 41d + 49s state is shown in blue. The 41d state is not excited by the laser, so the 41d + 49s state, depicted with a blue dotted line, does not play a role in our experiment. The energies and polarizabilities are calculated with the Numerov method [109] and we get \(W_0 = 25.15(13)\) MHz. The transition

\[
41d_{3/2,1/2} + 49s_{1/2,1/2} \leftrightarrow 42p_{1/2,1/2} + 49p_{3/2,1/2}
\]  

(7.54)

has a difference polarizability of \(\alpha_1 = 347.04(4)\) MHz/(V/cm)\(^2\) and is therefore resonant at a field of \(F_1 = 0.3807(15)\) V/cm. The transition

\[
41d_{3/2,1/2} + 49s_{1/2,1/2} \leftrightarrow 42p_{1/2,1/2} + 49p_{3/2,3/2}
\]  

(7.55)

has a difference polarizability of \(\alpha_2 = 297.40(4)\) MHz/(V/cm)\(^2\) and is resonant at a field of \(F_2 = 0.4113(16)\) V/cm.

The states couple through the dipole-dipole interaction. The single-atom states don’t have a dipole moment on their own (at zero field), but one atom oscillates between the 49s and the 49p state and the other atom between the 41d and the 42p state. These superpositions of two states do have a dipole moment, a so called transition dipole moment \(\mathbf{\mu}: \mathbf{\mu}_{sp} = \langle 49s | \mathbf{r} | 49p \rangle \) and \(\mathbf{\mu}_{dp} = \langle 41d | \mathbf{r} | 42p \rangle\). Transition dipole moments oscillate with a frequency that is given by the energy difference of the contributing states. Both dipole moments are around \(1000 \alpha_0 e\) but can have different polarizations; \(\mathbf{\mu} = \mu_z \hat{\mathbf{z}}\) for \(\Delta m_j = 0\) or \(\mathbf{\mu} = \mu_x \hat{\mathbf{x}} + i \mu_y \hat{\mathbf{y}}\) for \(\Delta m_j = \pm 1\). Note that, in contrast to the situation in a constant field, here the sign of \(m_j\) is relevant. The strength of the dipole-dipole interaction is given by the well-known expression

\[
V_{dd} = \frac{\mathbf{\mu}_{sp} \cdot \mathbf{\mu}_{dp} - 3(\mathbf{\mu}_{sp} \cdot \hat{\mathbf{R}})(\mathbf{\mu}_{sd} \cdot \hat{\mathbf{R}})}{R^3}
\]  

(7.56)
and is approximately equal to $2\pi \times 100$ kHz for a distance of 25 $\mu$m. This results in typical values of the adiabaticity parameter $\delta$ between $10^{-5}$ and $10^{-3}$ (depending on the values of $F_S$ and $F_{RF}$). This means that the crossings are traversed in a highly diabatic manner. The adiabatic transfer probability $\epsilon$ is between $10^{-4}$ and $10^{-2}$, so the approximations eqs. 7.47 and 7.51 are well validated. The Stokes phase $\phi$ lies between $0.248\pi$ and $0.25\pi$.

All 5 involved Rydberg states have a purely quadratic downwards Stark shift. This energy shift is due to the coupling with several $\ell$ states in a field. This means for the eigenstates in a field $\ell$ is not a good quantum number and the orbital angular momentum is not a conserved quantity. It is then not allowed to split the spatial and temporal dependence of the wavefunction (as in Eq. 7.14), because the field changes with time, and therefore $\psi(\mathbf{r})$ changes with time. This is not a problem with linearly Stark shifted states, because here these states have $k$ as a good quantum number, the parabolic quantum number which determines the dipole moment of the state. So in the linear case $\psi(\mathbf{r})$ does not change with the field (and therefore not with time). Fortunately, we need to use only very small fields. In the field of 0.4 V/cm we calculated the eigenstates of a whole range of Rydberg states and it appears that all involved states are more than 99.8% pure, e.g. the population of the 49p part of the wavefunction we call 49p is 99.8%. With this number we were confident that the separation of the space and time dependent part of the wavefunction is well validated (see Eq. 7.10).

In the experiment we explored the coupling strength as a function of the applied static field and the intensity of the radio frequency wave for this quadratically shifted system. In fact we made a few cuts though Fig. 7.8; a few horizontal scans, one scan along the vertical axis and finally several scans, where we followed a curved resonance line and could clearly observe the oscillations. The results (section 7.3.4) are discussed after we have described the experimental setup in section 7.3.2 and the simulations we have done.
7.3.2 Experimental Setup

To control the dipole-dipole interaction strength $V_{dd}$ (Eq. 7.56) we confined the atoms in space by using focused laser beams for the creation of the Rydberg atoms as described in chapters 3 and 5. Two sets of pulsed dye-lasers excite ground-state atoms in a magneto-optical trap (MOT) to the Rydberg states. By using cold atoms, we ensure that the atoms hardly move on the timescale of the experiment. The cylinder-like volumes that contain the Rydberg atoms are approximately 15 $\mu$m in diameter, determined by the laser focus. The length is about 0.5 mm, determined by the diameter of the MOT cloud. Each volume contains about 25 interacting atoms. The distance between the two laser beams is chosen to be 25 $\mu$m. Several ms before the laser excitation the magnetic field for the MOT is switched off, because this field broadens the resonances due to position-dependent and $m_j$ dependent variation of the Zeeman effect.

The atoms are located between two field plates, on which the 8-MHz oscillating field is applied as well as an ionizing field ramp to detect the atoms and their states (state selective field ionization or SFI [35]). The oscillating field is applied by an Agilent 33250A Arbitrary Waveform Generator, controlled by an NI Labview program. The distance vector between the cylinders is in the same direction as the electric field. We use the fraction of 49p atoms ($N_{49p}/[N_{49s} + N_{49p}]$) as a measure for the interaction strength. The exact field is calibrated with a static field scan over the known resonances $F_1$ and $F_2$. The atoms are detected after 20 $\mu$s of interaction time. All data are averaged over 200 realizations. More details on the experimental setup can be found in chapters 3, 5 and 6.

7.3.3 Simulations for multiple atom pairs

In section 7.2.4 we derived expressions for the coupling strength as a function of the oscillating field. However, what we measure in the experiment is the 49p fraction, which is a measure for the 49p+42p state (pp-state) population. The population of this pp-state on resonance is given by

$$P_{pp} = \sin^2 \frac{\Omega}{2} t,$$

since we start with 100% in the sd-state. The interaction strength $\Omega$ is given by Eq. 7.35, which contains the interaction $V$ (Eq. 7.9). This interaction is in this case the dipole-dipole interaction strength $V_{dd}$ (eqn. 7.56), which depends strongly on the atom-atom distance, which is restricted but not fixed. To match the theory better on the actual realistic situation, we performed simulations with multiple atom pairs. We have two elongated volumes, with the atoms sparsely distributed, giving varying distances for the atom pairs. $V$ is also angular dependent, this complicates the problem a lot, also because $\mu_{sp}$ and $\mu_{dp}$ have several possible polarizations. We will assume the angular dependence is averaged out and we use for $V_{dd}$

$$V_{dd} = \frac{\mu_{sd} H_{pp}}{(x^2 + (y + d)^2 + z^2)^{3/2}},$$

with $d$ the distance between the two cylinders.
We randomly placed one particle in an elongated ellipsoid, Gaussian in 3D with full length 400 $\mu$m and radius 8 $\mu$m (both numbers are $1/\sqrt{e}$ widths) at 25 $\mu$m distance of a probe particle. For the interaction strength we used Eq. 7.35, with $\Omega_0/2$ simply equal to $V_{dd}$ (Eq. 7.58). For $\mu_{dd}\mu_{pp}$ we used $800^2a_0^2e^2$. We varied the mixing angle, while keeping the effective field (Eq. 7.25) constant on resonance for different $n$ and $\omega=8$ MHz. The mixing angle is defined as

$$
\theta_F = \arctan \left( \frac{F_{RF}}{\sqrt{2}F_S} \right).
$$

(7.59)

In Fig. 7.8 $\theta_F$ runs from 0 to $\pi/2$, where 0 corresponds to the horizontal axis and $\pi/2$ corresponds to the vertical axis. We evaluated the $|pp\rangle$ probability after 20 $\mu$s for 10000 random cases. The result is shown in Fig. 7.16 as the red lines, where only the height is fitted to the experimental data.

### 7.3.4 Results

As a first experiment we measured the 49p fraction, which is a measure for the dipole-dipole interaction strength, as a function of static field, for various values of the RF field, given in Fig. 7.14. The measured data are depicted in black and are shown on top of a diagram of the resonances, like in Fig. 7.8. The red lines belong to the transition in Eq. 7.54 and the blue lines belong to Eq. 7.55. The green lines depict the exact RF amplitudes of the performed scans, which are $F_{RF} = 0.049, 0.17, 0.4$ and 0.585 V/cm. The green dots indicate where the resonances are expected, i.e. where the green lines cross the red and blue lines.

We observe that the positions of the peaks correspond nicely to the expected positions, the green dots. The exact field values $F_{RF}$ and $F_S$ have been slightly adjusted by a multiplication factor (maximum 2% different from 1) to fit the expected positions, because the exact effective plate distance is not accurately known. The measurements show that indeed more sidebands occur for a stronger RF field, or in other words, more multi-photon transitions can be observed. With an amplitude of only 0.049 V/cm just two sidebands are populated, the one above and the one below the original states. We can also say that we have observed the $n=1$ transition, where the two atom system absorbs one RF photon, and we have the $n=0$ transition, the original resonance, and there is the $n=-1$ transition, where the two atom system emits one RF photon through stimulated emission. When we increase the RF amplitude to 0.17 V/cm we observe sidebands from $n=2$ down to $n=-5$. Surprisingly, at this field the original state is completely depleted: the $n=0$ peaks are missing. At $F_{RF} = 0.4$ V/cm the peaks around $n=-4$ are missing, while at $F_{RF} = 0.585$ V/cm the peaks around $n=-9$ have disappeared. All these missing peaks are nicely predicted in Fig. 7.8 by the green, red and orange areas or the white parts in the resonance curves. Here the generalized Bessel function and therefore the strength of the transition goes to zero, or in other words destructive St¨uckelberg interference occurs.

As a second experiment, we measured the 49p fraction as a function of RF amplitude $F_{RF}$ for $F_S=0$. In other words, we made a vertical cut through Fig. 7.8, along the vertical axis. The results are depicted in Fig. 7.15. The observed peaks fit nicely with the expected resonances, depicted in red (transition in Eq. 7.54) and blue (transition in Eq. 7.55) again after a small adjustment of the field. This figure shows in fact the pure
Figure 7.14: The 49p fraction in arbitrary units, depicted in black, is measured as a function of static field for different RF amplitudes. The red and blue resonance lines correspond to respectively equation 7.54 and 7.55 and run from the +3 photon transition (lower left corner) to the -14 or -11 photon transition (upper right corner). The 0-photon transition is depicted with thicker lines. The resonance peaks are expected where the green lines cross with red and blue, indicated with green dots.

AC-Stark shift of the states, which is \(\frac{1}{4}\alpha F^2_{RF}\) (see Eq. 7.24). Interestingly, we observe only even-photon transitions. In the absence of a static field only an even number of photons can be transferred, due to the selection rules of photon transitions, as was the case in chapter 6 and is also visible in Fig. 7.8. Both atoms make a \(\Delta \ell = \pm 1\) transition, and one photon – with angular momentum 1 – goes from one atom to the other. Adding an even number of photons the transition is possible, because then the total angular momentum of these RF photons can add up to zero. With an odd number of RF photons, however, some angular momentum remains, and the photons can not be absorbed by the two atoms. Already if a small static field is added, odd-photon transitions are allowed, as can be seen in Fig. 7.8 because the static field can contain some angular momentum (but no energy). In other words, with a static field other \(\ell\) states are mixed in the atomic wavefunctions, as described in section 7.3.1, and the photons can couple these parts of the wavefunction. Note that only a tiny fraction of other \(\ell\) states is already enough to break up the selection rules. In other words, for a small field the radial part of the wavefunction is hardly changed, but the time dependent part is changed significantly.

A third experiment illustrates the Stückelberg oscillations most clearly. Here we keep the effective field (Eq. 7.25) constant and fixed to a resonance and we vary the mixing angle (Eq. 7.59). This requires adjustments of both the field amplitude and the static field. In Fig. 7.16 the result is depicted for mixing angles between 0 and \(\pi/2\) for the \(n = -3\) to \(n = +2\) photon transitions of Eq. 7.54. In fact the measurements run over the gray lines.
Figure 7.15: The 49p fraction as a function of RF amplitude at zero static field. The red and blue vertical lines depict the expected positions of the even photon transitions for both transitions given in equations 7.54 and 7.55.

in Fig. 7.8 starting on the bottom. The clear presence of the Stückelberg interference patterns in the data, especially at the positions of the destructive interference, proves that our system is coherent for at least one period of the RF oscillation, i.e. 125 ns. Since the crossing is traversed mostly diabatically, many oscillations are needed to build up the pp population. It can not be proven from these data if this build up occurs coherently or incoherently. However, if we look at figures 7.14 and 7.15, we can estimate the ratio of peak width to peak distance to be approximately 5. This means that $N$ in equation 7.51 is at least 5, so we can say that the interaction is coherent for at least 5 RF periods, i.e. 0.6 $\mu$s. In reality the coherence time could be longer, because the resonance peaks are probably broadened through other mechanisms.

The red lines in Fig. 7.16 show the results of the simulations, described in section 7.3.3. Here only the height and the vertical offset are adjusted to fit the data. The simulations fit the data nicely, proving that this straightforward calculation reflects the real experimental situation quite well. The blue lines Fig. 7.16 depict the squared generalized Bessel function directly (Eqs. 7.22 and 7.23). This reflects a situation of weak or short interaction, since the population of the pp-state (Eq. 7.57) then becomes $P_{pp} \approx V^2 J_n^2$. When the interaction is stronger, the pp probability oscillates between 0 and 1: the process is saturated. However, because multiple distances occur and therefore different interaction strengths, these oscillations are washed out. The result is that the bumps of the blue lines are broadened, as visible in the red lines of the simulation.

In the figure we see again that for small angles (DC-dominated cases) the population is fully in the $n = 0$ state. With increasing RF amplitude the population in this state
Observation of Stückelberg oscillations in dipole-dipole interactions

Figure 7.16: The 49p fraction measured for different mixing angles (Eq. 7.59). From left to right we have increasing $F_{RF}$ and decreasing $F_S$. The effective field is kept constant at the n-photon resonance for each plot. The red lines show the result of the simulation discussed in section 7.3.3 and the blue lines depict the squared generalized Bessel function (Eqs. 7.22 and 7.23) directly. The green vertical lines depict the boundaries of the classically allowed region and correspond to the lower blue lines in figure 7.8.

decreases and more and more higher order states are populated. For higher $|n|$ the start of the increasing population occurs at a larger RF amplitude (or larger mixing angle). This is also visible in the classical boundaries, depicted as green lines, equivalent to the blue lines in Fig. 7.8. Beyond these boundaries we observe the Stückelberg oscillations. On the right hand side of the plot we see again that the odd photon transitions go to a minimum and the even photon transitions go to a maximum, because on this side the static field is zero and the selection rules apply as explained in connection with Fig. 7.15.
7.4 Conclusions

We have studied both experimentally and theoretically interactions between atoms under the influence of an oscillating electromagnetic field. States with a quadratic Stark shift (polarizable states) show an essentially different behavior as states with a linear Stark shift (states with a permanent dipole moment). A comparison between the two cases has been made using the Floquet approach. Where in the linear case the resonance frequency depends solely on the static field and the coupling strength depends solely on the amplitude of the field, in the case of the quadratic shift, both the resonance frequency and the coupling strength depend on both the amplitude and the static offset of the field, and no separation of variables can be made. For the coupling strength in the quadratic case we invoked the generalized Bessel function. Its role is similar to the regular Bessel function for the linear case. The behavior of both functions is studied as a function of frequency and compared to the classical limit; the energy values that occur the most in the oscillating field correspond to the resonances with the strongest coupling strength, apart from some oscillatory behavior. The oscillatory behavior of the Bessel function as well as the generalized Bessel function can be explained in terms of Stückelberg oscillations, which is an interference effect between the developed phases of the two interacting states.

In the experiment the resonance positions and the interaction strength fully fit the described theory. Destructive Stückelberg interference is clearly observed, which proofs that our system is coherent for at least one period of the RF oscillation, 125 ns. From the ratio of the mutual distance the peaks and the width of the peaks, the lower bound is increased by a factor of 5 to 0.6 $\mu$s. Simulations with multiple atom pairs at different distances, corresponding to our experimental situation of two cylinders of atoms, fit the observed interference patterns well.
Observation of Stückelberg oscillations in dipole-dipole interactions
Summary

In this thesis we study dipole-dipole interactions between Rydberg atoms, in connection with feasibility studies of a quantum computer [46, 65]. We have built an experimental setup that allows us to control some important parameters of the dipole-dipole interaction between Rydberg atoms. The use of Rydberg atoms enables us to use relatively low-tech solutions, since these type of atoms have strongly enlarged properties. We excite laser-cooled ground-state atoms at $83(7) \mu K$ to Rydberg states with two tightly focused lasers, such that two separate elongated volumes of cold Rydberg atoms are created with a diameter of 15 $\mu m$. The Rydberg atoms in the two different volumes, typically 30 atoms per volume with principal quantum numbers between 40 and 50, interact with each other over distances of tens of micrometers. The interaction results in a change of the quantum state of the atoms, which occurs in several microseconds, and this state change is what we measure. The experiments described in this thesis are not yet real qubit operations, however, they provide us with more insight in the Rydberg interactions and they show which technological challenges will have to be overcome.

To be able to measure state-changing dipole-dipole interactions between atoms in (multiple) separated volumes like proposed in [81], a detection system is needed that reveals the state of the atoms as well as their position. We succeeded in making and testing such a detection system, which combined the techniques of state-selective field ionization [35] and time-of-flight. The system enables us to measure the atom’s position with an accuracy that ranges from 6 to 40 $\mu m$ over a length of 300 $\mu m$ and to simultaneously detect the state of the atom with a selectivity of 20:1 between $n = 53$ and 54.

However, in the experiments that followed, we did not need this twofold detection system, since we used a transition with four states involved and only two separate volumes; so this interaction can be probed solely with state-selective field ionization. Specifically, we used the transition

$$41d + 49s \leftrightarrow 42p + 49p,$$  \hspace{1cm} (8.1)

which we found after extensive calculations on the energy levels of a large number of Rydberg states in an electric field. The transition is resonant at a very low electric field, 0.4 V/cm, and the transition dipole moments between 41d and 42p and between 49s and 49p are both very large, approximately 1000 a.u. (2500 Debye).

With two cylinder-like volumes of Rydberg atoms, one volume with atoms in the 41d state and the other with atoms in the 49s state, we measure the amount of 49p atoms present after a certain interaction time, as a signature of the dipole-dipole interaction. In a first experiment, we varied the static electric field, the spatial separation between the volumes and the interaction time. We observed, for the first time, dipole-dipole interactions between atoms in separated volumes, and at distances that are enormous, up to 50 $\mu m$. All data are well understood and supported by well-fitting many-body simulations. For
short times or large distances the data fitted the scaling law of the transition probability $P \propto t^2 / d^4$ for infinitely thin and long cylinders at a distance $d$, as opposed to the case of two point particles, where $P \propto t^2 / d^6$.

In the following experiments we added a time-dependent electric field to the system, to explore more properties of the dipole-interacting atoms and to investigate the tunability and switchability of the dipole-dipole interaction. The transition eq. 8.1 is not only resonant for a static electric field of 0.4 V/cm, but also for several different combinations of specific frequencies, amplitudes and offsets of a radio-frequency field. In such cases, one or multiple RF-photons are absorbed or stimulatively emitted by the two-atom system, when undergoing the dipole transition.

In the RF experiments we observe transitions with up to about 10 photons involved. Secondly, having increased the parameter space, we could perform more extensive spectroscopy on the two-atom energy levels, with the resonances being 10 mV/cm (1.3 MHz). The involved states all have a purely quadratic Stark shift in the studied field regime, which results in a significant AC-Stark shift of the resonances. The exact positions of the resonances as well as the oscillatory behavior in the coupling strength are well reproduced with Floquet calculations. The coupling strength is given by the generalized Bessel function, which reflects our intuition in the classical limit: the energy values that occur the most in the oscillating field correspond to the resonances with the strongest coupling strength, apart from an oscillatory behavior. The oscillatory behavior is explained in terms of Stückelberg oscillations, which are due to interference between the two interacting two-atom states. In this view we can proof coherence for at least several RF periods, which is 0.6 $\mu$s.


In dit proefschrift wordt dipool-dipool wisselwerking tussen Rydbergatomen onderzocht. We willen onder meer weten of deze wisselwerkende Rydbergatomen gebruikt kunnen worden voor een quantumcomputer. Een quantumcomputer is een computer die rekent met quantumbits, verkort qubits. Zo’n qubit kan tegelijkertijd de waarde 0 en 1 hebben, dat heet superpositie, terwijl de klassieke (‘gewone’) computerbit óf 0 óf 1 is. Daarnaast zijn de qubits in een quantumcomputer met elkaar ‘verstrengeld’, dat wil zeggen dat de waarden van de qubits met elkaar samenhangen. In deze combinatie van eigenschappen schuilt de kracht van de quantumcomputer; hij kan met alle qubits tegelijk rekenen, terwijl deze ook nog eens alle mogelijke waarden tegelijk hebben.

Nanofysica op micrometerschaal

Qubits moeten met quantummechanische objecten gerealiseerd worden; dat zijn meestal heel kleine deeltjes zoals elektronen of atomen. Wij gebruiken een speciaal soort atomen, zogeheten Rydbergatomen - naar de Zweedse fysicus Johannes Rydberg (1854-1919). Dit zijn heel grote atomen. De grootte van een atoom wordt bepaald door de elektronenwolk die om de kern heen zit. Een elektronenwolk is normaliter ongeveer een Ångström of 0.1 nanometer groot (1 nanometer is $10^{-9}$ meter of 1 miljardste meter). De Rydbergatomen die wij gebruiken daarentegen hebben een elektronenwolk van wel 0.2 micrometer (1 micrometer is een miljoenste meter, of een duizendste millimeter). Dit is enorm, zo’n 1000 keer groter dan normaal. Door deze grootte zijn ze technologisch een stuk makkelijker bereikbaar, terwijl het nog steeds quantummechanische deeltjes zijn. We kunnen hiermee dus typische nano-natuurkunde doen op een micrometerschaal. Misschien is hiermee de ontwikkeling van een quantumcomputer op eenvoudiger wijze haalbaar.

Dipool-dipoolinteractie

Om een quantumcomputer te maken moeten we twee toestanden kiezen voor de Rydbergatomen, die we dan 0 en 1 noemen, en we moeten de Rydbergatomen met elkaar verstrengelen. De verstrengeling gebeurt door middel van elektrische dipool-dipoolinteractie. Deze interactie (wisselwerking) komt veel voor bij atomen en moleculen. De bekende Van der Waals-interactie is een vorm daarvan; het is de oorzaak dat druppels water bij een blijven en gekko’s tegen een muur op kunnen lopen. Een elektrische dipool is een object, waarbij de positieve en negatieve lading uit elkaar zit. Er bestaan ook magnetische dipolen; elke magneet is er een. Net als bij twee magneten, kunnen elektrische dipolen elkaar aan trekken of afstoten. De dipool-dipoolinteractie kan, naast deze afstotende en
aantrekkende krachten, er ook voor zorgen dat de atomen van toestand veranderen. Dat is de eigenschap waar wij in dit onderzoek gebruik van maken.

**Atoomtoestanden en licht**

De elektronen in een atoom kunnen zich in verschillende banen, ook wel 'schillen', bevinden. Deze banen zijn genummerd, beginnend bij 1 en oneindig oplopend. Hoe groter het nummer, hoe groter de baan en hoe zwakker het elektron gebonden is. De toestand van een atoom is de verdeling van alle elektronen over de verschillende banen. In ons geval varieert alleen de baan van het buitenste elektron. Wij gebruiken het element rubidium, dat heeft normaliter het buitenste elektron in de 5e baan; deze toestand heeft de laagste energie. Met licht van de juiste kleur kun je het atoom in een hogere toestand brengen, waarbij één foton wordt geabsorbeerd. Een foton is een lichtdeeltje, en zijn energie hangt samen met de kleur van het licht. Bij de juiste kleur komt de energie van het foton precies overeen met het verschil in energie tussen de hogere en de lagere toestand. Wij gebruiken een laser om de atomen in een hogere toestand te brengen, omdat een laser heel veel licht uitzendt met een hele smalle bandbreedte, oftewel heel precies die ene kleur die je wilt.

In ons experiment gebruiken we twee zorgvuldig uitgekozen toestanden, 41d en 49s. Het cijfer is het baannummer, de letter zegt iets over de draaiing van het elektron. Bij een zwak elektrisch veld kunnen de twee atomen van toestand veranderen door middel van de dipool-dipoolinteractie. Het 41d atoom gaat in energie omhoog naar de toestand 42p (vreemd genoeg een schil hoger, maar toch een lagere energie) en het 49s atoom gaat omhoog naar 49p. Dit proces is een quantummechanisch proces en er is eigenlijk niet één moment waarop de toestandsverandering plaatsvindt; het ene atoom zit in de 41d en 42p toestand tegelijk, terwijl het andere atoom in de 49s en 49p toestand tegelijk zit. Of beter, door de verstregeling kunnen we beter spreken over het systeem van de twee atomen samen, dit systeem zit tegelijkertijd in de 41d+49s toestand én in de 42p+49p toestand.

**Experiment**

We creëren eerst een koud wolkje atomen. Het is belangrijk dat de atomen koud zijn, want dan blijven ze een tijdje op hun plaats, want hoe lager de temperatuur, hoe langzamer atomen en moleculen bewegen. Door middel van laserbundels en een magnetisch veld hebben we de rubidiumatomen op ongeveer één tienduizendste graad boven het absolute nulpunt van temperatuur gebracht: behoorlijk koud dus. Vervolgens schijnen we twee andere laserbundels in het koude wolkje (zie figuur 1). De ene laser brengt atomen in de 41d toestand en de andere in de 49s toestand. Hierdoor ontstaan twee langgerekte gebieden met elk ongeveer 30 Rydbergatomen. We detecteren de atomen na ze een tijdje te hebben laten wisselwerken. De detectie gebeurt door een elektrisch veld aan te brengen dat toeneemt in de tijd. Hierdoor komen één voor één de elektronen vrij, eerst van de atomen in de hoogste toestanden, want die zijn het zwakst gebonden, en daarna die van lagere toestanden. Die elektronen komen op een detector en uit hun aankomsttijd kunnen we herleiden in welke toestand de atomen zaten. We hebben ook een techniek ontwikkeld om tegelijkertijd de plaats van de atomen te herleiden, echter, dit is enigszins complex en we hebben het pas nodig bij meerdere gebiedjes met atomen.
Figuur 1: Een schets van het experiment. Twee verschillende laserbundels maken Rydbergatomen uit een koud wolkje atomen. De atomen uit de twee gebiedjes kunnen vervolgens een toestandsverandering ondergaan van 49s naar 49p en van 41d naar 42p. μm is kort voor micrometer.

In de experimenten meten we het aantal atomen in de 49p toestand terwijl we ofwel de afstand tussen de laserbundels, ofwel de interactietijd, of het elektrisch veld variëren. We kunnen dus verschillende aspecten van de dipooldipoolinteractie bestuderen, zodat we het proces beter kunnen begrijpen. Het elektrisch veld is belangrijk, omdat de atomen een iets andere energie krijgen in het veld. Deze verandering in energie is grafisch afgebeeld op het omslag van dit proefschrift. Bij een bepaald elektrisch veld, het resonantieveld, is het energieverschil tussen 41d en 42p precies gelijk aan het energieverschil tussen 49s en 49p. Omdat energie altijd behouden moet blijven, kan alleen bij deze waarde van het veld de interactie plaatsvinden.

Als het elektrisch veld goed is afgesteld variëren we de afstand tussen de laserbundels (zie figuur 2). Bij zeer grote afstanden, bijvoorbeeld 50 micrometer – ongeveer een haardikte, vindt er nog steeds interactie plaats tussen de atomen in de gescheiden gebieden. Deze meting van atomaire interactie op zulke afstanden is heel bijzonder en nog nooit eerder vertoond. Wanneer we de hoeveelheid 49p atomen meten als functie van de

Figuur 2: De 49p populatie na 10 microseconden in blauw (1 microseconde is 1 miljoenste seconde). In rood de overlap tussen de twee atoomgebiedjes. Boven de figuur de bijbehorende foto’s van de doorsneden van de laserbundels.

Figuur 3: De gemeten 49p populatie als functie van de tijd voor verschillende afstanden; 0, 20, 30, 40 en 50 micrometer.
tijd zien we een gestage groei en daarna een afvlakkning (figuur 3). De snelheid van de groei heeft te maken met de sterkte van de interactie: hoe kleiner de afstand, hoe sterker de interactie en hoe sneller de groei van de 49p populatie. Echter, in het geval van twee atomen op een vaste afstand, zou je niet een gestage groei verwachten, maar een oscillerende: de populatie gaat op en neer tussen 0 en 1 op regelmatige wijze. Dit heeft te maken met de quantummechanische processen van superpositie en verstrengeling. In ons experiment is dit niet zichtbaar, omdat we niet twee atomen, maar twee langgerekt gebieden hebben met meerdere atomen. Hierdoor komen verschillende afstanden en daarmee verschillende interactiesterktes voor en zodoende kunnen we het quantumkarakter niet goed aantonen. In de nabije toekomst is wel het doel om slechts twee atomen te maken in veel kleinere gebiedjes, zodat we deze oscillerende kunnen zien.

In vervolgexperimenten hebben we de afstand en de interactietijd vast gehouden en een oscillerend elektrisch veld in het radio-frequentiegebied (RF) aangebracht. Hierbij kunnen de twee atomen samen één of meerdere RF-fotonen absorberen of uitzenden. Met deze metingen konden we interferentie aantonen tussen de twee toestanden 41d+49s en 42p+49p. Interferentie is een effect dat plaatsvindt tussen golven. Als twee gelijke golven samen komen tellen twee toppen op tot een twee keer zo grote top en twee dalen tot een twee keer zo diep dal (constructieve interferentie). Als de ene golf daarentegen een stukje achter loopt op de ander, dat wil zeggen, ze hebben een verschillende fase, kan een top van de één precies het dal van de ander opheffen en is het resultaat nul (destructieve interferentie). De intensiteit van het resultaat is dus afhankelijk van het faseverschil van de golven. Quantummechanische objecten gedragen zich ook als golven. In het oscillerende elektrische veld kunnen de twee toestanden 41d+49s en 42p+49p een verschillende fase ontwikkelen en op het punt dat het elektrisch veld gelijk is aan het resonantieveld (waar de dipool-dipoolinteractie plaats kan vinden) interfereren de twee toestanden. Afhankelijk van de eigenschappen van het oscillerende veld is deze interferentie constructief of destructief, en dit betekent dat je meer of juist minder in de 42p+49p toestand meet (figuur 4). Met deze meting hebben we bewezen dat ons systeem zich daadwerkelijk quantummechanisch gedraagt gedurende ten minste een halve microseconde.

**Figuur 4:** Interferentiepatronen tussen de 41d+49s toestand en de 42p+49p toestand voor een verschillend aantal uitgezonden RF fotonen.
Bij dit proefschrift zijn vele mensen betrokken, mensen die het mogelijk hebben gemaakt, mensen die geholpen hebben, mensen die een aandeel hebben geleverd en mensen die voor afleiding, gezelligheid en steun hebben gezorgd. Ik wil hen allen bedanken.

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List of publications

C. S. E. van Ditzhuijzen, A. F. Koenderink, L. D. Noordam and H. B. van Linden van den Heuvell

*Simultaneous position and state measurement of Rydberg atoms*


C. S. E. van Ditzhuijzen, A. F. Koenderink, J. V. Hernández, F. Robicheaux, L. D. Noordam, and H. B. van Linden van den Heuvell

*Spatially Resolved Observation of Dipole-Dipole Interaction between Rydberg Atoms*


Atreju Tauschinsky, C. S. E. van Ditzhuijzen, L. D. Noordam, and H. B. van Linden van den Heuvell

*Radio-frequency-driven dipole-dipole interactions in spatially separated volumes*


C. S. E. van Ditzhuijzen, Atreju Tauschinsky, and H. B. van Linden van den Heuvell

*Observation of Stückelberg oscillations in dipole-dipole interactions*