LIFETIME OF RUBIDIUM RYDBERG ATOMS IN A MAGNETO-OPTICAL TRAP

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Abstract

I report my research on the lifetime of rubidium Rydberg atoms in a magneto optical trap. I cover the relevant theoretical aspects of Rydberg atoms, as well as the experimental setup and procedures used to measure actual lifetimes. I measure the lifetimes for rubidium Rydberg states from principal quantum number $n = 35$ to $n = 50$ with $l = 2$, and find they roughly match theoretical predictions for a combination of spontaneous radiative decay and decay induced by thermal background radiation. Measured lifetimes range from of 28.0(7) µs for $n = 35$ to 56(4) µs for $n = 50$. Lifetimes seem to increase at higher densities of Rydberg atoms, by as much as a factor of two for high density $n = 50$ atoms. However, a reasonable explanation for this counterintuitive effect can be given through dipole-dipole interactions between $nD$ states. These interactions create states of different angular momentum quantum number $l$ with longer lifetimes, part of whom can not be experimentally distinguished from the originally created D states. This seemingly increases the lifetime of the original states. As the likelihood of these interactions occurring increases with density, this explains the found relation between density and lifetime.
1 Introduction

Rydberg atoms, atoms with one or more outer electrons in states with high principal quantum number \( n \), are a popular subject in current atom physics. Their unique properties make Rydberg atoms a testing ground for several quantum mechanical phenomena. The ongoing research in the group I joined for this project is directed towards quantum hopping, a name given to the quantum mechanical dipole-dipole interaction of two or more Rydberg atoms at a relatively large distance from each other. To study interactions, the Rydberg atoms have to remain in the same position on relevant timescales. This is achieved by cooling them down to temperatures below 100\( \mu \)K using a magneto-optical trap. In such a system of interacting Rydberg atoms in a magneto-optical trap the lifetime of the atoms is a very interesting property, as it is important to know whether there is significant decay on the timescale of the experiment. Some previous work has been done on these lifetimes, both experimental and theoretical. However, the experimental work is quite rare, and does not deal with exactly the same kind of system as the one we use. The theoretical work on the other hand, deals mainly with single-atom decay and is not very clear on the lifetime of a dense cloud of Rydberg atoms. Therefore I have investigated the lifetime of rubidium (the element used in the ongoing research) Rydberg atoms in a dense magneto-optical trap, both for different \( n \) and for different Rydberg atom density.

In the following section some theoretical background information on relevant properties of Rydberg atoms will be provided, with particular emphasis on their lifetimes. After that, I will go over the experimental setup, with some theoretical background on laser cooling and a detailed description of all aspects of the setup, as well as the procedures used to collect data. Finally I will discuss the results, and draw some conclusions.

2 Rydberg atoms

2.1 Introduction

Rydberg atoms of alkali metals, the group of atoms usually used to study Rydberg states, have one electron in a wide orbit around a compact core with charge +1. Therefore, alkali Rydberg atoms can be treated classically with the Bohr model in many aspects. Two of these aspects are the binding energy of the outer electron, which scales with \( \frac{1}{n^2} \), and the orbital radius which scales with \( n^2 \), meaning a very loosely bound valence electron in a very wide orbit. The low binding energy makes Rydberg atoms very sensitive to electric fields. The wide orbit gives a very long lifetime, as there is little spatial overlap between the excited Rydberg state and the ground states, which makes radiative decay relatively unlikely.

One of the main reasons to study Rydberg atoms is the fact that they can have a large dipole moment. This makes dipole-dipole interactions over distances of tens of microns possible. Coupled to the long lifetime, this makes it possible to study these dipole-dipole interactions extensively, and make them occur in a controlled way. Controlled dipole-dipole interactions may eventually lead to the implementation of the Controlled NOT operation in a system.
of quantum bits, the only operation that requires the interaction of multiple quantum bits, which makes its implementation problematic.

2.2 Lifetime

2.2.1 Spontaneous radiative decay

The first and most basic contribution to Rydberg decay is spontaneous radiative decay. An electron in a high-\( n \) state spontaneously falls to a lower \( n \) state, emitting a photon. Because of their large orbital radius Rydberg states have very little spatial overlap with ground states, making decay unlikely and creating long lifetimes. This effect becomes even more pronounced for states with high angular momentum, which can be explained in a classical manner. An electron with high angular momentum spends less time near the core, making the window of opportunity for decay to the ground state even smaller. To analytically calculate the radiative lifetime of Rydberg atoms it is necessary to calculate the decay rate from a given state to all available states of lower energy, and add all these rates. Calculating the decay rate for the transition between two states involves calculating and overlapping the wavefunctions of the states, which makes the complete calculation quite complex. These calculations have only been performed numerically. However, a simple approximation equation for the radiative lifetime is available that gives excellent results. [1]

\[
\tau_{\text{rad}} = \tau_0 n_{\text{eff}}^\alpha
\]  

(1)

Here \( n_{\text{eff}} \) is the effective principal quantum number as given by \( n_{\text{eff}} = n - \mu_l \), with \( \mu_l \) the quantum defect of an \( nl \) state. In this paper I will use values for \( \tau_0 \) and \( \alpha \) from ref [2] and values for \( \mu_l \) taken from ref [3]. All values for \( \alpha \) are very close to 3, so radiative lifetimes roughly scale with \( n^3 \), which shows the long lifetimes of high-\( n \) states. As the radiative decay rate from one state to another depends on \( \omega^3 \), with \( \omega \) the frequency of the radiated photon, the transitions with the highest frequency will dominate the total decay rate. [4] The highest frequency transitions are those to the lowest \( n \)-values, which means that radiative decay is most likely to occur to the ground state and other states of low \( n \).

2.2.2 Black body radiation

In a system of Rydberg atoms at 0K with no outside influences the observed lifetime should equal the radiative lifetime as given in section 2.2.1. However, outside influences can never be completely annihilated. In a magneto-optical trap, the main factor is thermal radiation coming from the walls of the vacuum system. This blackbody radiation (BBR) can induce state transitions. Rydberg atoms can not be created by excitation through thermal radiation, as the Rydberg energy levels are too far above the ground state. Consecutive Rydberg levels, however, are quite close together. At room temperature the BBR energy \( kT = 25mEv \) is much larger than the difference in energy between consecutive Rydberg states. For example, the energy difference between 44d and 45d is only 0.3 mEv. This means that Rydberg atoms are quite likely to undergo BBR-induced transitions to states of nearby \( n \). Where in other atoms much faster radiative decay would make this effect insignificant, the long
radiative lifetime of Rydberg atoms means BBR-induced decay plays a significant role. As with the radiative decay, exact calculation involves overlapping wavefunctions and summing over possible transitions. Several relatively simple approximation equations for the BBR-induced lifetime are available in the literature. One which is shown to be in good agreement with experimental as well as numerical results is the equation provided in ref. [2].

\[ \tau_{BBR} = \frac{n_{eff}^D}{A} \left( e^{B/(n_{eff}^C T)} - 1 \right) \]  

Here \( T \) is the temperature of the blackbody radiation, which is assumed to be 293K in our experiment. Values for the parameters \( A, B, C \) and \( D \) are provided in ref. [2]. These are fitting parameters, created by fitting this equation to numerical results. To give an idea of the shape of this function it is interesting to note that the simpler equation provided in ref. [4] scales with \( n^2 \). This equation is less accurate mainly at low \( n \). Compared to the spontaneous decay lifetime which scales roughly with \( n^3 \), this means the BBR decay becomes dominant at high \( n \). The crossover point, where spontaneous and BBR-induced lifetimes are equal, lies at \( n = 59 \) (for \( l = 2 \)). As blackbody decay occurs mainly to neighboring states, while radiative decay occurs mainly to the ground states, these two decay mechanisms are easy to distinguish in a state selective measurement, making it possible to test the radiative and blackbody decay separately.

The radiative and BBR-induced lifetimes can be combined to create an effective lifetime in a simple manner. [4]

\[ \frac{1}{\tau_{eff}} = \frac{1}{\tau_{rad}} + \frac{1}{\tau_{BBR}} \]  

This is the lifetime one would expect to find for a single Rydberg atom, isolated from any outside influences except uniform thermal radiation with temperature \( T \), which is in very good agreement with observed lifetimes for a low density magneto-optical trap. [5]

### 2.2.3 Collective effects

While sections 2.2.1 and 2.2.2 should completely cover the decay of a single Rydberg atom, it is quite possible that the lifetime differs for systems of multiple atoms. For instance, some groups have reported superradiance, a form of stimulated radiation, in systems of cold Rydberg atoms; decreasing lifetimes by as much as two orders of magnitude. [6] Other density dependent effects, most likely involving collisions, might also be found to influence Rydberg lifetimes.

### 2.3 Energy levels

Without any electric fields the binding energy of the outer electron is given by.

\[ E = -\frac{1}{2n_{eff}^2} \]  

However, these energy levels change significantly in the presence of electric fields. As we require an electric field to detect Rydberg atoms, their energy levels in an
electric field are important to know. In the presence of such a field the Rydberg atoms become coupled to the field due to their induced dipole moment, causing a shift in the energy levels. This shift of the energy levels in an electric field is called the Stark effect. For states which are degenerate at zero field the first order Stark effect creates a splitting of the energy levels which is linear with the field, creating a series of states with increasing separation at increasing field. States which are not degenerate at zero field are instead affected by the second order Stark effect, which shifts the states quadratically with the field. In rubidium, as well as other alkali metals, $nS$, $nP$ and $nD$ states are affected by the second order Stark effect, and all states of higher $l$ by the first order Stark effect. This is illustrated in figure 1, which shows a so-called Stark map.

![Figure 1: A rubidium Stark map around $n = 21$, showing the binding energy of different states versus the electric field. The blue line depicts the classical ionization limit. Image taken from ref [9].](image-url)

As can be seen from in figure 1, the energy levels of the high-$l$ states, also called the manifold (M) states, cross those of the other states. However, looking closely one can see that there are no actual crossings, instead the energy levels seem to ‘reflect’ off each other. These so-called avoided crossings are caused by deviations of the core potential from simple Coulombic behaviour, as near the core the hydrogen-like picture of a core of charge +1 circled by an electron does not hold up. If the field is varying rather than static these avoided crossings can be ignored, depending on the speed at which the field changes. The combination of avoided and regular crossings, depending on the time derivative of the field, makes a precise determination of the energy a certain state will have in a certain field very complex. For our purposes, the main question is at what field a certain state will ionize. The classical ionization limit is given by $E_{\text{ion}} = -2\sqrt{F}$ with $E_{\text{ion}}$ the ionization energy and $F$ the electric field. This limit is depicted as a blue line in figure 1. An atom will ionize when it crosses this limit. However, as we can not be certain of the exact path of regular and avoided crossings a state takes through the Stark map, we can not calculate the exact field at which it
ionizes. Two theoretical limits exist for this field. These are the diabatic limit, which assumes regular crossings and gives an ionization field of $F = \frac{1}{16n^4 eff}$, and the adiabatic limit, which assumes all crossings are avoided and gives a field of $F = \frac{1}{9n^4 eff}$.

2.4 Dipole-dipole interactions

The combination of a positively charged core and an outer electron relatively far from this core gives Rydberg atoms strong dipole characteristics. Interactions between dipoles are very well known, mainly as producing attractive or repellant forces. If the dipoles are resonant, they can also exchange energy in the form of state changes. For example, two Rydberg atoms in D states can exchange energy and angular momentum, to leave one of the atoms in a lower lying P state and the other in a higher M state [10]. For this kind of interaction to occur the sum of the individual energy levels of the atoms, as given by equation (4), has to remain almost exactly the same, and the $n$ values of the created states should be close to those of the old states. This makes the most likely transition for two atoms in the same $nD$ state the one to $(n+2)P$ and $(n-2)M$. These interactions are more likely at higher $n$, and their probability scales with $\frac{1}{R^3}$, where $R$ is the distance between the dipoles [10].

3 Experiment

3.1 Experimental background

3.1.1 Magneto-optical trap

To be able to study Rydberg atoms effectively we want them to be confined to a defined region of space during the experiment. This is achieved by creating a magneto-optical trap of rubidium atoms, which both traps the atoms and cools them to around 90 $\mu$K [7] [8]. These cold trapped atoms are then excited to Rydberg states.

A magneto-optical trap works on the principle that a photon being absorbed by an atom transfers its momentum as well as its energy to the atom. Therefore, a cloud of atoms bombarded by a constant stream of photons will gain momentum in the direction of the light. The atoms, now in an excited state, will then spontaneously emit a photon and lose the gained energy and momentum again. However, since the direction of this emission is random, the net effect will be an increase of momentum in the beam direction.

By tuning the wavelength of the light slightly to the red of a known resonance of the atoms, one can ensure that only atoms moving towards the light absorb the photons, due to the Doppler effect. This means the atoms moving towards the light are slowed down, while the other atoms are unaffected. In this experiment we use the $5S_{1/2} (F=3) \rightarrow 5P_{3/2} (F=4)$ transition in $^{85}$Rb. Aiming six of these beams orthogonally on the same cloud of atoms will slow the atoms down in all directions, cooling them dramatically. This effect alone, while cooling the atoms down, will not confine them. Atoms moving slowly, with velocity too low to be in resonance with the light, can escape the laser beams. This is where the magnetic field comes in. The trap is created at the center and zero-
point of a magnetic field created by two parallel coils with opposite currents, the so-called anti-Helmholtz configuration. An atom moving out of the zero-point will experience an increasing magnetic field, which causes Zeeman-splitting of its energy levels. This Zeeman splitting will cause the atomic transition to move towards the frequency of the light, enabling slow moving atoms to absorb photons as well. Summarizing, the magnetic field combined with the cooling lasers acts as a position dependent force through the Zeeman-splitting, confining the atoms to the center of the configuration. This constitutes a magneto-optical trap (MOT).

Most alkali metals, such as the rubidium used in this experiment, have two different ground states, due to hyperfine splitting. In our experiment these are the $5S_{1/2} (F=2)$ and $5S_{1/2} (F=3)$ states. The cooling laser is tuned to a transition from the $(F=3)$ ground state to a certain excited state, but spontaneous emission can decay the atom to either of the ground states. An atom that decays to the wrong ground state will not be affected by the cooling laser anymore and escape the trap, destroying the MOT. Therefore, another laser beam is required to bring the atoms decaying into the wrong ground state back to the one used for the cooling transition. This laser is called the repumper. Our repumper excites the $5S_{1/2} (F=2)$ atoms to the $5P_{1/2} (F=3)$ state. From there they can decay into either ground state, but as long as the atoms in the wrong ground state keep getting excited they will eventually end up in the cooling state again.

3.1.2 Rydberg excitation

There are several ways of exciting rubidium atoms to Rydberg states. I will outline the two most used methods. The first is by using a blue 480 nm laser to excite rubidium atoms from the 5P state created by the cooling laser to any of the high-\textit{n} S or D states. The second is through a two-photon process with a 594 nm laser, which excites the atoms from the 5S state to a virtual P state in between 5P and 6P, and from there to the high S or D states. Both processes are schematically depicted in figure 2. The exact state created can be determined by fine-tuning the laser to the wavelength associated with that particular state. In our experiment we excite the atoms through the second process, using a pulsed Lambda Physik ScanMate Dye Laser, pumped by a Nd:YAG Laser. Because we use a two-photon process, the excitation probability scales with the laser intensity squared [9]. A side-effect of using a two-photon process is that three-photon processes can also occur, ionizing the rubidium atoms. Depending on the laser power this creates a noticeable signal on the detector, however this does not interfere with the Rydberg detection.

3.1.3 Rydberg detection

To detect the Rydberg atoms we use the method of state-selective field ionization (SFI). The Rydberg atoms are ionized by an electric field, which also serves to accelerate the now unbound electrons towards a micro-channel plate detector. The state-selective part works on the principle that different states ionize at different electric fields, due to different binding energies of the electron in the highest state. This means that the gradually increasing electric field will ionize different Rydberg states at different times, which can be seen by looking at the electron detection versus the time.
Rather than attempt to trace different paths through the Stark map to calculate the fields for which different states ionize (see section 2.3), we calibrate so-called timing windows for states of interest by creating different $nD$ states and measuring them without any delay between the Rydberg excitation and the start of the electric field ramp. States of different $l$ are then expected to ionize in between these $D$ states. This does of course require a well-calibrated laser to ensure we know what state we are creating, but this is essential anyway as we want to measure the lifetime of particular states, so we need to be able to selectively create these states. This also requires a narrow enough linewidth, to prevent the creation of multiple states at the same time. Calibration of the laser will be explained in section 3.3.1.

### 3.2 Experimental setup

A general overview of the vacuum setup the MOT is created in can be seen in figure 3. The system is kept at a pressure of roughly $3 \times 10^{-8}$ millibar by a turbo pump. The MOT cooling beams are provided by a 780 nm laser diode, the setup of which will be explained in section 3.2.1. The beam from this laser is split three ways, each beam entering the chamber orthogonal to the other two, and being reflected back through the chamber for a total of six cooling
Figure 3: a) Overview of the vacuum system. Detailed explanation is provided in the main text. b) Close-up of the MOT. Field plates P1 and P2 create the electric field required for Rydberg ionization and electron acceleration. c) Close-up of the dispenser. A voltage is applied on the two plates to catch off any produced ions, and a grounded plate around this setup shields the resultant field from the rest of the system. Image taken from ref. [9]
beams crossing at the center of the chamber. The seventh beam required for the MOT, the repumper, comes from a 795 nm laser diode, which corresponds to the transition from the ground state 5S_{1/2} (F=2) to 5P_{1/2} (F=3), from where the atoms will decay into the 5S_{1/2} (F=3) state required for the cooling.

Rubidium atoms are supplied by a SAES Getters rubidium dispenser positioned just underneath the location of the MOT. This is a 1 mm wide and 1 cm long electrical filament containing rubidium chromate (Rb_2CrO_4) and a reducing agent, which gives off rubidium atoms when we run a current of 4.7A through it. However, it also seems to give off charged particles, which is why two metal plates with potentials of 6 V and -6 V are put in front of it to catch off any charged particles. To shield this electrical field from the rest of the system, a grounded metal plate is placed around it, with a slit in it for the rubidium atoms to pass through. The dispenser setup is shown in figure 3c.

In the middle of the chamber, around the position of the MOT, two circular metal plates provide the electric field required to ionize the Rydberg atoms. These plates have a diameter of 55 mm, with a 14 mm diameter hole in the middle to allow the cooling beams as well as the ionization products to go through, and are 25 mm apart (see figure 3b). Plate P1 is ramped to several hundred volts in a couple of microseconds using a high-voltage power supply triggered by a Hewlett Packard pulse generator. The exact voltages are unimportant to this experiment, as timing windows are compared to calibrated measurement traces rather than calculated ionization voltages. The HP pulse generator adds an adjustable delay to the trigger of a central function generator and also triggers the oscilloscope used to gather data. The central function generator, which also triggers the dye laser that excites the Rydberg atoms, is set at 10 Hz, creating a refresh rate of 10 Hz for the entire experiment. This triggering setup means that the moment the ionization ramp starts and the moment the scope reads out the signal stay fixed relative to each other, but can be delayed with respect to the laser pulse, which always occurs at a fixed point in time.

In the side arm of the vacuum chamber a Hamamatsu micro-channel plate detector (MCP) is positioned to detect the electrons coming from the ionized Rydberg atoms. The electrons are accelerated by the potential over the plates P1 and P2, and fly a distance of 28 cm to the MCP. By equating the potential energy on the atom from the electric field to kinetic energy [9], the time this takes follows as \( t = \frac{668}{\sqrt{V}} \) ns. The MCP, which is connected to a power supply providing 2.5 kV, multiplies every electron hit by a factor \( \sim 10^6 \), and this signal is fed into a HP Infinium oscilloscope for direct observation or readout with a computer.

Around the main vacuum chamber two large MOT coils provide the magnetic field required for the MOT. Four smaller coils make it possible to move the MOT around, by altering the position of the minimum of the field created by the main MOT coils. Four rectangular compensation coils around the entire setup compensate for the earth magnetic field and other magnetic fields caused by electronics around our setup.

3.2.1 Laser setup

The 780 nm cooling beam and the 795 nm repumper beam both come from the same kind of setup, illustrated in figure 4. Light from a Toptica DL 100 diode laser hits a reflection grating, which reflects the first order back into the
diode. The grating and the back of the diode now form the lasing cavity. The zeroth order is reflected off the grating and towards the rest of the setup by a mirror. The laser frequency is adjusted by changing the angle of the grating, either manually with a screw on the grating, or automatically through a piezo actuator. The beam goes through an achromatic prism pair to change the beam profile from elliptical to circular, and then through an isolator to prevent reflection back into the diode, which could cause damage to the diode. Then it is split by a $\lambda/2$ plate and a polarizing beam splitter, with most of the beam going towards the MOT setup and a small part into a Doppler-free frequency modulated spectroscopy setup to control the laser frequency.

![Figure 4: Schematic of the laser setup. The beam in the top right goes into the main experiment. The bottom half constitutes the spectroscopy setup. Detailed explanation in the main text. Image taken from ref. [9].](image)

In the spectroscopy setup the beam is split again, into a low intensity probe beam and a higher intensity pump beam. Both are directed through a rubidium vapor cell, with the probe beam ending up on a photodiode. The counterpropagating pump beam creates small Doppler-free peaks on the Doppler broadened absorption spectrum of the probe beam. These are caused by atoms with no velocity in the direction of the beams, which absorb photons from both beams, thereby absorbing slightly less of the probe beam than other atoms. These Doppler-free peaks make it possible to resolve the hyperfine structure of the atoms. Some additional peaks are created by atoms moving with a velocity that causes them to be in resonance with both beams for two different transitions, the so-called crossover peaks. A 20 MHz signal is superimposed on the signal from the photodiode, creating the derivative of the absorption spectrum, with zero crossings at the resonances. This signal is fed into the piezo actuator of the laser grating through a lock-in amplifier, keeping the frequency at the selected resonance.

We lock the cooling laser to the $F=2$, $F=4$ crossover peak located at -93 MHz from the $5S_{1/2}$ ($F=3) \rightarrow 5P_{3/2}$ ($F=4)$ resonance used for cooling, and the repumper directly to the required $5S_{1/2}$ ($F=2) \rightarrow 5P_{1/2}$ resonance. The repumper is directed straight into the vacuum chamber. The cooling beam first goes through an acousto-optical modulator which adds 80 MHz to the signal, so that it ends up at -13 MHz from the used transition. This beam is then split three ways, directed through the chamber in the x-, y-, and z-direction and reflected at all ends to create the six required beams.
3.3 Method

3.3.1 Wavelength scan

To calibrate the dye laser we perform a wavelength scan before each series of measurements, to correct for any changed conditions or laser drift. This is an automated procedure, controlled by a Labview interface. The wavelength of the excitation laser is altered in steps of $10^{-12}$ m, and for each wavelength the total number of detected electrons, and therefore Rydberg atoms, is counted. This gives a regular peak structure, with major peaks for each $nD$ state created and (if the linewidth is narrow enough) smaller peaks for the $nS$ states. Such a wavelength scan can be seen in figure 5. By comparing this peak structure to calculated resonance wavelengths we are able to calibrate the wavelength of the laser. The resonance wavelengths in nm are given by $\lambda = \frac{2 \times 10^7}{E_n - E_g}$, with $E_n$ the binding energy of the state and $E_g$ the energy of the ground state, $E_g = -33690.798(2) \text{ cm}^{-1}$ [9]. The binding energy of the states is given by $E_n = -\frac{R_y}{n_{\text{eff}}^2}$ with $R_y = 109736.605 \text{ cm}^{-1}$ the Rydberg constant for rubidium [4] and $n_{\text{eff}}$ the effective principal quantum number. This also has to be corrected for the refraction index of air, as the laser operates in air while the Rydberg atoms are in vacuum.

![Wavelength scan graph](image)

Figure 5: Dye laser wavelength scan from 594.5 to 595 nm, with peaks corresponding to 46D to 39D. Small S-state ‘shoulders’ can be seen on the left hand slope of some of the peaks.

The spacings between the peaks found in the wavelength scan are compared with the theoretical distances between the peaks. Because the binding energy of the states decreases quadratically with increasing $n$, no two states have the same energy difference between consecutive states, so this examination of the energy spacing between created states conclusively determines what states are created at which wavelength input value, which allows us to fine-tune the laser.

3.3.2 Lifetime measurements

It is clear from figure 5 that at the time of my experiments the linewidth of the dye laser was not good enough to properly resolve S-states. Therefore I restricted my measurements to the lifetime of different $nD$ states. The lifetime measurements are performed using a labview program, which I created specifically for this purpose. The interface and entire code can be seen in appendix A.
The program controls both the excitation laser frequency and the delay between the excitation pulse and the field ramp and oscilloscope trigger. It changes the delay in pre-set steps and for each delay reads out the oscilloscope a set number of times, averages the traces and saves the result. Then it tunes the laser to the next pre-set wavelength. In this way the lifetime of multiple states can be measured in one go.

![Figure 6: Calibration picture showing scope traces for states 37D to 46D, with 46D on the left and 37D on the right. The SFI ramp starts at t=0.](image)

To analyze the results, traces of all relevant decay states that might be created by decay of the states to be measured are required, so that these decay states can be recognized in the results. This is done by taking a measurement for any number of likely and measurable decay states without any delay. A calibration graph created in this manner for the lifetime measurement of the 40D to 44D states can be seen in figure 6. Calibration measurements are performed with 50 averages for each data point, lifetime measurements with 200 averages. Figure 7 shows the saved oscilloscope traces for several delay times of the 42D state. It is clear that the signal decreases over time, and it is also possible to see a minor peak appear to the left of the main peak. To determine the lifetime of a single state, I determined integration boundaries within which I considered the signal to be coming from that one state, based on both the calibration and the measurement scope traces. An example of these integration boundaries can be seen in figure 7. I then integrated the signal over this region, creating a value for the total number of Rydbergs in a specified state detected as a function of delay time. To determine the lifetime, I fitted an exponential decay function to this graph. The number of Rydbergs versus delay time graph for 42d can be seen in figure 8. How well the exponential function fits gives an error value for the lifetime.
3.3.3 Lifetime versus Rydberg density

Apart from measuring lifetimes for various $nD$ states I also measured lifetimes for single $n$ states with varying excitation laser intensity. Using a polarizing beam splitter and a $\lambda/2$ plate I altered the intensity of the beam going into the vacuum chamber, while measuring the power of the beam with a power meter. Using the duration of the laser pulse, 8 ns, and the beam diameter inside the MOT, 15 $\mu$m, I calculated the beam intensity inside the MOT. Squaring this intensity (see section 3.1.2) gives the probability an atom has of being excited by the laser, barring some constant. The number of excited atoms, and therefore
the Rydberg atom density, is directly proportional to this probability.

4 Results and discussion

4.1 Decay of $nD$ states

4.1.1 Lifetimes

The main result of this section, the lifetimes of rubidium D states from $n = 35$ to $n = 50$, can be seen in figure 9. It is clear from this picture that at regular Rydberg density our system does not exhibit any effects that greatly change the lifetime from the value predicted for a single atom.

![Graph of the lifetime of rubidium D states 35D up to 50D. Error bars indicate how well the measured data fitted to exponential decay. The solid line is a prediction based on equations (1), (2) and (3).](image)

However, at most $n$ values, the predicted values do not fall into the error bars. This can be partly explained by the results of section 4.2, as I did not extensively monitor the dye laser power, and thereby the Rydberg density during these measurements. It should also be kept in mind that the depicted error bars solely give information about the quality of the exponential fit to the data, without dealing with any errors in the data itself. Possible other errors might include fluctuations in the MOT and a nonuniform blackbody radiation field due to the shape of the vacuum chamber. Furthermore, different parts of these measurements were performed on different days, which might cause some variation.

4.1.2 Decay products

Apart from looking at the decay of a single state, I also studied the scope traces to look at the states being produced in the decay. Figure 10 shows a trace for the 42D state ionized after a 50µs delay, with the calibration measurements in
Figure 10: Scope trace of rubidium 42D ionized 50µs after excitation, plotted over calibration measurements for states 40D to 44D, with a vertical shift.

the same graph for comparison. This picture shows several interesting features, which also show up clearly in other studied scope traces. There are two minor peaks visible, to the left and right of the main peak. The peak on the left exactly matches the 43D calibration peak. However, due to conservation of angular momentum a D state cannot decay directly into another D state. The main nearby states 42D should decay into nearby P and manifold states. Looking at the energy levels (see section 2.3, in particular figure 1) we expect to find the 41M states and then the 44P state when going towards lower ionization field (left in the graph), and the 43P state followed by the 40M states towards higher ionization field. The P states should give a clearly defined ionization peak, while the manifold states ionize over a broad range because of energy level splitting. Decay to another D state can only happen from a P or manifold state, which makes it unlikely that another D state gives a noticeable peak. This leads me to conclude that the left peak matching 43D is given by the 44P state, and more generally that in the range of this experiment an nD state and an \((n + 1)\)P state ionize at the same field. The manifold states are most likely contained in the tails on the right of the visible peaks. This last point is strengthened by figure 11, which shows the calculated lifetime for each point in the oscilloscope traces, within a relevant range. This plot shows increasing lifetimes at later times, or higher ionization field, around the right integration boundary. These lifetimes should not be interpreted directly as atom lifetimes, but rather as the decay time of the signal. If the created 42D atoms decay into 40M states, which ionize around the right integration boundary, then it is natural that the signal in this area remains relatively large over time.
Figure 11: Lifetime of the 42D state signal versus the actual time (also interpretable as electric field). Dashed lines are the used integration limits (same as in figure 7).

Summarizing the information extracted from figures 10 and 11 I conclude that the 42D state definitely decays into the 44P and 40M states, observable by the peak to the left of the main peak and the tail on the right side of the main peak. The likely decay into 43P and 41M states is not observable, as these signals are contained in the main 43D peak and the small tail of the 44P peak. The small peak visible to the right of the main peak could be 42P, although it does not match the overlapping D peak as well as the other P states do. Perhaps a more likely candidate is 43S, since this state should ionize at a slightly lower field than 41D, which would account for the signal lying on the left edge of the 41D calibration peak. All these decays are caused by blackbody radiation, as spontaneous decay occurs mainly to the ground state and only to states of lower energy, while BBR-induced decay occurs to nearby states, both higher and lower in energy.

4.2 Lifetimes at various Rydberg densities

I measured the lifetimes of the 40D, 45D and 50D states for several laser intensity values. The results of these measurements can be seen in figures 12, 13 and 14. These figures show some surprising behavior. Rather than decreasing lifetimes at higher density, which could be caused by collision or stimulated radiation effects, they seem to show increasing lifetime at higher Rydberg density, up to double the predicted lifetime. Also, at low Rydberg density, the obtained values are below the predicted values. However, while some of the error bars at low density are quite small, suggesting a high quality fit, these fits are based on very low signal, and don’t appear to be very reliable on closer examination.
Figure 12: Lifetime of the rubidium 40D state for differing excitation laser intensity squared, which is proportional to Rydberg density. The horizontal line corresponds to the value given by equations (1), (2) and (3).

Figure 13: Lifetime versus Rydberg density of the 45D state.
Figure 14: Lifetime versus Rydberg density of the 50D state.

The apparent increase of the lifetime of all three measured D states at high density is a much more significant result. However, an actual increase of the lifetime of the same state is impossible. The spontaneous and BBR-induced decay can not be repressed by a high atom density. Therefore, it is most likely that this result actually stems from the production of different states, in some density related process. From section 4.1.1 we know that the nD state is indistinguishable from the (n + 1)P state and overlaps with the (n − 2)M states. Both P states and manifold states live longer than D states, so if these states are created they could account for the observed longer lifetimes. We also know of a process that can create these states: dipole-dipole interaction. From section 2.4 we know that the most likely interaction between two nD atoms sees them end up in the (n + 2)P and (n − 2)M state, but (n + 1)P and (n − 1)M are also possible. However, as the lifetime of 51P is not long enough to account for the last point in figure 14 the former option, with the (n − 2)M states creating the longer lifetimes, gives a more complete explanation. Evidence for this interaction taking place can also be found in the scope traces for the high density measurements, as they all show significantly larger peaks at (n + 2)P than their low density equivalents. Because the dipole-dipole interaction probability scales with $\frac{1}{R^3}$, with $R$ the distance between the atoms, the observed increase in lifetime with increasing density makes sense. At higher densities a greater number of long-living manifold states are created. This also goes for higher n (see section 2.4), which explains why the measured lifetime increases much more for 50D than it does for 40D and 45D.

5 Conclusion

My first conclusion is that the decay of rubidium Rydberg nD atoms in our magneto-optical trap does not differ significantly from theoretical predictions considering only spontaneous and blackbody radiation induced decay. There is no evidence for superradiance or other lifetime reducing effects, even at high Rydberg density. My second conclusion is that at high Rydberg density dipole-dipole interactions occur between nD states, creating (n + 2)P and (n − 2)
manifold states, and perhaps also \((n + 1)P\) states and \((n - 1)M\). As these states partly ionize at the same field as the original D states this process seemingly increases the lifetime of the D states.

To perform more accurate measurements of single-state lifetimes as well as confirm my observation of dipole-dipole interactions a detection method is required that can fully distinguish between S, P, D and M states. The general lifetime measurements could be further improved by making sure they are all carried out at the same Rydberg density. The accurate measurements of Rydberg density this would require would of course also improve the lifetime versus Rydberg density measurements.

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References

Appendices

A Labview interface

This appendix contains the front-end of the labview interface I created (figure 15) as well as all of the code. Figure 16 shows the overall code structure, with all consecutive block elements shown in figures 17 to 24. The code includes several sub-programs created by others, which are shown as question marks in the code structure.

Figure 15: Front-end of the labview interface
Figure 16: Labview code outer shell with element A0. Question mark components are oscilloscope and laser initialization programmes.

Figure 17: Code element B1
Figure 18: Code element A1. The question mark component sets the wavelength of the dye-laser.

Figure 19: Code element C1

Figure 20: Code element E1
Figure 21: Code element D1
Figure 22: Code element F1. The question mark component reads out the oscilloscope.
Figure 23: Code element F2

Figure 24: Code element D2