Dipole-dipole interaction between cold Rydberg atoms

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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) µK); 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) µm and 16.3(5) µm. 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 important parameters is depicted in figure 3.1. The vacuum chamber with all its components is shown in figure 3.2.

![Diagram](image)

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 $\tau \sim 5 \mu s$ 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 focus 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 interaction 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 ~1 mm cloud with a temperature of ~100 $\mu$K. The working principle of the MOT, as well as the experimental
3.1 Introduction

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 (~3·10⁻⁸ 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 P₁ and P₂ 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 \( \sim 594 \text{ nm} \), 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 \( \frac{3}{2}nk \), where \( k \) can have values between

![Figure 3.3](image1.png)

**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](image2.png)

**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.
−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 type 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](image)

**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.

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).

![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.](image)

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 5²S₁/₂ (F=3) → 5²P₃/₂ (F=4) resonance in ⁸⁵Rb. This transition is de-
3.2 The Magneto-Optical Trap

Pictured 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 $5^3P_{3/2}$ (F=4) state shift down in energy in a negative B-field – due to the Zeeman effect – and $\sigma^+$ polarized light couples to these states, because of the selection rule $\Delta m=+1$. At the same field, negative m states are shifted upwards and $\sigma^-$ light couples to them. So an atom at $+z$ in figure 3.6 (negative field) will be more resonant to the $\sigma^+$ 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,$m_F=\pm 3$) and $5^2P_{3/2}$ (F=4,$m_F=\pm 4$), which have a shift of respectively $\pm \mu_B B$ and $\pm 2\mu_B B$. At the edge of a beam, 0.5 cm away from the center on the z-axis, the shift of the resonance frequency becomes $\pm 8.4$ MHz. This number is comparable to the laser detuning as well as to the Doppler shift for an atom velocity $v=10$ 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 $5.5(2.2) \cdot 10^{10}$ 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) $\mu$K. In all experiments described in this thesis the temperature was assumed to be below 300 $\mu$K, which is typical for a ru-
bidium MOT \cite{100}. These temperatures of 300 $\mu$K resp. 83 $\mu$K correspond to an average speed of 0.27 $\mu$m/$\mu$s resp. 0.14 $\mu$m/$\mu$s. Both speeds are low enough to regard the particles as "frozen", when studying dipole-dipole interactions at distances of 20 - 50 $\mu$m in 10 - 20 $\mu$s.

### 3.2.1 Diode laser system for the MOT

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{diode_laser_setup.png}
\caption{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 $\lambda/2$ 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 $+1^{\text{st}}$ 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 $\lambda/4$ 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 $\lambda/4$ twice.}
\end{figure}
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 ∼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 +1st 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/cm² and for the z-beam it is about 7 mW/cm². These intensities are a few times larger than the saturation intensity of the \(5^2S_{1/2}-5^2P_{3/2}\) transition, which is \(I_s=1.64\) mW/cm².

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 incoming 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 a 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]

$$N = \frac{F}{\eta \hbar \omega \rho \Gamma},$$

(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 480x752 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 $\rho = \frac{1}{2} \frac{C^2 I/I_s}{1 + C^2 I/I_s + (2\delta/\Gamma)^2}$. \hfill (3.2)

Here $I_s = 1.64$ mW/cm$^2$ is the saturation intensity for the Rb $5S_{1/2} \rightarrow 5P_{3/2}$ transition, $I = 30(3)$ mW/cm$^2$ is the total intensity of all six laser beams together and $\delta = 13(1)$ 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 $19.4(2) \mu m$ in reality, calibrated from the 14 mm holes in the plates P$_1$ and P$_2$, visible in the complete 752×480 picture. We obtained 0.54(6) mm for the 1/√2 diameter, which results in a center density of $5.5(2.2) \cdot 10^{10}$ cm$^{-3}$. This density is sufficient for the experiment. It corresponds to an inter-particle distance of approximately 1.5 $\mu m$.

### 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

$$\Omega = \frac{\Omega_1\Omega_2}{2\delta}, \hfill (3.3)$$
3.3 Rydberg excitation

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\(^2\)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) [56] 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/√e diameter of the intensity profile, is given by

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

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.
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 $\text{Ry} = 109736.605$ cm$^{-1}$ the Rydberg constant for rubidium [35], and $n^*$ the effective principal quantum number $n^* = n - \delta_\ell$, 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 $E_g = -33690.798(2)$ cm$^{-1}$ [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 [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 [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
3.3 Rydberg excitation

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 $0.017(1) \text{ cm}^{-1}$ or $0.51(3) \text{ GHz}$. This linewidth is actually smaller than the value specified in the manual [56], which is $0.024 \text{ cm}^{-1}$ 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 $\sim 500 \mu \text{m}$. 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).

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

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

\[ \rho(r) = \frac{1}{2} \left( 1 - e^{-\left(\frac{se^{-r^2/w^2}}{\sqrt{2}}\right)^2} \right), \quad (3.7) \]

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

![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

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

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

\[ \rho(r) = \frac{1}{2} \left( 1 - e^{-\left(\frac{se^{-r^2/w^2}}{\sqrt{2}}\right)^2} \right), \]

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) \( \mu m \).

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

\[
\int \frac{1}{2} \left( 1 - e^{-F_1(x,y) F_2(x-x_0,y)/F_{\text{sat}}^2} \right) dxdy,
\]  

(3.9)

which is known analytically if the fluence profiles \( F_1 \) and \( F_2 \) 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) \( \mu m \). Assuming identical laser beams the waists are \( 1/\sqrt{2} \) times this number or 13.7(4) \( \mu m \). Including the saturation effect, this gives Rydberg volume diameters of 11.6(4) \( \mu m \) for 49s and 16.3(5) \( \mu m \) 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 \( \mu m \), without assumptions on
the separate beams. Due to the finite temperature of the atoms (\( \leq 300 \, \mu K \)) the diameter of the cylinders increases at most 2 \( \mu \text{m} \) in 25 \( \mu \text{s} \).

These widths of 11.6(4) \( \mu \text{m} \) and 16.3(5) \( \mu \text{m} \) are small enough to measure the dipole-dipole interaction between separate volumes. In chapter 5 we measured a significant signal at a distance between the volumes of 40 \( \mu \text{m} \), which is significantly larger than the upper limit of the convolution of the Rydberg volumes of 23 \( \mu \text{m} \). In the future we would like to decrease the volume size. If the volumes would be 5 \( \mu \text{m} \) in every dimension it should be possible to observe coherence, as described in [81]. This could be achieved by using two crossed laser beams, where one photon from each beam results in excitation to the Rydberg state, drastically shortening the length of the volumes. Furthermore the focus size should be decreased by using a lens with a smaller focal length (inside the vacuum chamber) and/or improving the beam profile.

### 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 \( \frac{3}{2} n k \), which means their energy changes by \( \frac{3}{2} n k \) 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 \( \sim 190 \text{ kHz} \) for a field change of only 1 mV/cm. For the 41d + 49s interaction this number is \( \sim 130 \text{ kHz} \). 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
\begin{equation}
\frac{\partial^2 V}{\partial x^2} + \frac{\partial^2 V}{\partial z^2} = 0, \tag{3.10}
\end{equation}
with $V$ the electric potential or voltage \cite{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.
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 \( \sim 200 \text{ kHz} \). 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]

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

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]

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

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

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]

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

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 \( \tau_{49s} = 9.1 \text{ kHz} \), \( \tau_{49p} = 3.4 \text{ kHz} \), \( \tau_{41d} = 13.3 \text{ kHz} \) and \( \tau_{42p} = 5.5 \text{ kHz} \). 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

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

$$F = \frac{1}{16n^4}. \quad (3.15)$$

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 $\Delta E = \pm \frac{3}{2}n^2F$ 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 Infiniium 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].
The 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 (V/cm)/μs. 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 $0.668 \, \mu s \sqrt{V}$. The ionization time, the time for the electron to find the saddle

![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.](image-url)
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.

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.

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) \(\mu m\) resp. 16.3(5) \(\mu m\) in diameter. The atoms remain in position during the \(\mu s\) experiment, because they are ultra-cold (83(7) \(\mu K\), \(\bar{v} = 0.14 \mu m/\mu s\)), 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 \(nd\) and \(ns\) 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 \(mV/cm^2\)) 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
interaction.