# Time-resolved coherent excitation dynamics of Rydberg states in thermal vapors

MASTERARBEIT

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## Declaration

I declare that I authored this work independently. I have not used any further sources or means than those mentioned in this thesis, and all citations are marked as such.

Stuttgart, October 24, 2013

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# **1** Introduction

Rydberg atoms are atoms with a highly excited outermost electron, which can be brought into this state by interaction with light. This electron is characterized by quantum numbers, which correspond to its energy and angular momentum. If both are large, the electron will be far distant to the core. Due to the correspondence principle the electron then behaves like a classical particle.

This circumstance helped the early atomic physicists to understand the structure of the atom. The hydrogen atom was well understood by analytical solutions of the Schrödinger equation, as it comprises only two particles: the core and one electron. However, larger atoms consist of more than two electrons, which tremendously complicates the treatment. Such atoms excited to Rydberg states, in turn, can be similarly described to the hydrogen atom, as a separation of the outermost electron from the core and inner electrons is possible.

If only the energy of the outermost electron is large, but not the angular momentum, the electron can also be found far distant to the core, but not necessarily: It is nonclassically distributed in a large "cloud" overlapping the core atom. It turned out that atoms in such states are extremely interesting systems, as they are highly sensitive to their surroundings. For instance, Haroche and coworkers used Rydberg atoms in order to detected single photons without changing their wave nature [1]. Thereby fundamental laws of quantum mechanics could be manifested. For appreciation of this work, Haroche obtained the Nobel Prize in 2012, shared with D. Wineland.

Related to their sensitivity, Rydberg atoms show further extraordinary behaviors. For instance, Rydberg atoms have long-range potentials. For this reason, they prevent any excitation of other atoms to Rydberg states if they are in close vicinity. This effect is called blockade [2, 3]. The blockade is the reason that in large ensemble of atoms only one atom can be excited to a certain Rydberg state. Thereby, with respect to certain aspects of atom-light interaction, macroscopic units consisting of atoms can behave like single atoms and hence quantum-mechanically. This opens a wide range of applications in novel technologies basing on quantum mechanics, like quantum computation or quantum communication [4].

Moreover aspects of fundamental interest in modern physics can be investigated using Rydberg atoms, again using its long-ranging interactions. For example, solid state systems can be experimentally modeled by Rydberg atoms.

Usually experiments with Rydberg atoms are done by cooling them close to absolute zero temperature by expensive and laborious cooling techniques. This is done in order to prevent atomic movement and thereby different energies of the atomic levels due to the Doppler effect, as well as other effects like the transit-time effect or collisions. Then the spectral lines of the atom ensemble is narrow and effects of Rydberg interaction can be observed clearly. However, such cooling limits the practicability of such systems. Fortunately, all effects corresponding to Rydberg atoms principally occur also in ensembles at room temperature. In order to circumvent the effects due to the movement of the atoms, such as spectral Doppler broadening, Rydberg excitation and probing processes have to be executed on very short time scales (frozen gas regime).

Work at PI5 MicCell group This is a way which is pursued within the Micro cell group (at which the author carried out this thesis). In this group, important results with respect of Rydberg atoms in thermal vapors have been obtained prior to this thesis [e.g. 5, 6].

The established method to detect Rydberg excitations in thermal atomic clouds is to probe the atoms by exciting with resonant laser irradiation.

A frequently used way to create Rydberg atoms is a two-photon excitation by radiation near resonant to atomic level transitions. By excitation to the Rydberg state using pulses of few nanoseconds, coherent Rabi oscillations could be observed, beating the Doppler broadening bandwidth [5]. Hence evidence of strong Rydberg-Rydberg van der Waals interaction was found [6].

This means an important step towards the combination of strong Rydberg-Rydberg interactions and four-wave mixing [7]. Thereby, non-classical photon sources may be realized, potentially towards single photons [8]. This technique is a promising candidate for a raising applicability of single photon sources, since individual systems are intrinsically identical, which means photons produced by different sources are indistinguishable, especially with respect to the frequency. Competing techniques basing on solid state systems do not provide intrinsic indistinguishability.

**Cesium experiment** In order to circumvent the pulsed laser system, which has only low repetition rates, a second experiment has been set up recently. The new aspect of this system is a Pockels cell [9], which allows a more controllable pulse length and thereby enables an investigation of time scales of few nanoseconds. In order to enable the application of this device, a different excitation scheme (inverted excitation scheme) was chosen. After the setup of the laser system [10] first experiments [11, 12] exhibited effects caused by the modified excitation scheme, such as double resonance optical pumping.

**This thesis** In the scope of this thesis, the near-term task has been time-resolved measurements of the coherences of Rydberg states.

In section 2, the set up of the two-photon excitation experiment is described.

The relevant features of cesium, especially spectral properties, as well as the excitation paths to Rydberg states are described in section 3.

Basics of atom light interaction accompanied by simulations neglecting Rydberg interactions are introduced in section 4. In section 5 we outline optical aspects, especially with respect to imaging optics.

A strong coupling pulse leads to a.c. Stark shifts. This is investigated in section 6.

Principals of Rydberg interactions are introduced in section 7, where they are also applied for modeling of Rydberg interactions in atomic ensembles and compared to measurements at low densities.

In section 8, finally, measurements at high densities and thereby in the strong Rydberg interaction regime are discussed. The discussions of the measurement results are induced by theoretical aspects concerning the Rydberg potential structures and critical behavior.

Section 9 concludes the presented results and gives an outlook.

# 2 Experimental setup

The experimental setup basically consisted of three parts:

- the laser system, containing light generation as well as frequency locking and shifting for the wavelength 455 nm, light generation, laser amplification, and pulse shaping for the wavelength 1066 nm, and light generation for the wavelength 852 nm;
- the pulsed experiment part containing basically the vapor cell and the signal detection;
- the reference spectroscopy system, integrated in the pulsed experiment, necessary for absorption measurements.

## 2.1 Laser system setup

In the experiment, three different wavelengths were involved:  $455 \text{ nm} (6S_{1/2}-7P_{3/2})$ , around  $1066 \text{ nm} (7P_{3/2}-nS_{1/2})$ , and  $852 \text{ nm} (6S_{1/2}-6P_{3/2})$ . The first two wavelengths correspond to the Rydberg excitation scheme, whereas the latter corresponds to the  $D_2$  line of cesium that was used for density reference measurements (c.f. figure 3.1).

## 455 nm laser system<sup>1</sup>

The 455 nm light was generated by a Toptica Photonics TA-SHG pro high power, frequency-doubled, tunable diode laser system. The system contains an internal frequency stabilization by an external cavity diode laser (ECDL) as well as amplification by an tampered amplifier (TA).

The atoms are excited off-resonantly by a two-photon excitation with a detuning of 1.5 GHz from the  $7P_{3/2}$  state. For the frequency locking, some light was split off and was then red-shifted by -1.5 GHz. This light was then used for locking the laser via dichroic atomic vapor laser lock (DAVLL) spectroscopy to the  $6S-7P_{3/2}(F = 4 - F' = 5)$  hyperfine transition in a cesium cell [13]. In order to be used for the pulsed experiment in a different location, it was coupled into an optical fiber. This laser system provided powers of about 250 mW, however, due to the reference and fiber losses, about 120 mW were available in the experiment.

<sup>&</sup>lt;sup>1</sup>The setup of this laser system is described in detail in [10, ch. 3.2].

## 1066 nm laser system<sup>2</sup>

The infrared light around 1066 nm was generated by a Toptica Photonics DL 100 pro design, which is a grating-stabilized, tunable single-mode diode laser. Its output wavelength ranged from 1040.6 nm to maximally 1078.5 nm, where the maximum gain was at 1066.0 nm.

In the experiment this laser is typically scanned spanning a frequency range of few GHz, where a Fabry-Perot resonator is used for a relative frequency scale. In order to get a zero frequency, a Rydberg electromagnetically induced transparency (EIT) signal is used [14]. In order to drive the 6S-7P transition, a light beam coming from the 455 nm laser system was used. The remaining beam was amplified by an external TA, before it was coupled into a fiber going to a fiber amplifier (Keopsys). This device provides an output power of 42.5 dBm, which corresponds to 15 W.

In order to obtain time-resolved dynamics, the upper transition was laser driven in a pulsed way. Therefore the light was shaped by a Leysop Pockels cell<sup>3</sup>. Using this device, pulses of nearly rectangular temporal shapes could be obtained. In the experiment the typical pulse length was 100 ns at a repetition rate of 10 kHz. The rise time was about 1.5 ns and the background level less than 1% of the peak power.

#### 852 nm laser system

The laser light at 852 nm for the  $D_2$ -line density reference was produced by a Toptica Photonics DL pro grating-stabilized, tunable single-mode diode laser. For the density measurement only few micro-watts were needed, and the laser was frequency scanned across the  $6S_{1/2}$ - $6P_{3/2}$  transition.

# 2.2 Pulsed two-photon excitation setup

#### Microcell

The central part of the pulsed experiment was a self-built glass cell with an atomic vapor layer inside with a thickness of  $220\mu m$ . The cell was composed of a rectangular "science chamber", connected to a cylindrical reservoir. Latter was filled with pure cesium and the density could be controlled by the temperature of the reservoir. For heating purposes, an oven was built surrounding the cell.

#### Beam guidance, imaging, and detection

The infrared light beam entered the experiment from the fiber with a waist of 1.33 mm  $(1/e^2 \text{ diameter})$ . An f = 30 mm lens then focused the beam into the cell down to 31 µm.

<sup>&</sup>lt;sup>2</sup>The setup of this laser system is described in detail in [10, ch. 4.2].

<sup>&</sup>lt;sup>3</sup>The setup, operating mode, and performance of the Pockels cell are described in detail in [9].

#### 2 Experimental setup



Figure 2.1: Glass cell of 220 µm containing a u-formed spacer (left part), and a Cs reservoir (right part).

Behind the cell an f = 75 mm lens recollimated the beam to about 3.3 mm. It then was attenuated, and detected by a Thorlabs Det02AFC photodetector with 1.2 GHz bandwidth.

The blue beam came out of the fiber with a beam waist of 1.25 mm, was focused into the cell by an f = 75 mm lens to a waist of  $35 \mu\text{m}$  and recollimated by an f = 30 mmlens to 0.5 mm. In order to image a homogeneous part of the beam, it was focused onto a  $25 \mu\text{m}$  circular high power pinhole at a beam diameter of  $73 \mu\text{m}$  and the transmitted light was focused by a 2 f imaging configuration on the detector. For this purpose a Femto ultra fast photo-receiver HSA-X-S was chosen. This Si detector is internally amplified by a conversion gain of  $2.5 \cdot 10^3 V/W$  and has a bandwidth from 10 kHz to 2 GHz.

Both beams of the two-photon transition scheme passed the cell in opposite direction ("counter-propagating").

As mentioned above the available infrared beam power was 15 W, which corresponds to a peak intensity of  $40 \text{ kW/mm}^2$  at the focal point. The power could be controlled either by the fiber amplifier or by a half wave plate plus high power polarizing beam splitter cube (PBSC), whereas the latter was polarization sensitive and thereby not favorable. The blue beam provided at most about 150 mW, corresponding to  $0.3 \text{ kW/mm}^2$  peak intensity in the cell. Here power control had to be realized by a sequence of polarization optics, in order to hold the power level stable. It consisted of a cascade of a quarter wave plate, half wave plate, PBSC, half wave plate, and PBSC elements.

Both beams had to pass a PBSC before passing the cell, not only for power control (even not in the case of the infrared beam), but also to have a well defined linear



Figure 2.2: Experimental setup used in order to Rydberg excite atomic cesium and to measure the atomic density in the cell.

polarization of the light.

The detected signals were processed by a Teledyne LeCroy Wavesurfer MXs-B oscilloscope, which has a spectral bandwidth of 1 GHz and a specified sampling rate of 10 giga-samples per second<sup>4</sup>.

## 2.3 Reference spectroscopy setup

In order to measure the density in the cell, a third beam passed the cell. This was a 852 nm beam at a power of few micro Watts only, as power broadening had to be avoided in the reference spectrum measurements. In order to be collimated in the cell at a quite large beam diameter, the beam had to be expanded by a telescope, and again focused in order to compensate for the focusing lens before the cell. In this way, the Rabi frequency in the cell was kept in the MHz range. After passing the cell the light signal was detected by a Thorlabs PDA36A-EC amplified photodetector.

<sup>&</sup>lt;sup>4</sup>The sampling rate could be hold only for single bundles of events and not continuously, as the graphic processing blockaded further incoming data processing. Therefore the effective sampling rate, which was provided for continuous sampling, was about 1.5 kilo-samples per second.

# 3 Cesium

## General properties<sup>1</sup>.

group	alkali metal
proton number	55
isotope	Cs-133
relative natural abundance	100%
atomic mass	132.905451931(27) u
nuclear spin	7/2
melting point	$28.5^{\circ}\mathrm{C}$
boiling point	$671^{\circ}\mathrm{C}$

# 3.1 Level scheme

The level energies of atomic cesium are obtained from [16, table 2]. In figure 3.1 the fine structure levels up to D (L = 2) are shown.

#### Quantum defects

Looking at the binding energies or the polarization, many principles of the treatment of the hydrogen atom can be adapted to the alkali atoms like cesium. The reason is that all of them are similar due to the outermost electron. However, the effective potentials for this electron are slightly different. This can be accounted for an effective quantum number

$$n^* = n - \delta_{LJ} \,, \tag{3.1}$$

where  $\delta_{LJ}$  is called quantum defect. It depends on the angular and total angular momentum L, but not on the principal quantum number n. The quantum defects of cesium are listed in table 3.1, whereas the results of several groups differ slightly.

# 3.2 Excitation scheme

In the experiment dealt with in this thesis, an inverted excitation scheme is chosen. The participating levels are  $6S_{1/2}$ ,  $7P_{3/2}$  coupled by light of 455.6556 nm wavelength [16,

<sup>&</sup>lt;sup>1</sup>The values are taken from [15, table 2]



Figure 3.1: Level scheme of Cesium [adapted from 10]. The energy values are taken from [16, table 2].



Figure 3.2: Inverted excitation scheme of Cesium. The left scheme provides a larger probe signal, whereas the right one enables to reach lower Rydberg states. The wavelength regime of the upper transition is limited by the laser system.

table 2], and a certain Rydberg state  $nS_{1/2}$ . The excitation scheme is shown in figure 3.2. The laser system provides a maximal wavelength of about 1075 nm, which corresponds to  $30S_{1/2}$  up to 1040 nm, and is far above the ionization threshold of atomic cesium.

In comparison, conventional two-photon excitation schemes contain a D line as probe transition [e.g. in comparable experiments of 5]. This implies a Rydberg-coupling wavelength that is shorter than the D line wavelength. This fact causes differences, for instance with respect to the Doppler broadening. Looking at the transition strengths, the probe signal is stronger, whereas the coupling to the Rydberg state is weaker<sup>2</sup> than in the inverted scheme. Furthermore the D-line is a closed transition, i.e. there are not any other levels between the driven levels. This avoids dephasing and depopulation effects.

<sup>2</sup>The difference is a factor of about two (S. Hofferberth: private communication).

Table 3.1: Quantum defects of cesium in comparison.

Ref.	$S_{1/2}$	$P_{1/2}$	$P_{3/2}$
[17]	4.06	3.	59
[18]	4.0493527	3.5914856	3.5589599
[19]	$4.05219 \pm 0.02786$	-	

To reach states of lower Rydberg quantum number a modified excitation scheme has to be chosen, in which  $7P_{3/2}$  is replaced by  $7P_{1/2}$ . That means a wavelength of 459.4459 nm [16, table 2] for the lower transition. Here the maximally reachable wavelength corresponds to  $22S_{1/2}$ . However, the transition strength of the probe transition is only half as large as for the  $7P_{3/2}$  state [c.f. e.g. 20].

# 4 Atom-light interaction

## 4.1 Theory

The influence of light on a single atom can be described by the Schrödinger equation. Here the atom-light coupling is included by an interaction term  $H_{int}$  in the Hamiltonian H, i.e.

$$H = H_{\text{atom}} + H_{\text{int}} \,. \tag{4.1}$$

The electric dipole approximation is used, which means that the atomic extension has to be much smaller than the wavelength of the light. The equation of motion for the density operator  $\rho$  is [21, (9)]

$$\frac{d}{dt}\rho = -\frac{i}{\hbar} \left[H,\rho\right] + L\left(\rho\right) \,, \tag{4.2}$$

where the Lindblad operator L accounts for decays and dephasing mechanisms of the levels. Equation (4.2) is often called master equation, or von-Neumann equation.

In the following, the Hamiltonian and the density operator are treated within the Schrödinger picture and in the rotating frame<sup>1</sup>.

The atom is influenced by external light. In turn, the atom influences the light field due to its induced dipole moment

$$\mu = \langle er \rangle_{\varrho} \,. \tag{4.3}$$

The polarization of an atomic ensemble with density  $\rho$  is then  $P = \rho \mu$  [Detailed treatment in e.g. 22].

Using the framework of equation (4.2), many effects can be described, such as Rabi oscillations, the near-resonant a.c. Stark effect, or electromagnetically induced transparency in the case of a three-level system. They all are single atom effects, i.e. they are experienced by single atoms, or, at least, by single-atom-like behaving ensembles.

<sup>&</sup>lt;sup>1</sup>Rotating frame means rotating at light frequency [Details e.g. in 22]; rotating at atomic transition frequency corresponds to the Dirac or interaction picture. Using latter picture is also a way to treat the system as it is done in [21, p. 639, (8)].

#### **Two-level system**

For a two-level atom, the density operator can be represented by a density matrix  $\rho = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix}$ . The two-level Hamiltonian in the rotating frame is

$$H = \frac{\hbar}{2} \begin{pmatrix} 0 & \Omega\\ \Omega & -2\Delta_{12} \end{pmatrix}$$
(4.4)

and the Lindblad operator is

$$L = \Gamma_{21} \begin{pmatrix} \rho_{22} & -\frac{1}{2}\rho_{12} \\ -\frac{1}{2}\rho_{21} & -\rho_{22} \end{pmatrix}, \qquad (4.5)$$

with the population decay rate  $\Gamma_{21}$ . Here the Rabi frequency is defined by

$$\Omega \equiv \frac{dE_0}{\hbar} \,, \tag{4.6}$$

where d is the effective dipole matrix transition element and  $E_0$  is the amplitude of the electric field  $E(t) = E_0 \cos \omega t$ .

#### Three-level system

For three-level systems, there are few different configurations connected to different level couplings and decay paths, whereas we here restrict to the ladder system. For that case the Hamilton operator is [e.g. 5]

$$H = \frac{\hbar}{2} \begin{pmatrix} 0 & \Omega_{12} & 0 \\ \Omega_{12}^* & -2\Delta_{12} & \Omega_{23} \\ 0 & \Omega_{23}^* & -2\left(\Delta_{12} + \Delta_{23}\right) \end{pmatrix}$$
(4.7)

and the Lindblad operator is [5, p. 2]

$$L = \Gamma_{21} \begin{pmatrix} \rho_{22} & -\frac{1}{2}\rho_{12} & 0\\ -\frac{1}{2}\rho_{21} & -\rho_{22} & -\frac{1}{2}\rho_{23}\\ 0 & -\frac{1}{2}\rho_{32} & 0 \end{pmatrix} + \Gamma_{32} \begin{pmatrix} 0 & 0 & -\frac{1}{2}\rho_{13}\\ 0 & \rho_{33} & -\frac{1}{2}\rho_{23}\\ -\frac{1}{2}\rho_{31} & -\frac{1}{2}\rho_{32} & -\rho_{33} \end{pmatrix}$$
(4.8)

In the following, we label the levels in the ladder system ground state, excited state and Rydberg state.

#### Adiabatic elimination

If the lasers are in two-photon resonance and far detuned from the excited level, the three-level system can be approximated by an effective two-level system. The effective Rabi frequency then is [c.f. 23]

$$\Omega_{\rm eff} \approx \frac{\Omega_{12}\Omega_{23}}{2\Delta_{12}} \tag{4.9}$$

and an effective frequency detuning can be defined as

$$\Delta_{\text{eff}} = \Delta_{12} + \Delta_{23} \,, \tag{4.10}$$

which is also called two-photon detuning.

#### Doppler broadening

In atomic ensembles at finite temperature, the movement of the atoms has to be taken into account. Due to the Doppler effect, atoms moving at a certain velocity see a shifted driving light frequency, so that the Doppler shift  $\Delta \omega = kv$  has to be included to the detuning, i.e.  $\Delta \longrightarrow \Delta \pm kv$ . The detuning distribution of the atoms in the ensemble is due to the one-dimensional Maxwell-Boltzmann distribution [e.g. 22, ch. 16]

$$g(\Delta) = \frac{1}{\sqrt{2\pi\sigma}} \exp\left(-\frac{(\Delta \pm kv)^2}{2\sigma^2}\right), \qquad (4.11)$$

where m is the mass of an atom and T the temperature in K. The width of the Doppler broadening is given by

$$\sigma = \frac{\omega_0}{c_0} \sqrt{\frac{k_B T}{2m}} \,. \tag{4.12}$$

So the atomic vapor can be seen as an atomic ensemble of classes of different resonance frequencies, which induces inhomogeneous broadening.

#### Level transitions

Due to spontaneous emission, an atom in a certain level can decay into lower levels. This implies a natural lifetime  $\tau = 1/\Gamma$ . The line width is given by [15, (4)]

$$\Gamma_{J'J} = \frac{\omega_0^3}{3\pi\epsilon_0\hbar c^3} \frac{2J+1}{2J'+1} \left| \langle J \| er \| J' \rangle \right|^2 \,, \tag{4.13}$$

where  $\omega_0$  is the frequency difference and  $|\langle J || er || J' \rangle|$  is the reduced dipole matrix transition element<sup>2</sup> between the levels with total angular momenta J and J'.

# 4.2 Values

#### **Doppler widths**

The Doppler width depends on the wavelength of the exciting radiation. Thereby, the Doppler broadening of both transitions are different.

<sup>&</sup>lt;sup>2</sup>More details about the formalism of dipole matrix transition elements in [10] and [15].

Table 4.1: Dipole matrix transition element  $|\langle J || er || J' \rangle| = |\langle 6S_{1/2} || er || 7P_{3/2} \rangle|$  obtained from different publications in comparison.

	=	-
Ref.	$\left \left<6S_{1/2}\ er\ 7P_{3/2}\right>\right $	
[20, table 10]	$3.151 \cdot 10^{-30} \mathrm{Cm}$	theoretical
[25, table 4]	$3.063 \cdot 10^{-30}  \mathrm{Cm}$	experimental
[26, table 1]	$3.51(0) \cdot 10^{-30} \mathrm{Cm}$	experimental

At a ensemble temperature  $T = 100 \,^{\circ}\text{C}$  the root-mean-square variance (4.12) of the Doppler distribution of the probe transition at wavelength 455 nm is

$$\sigma_{455\,\rm nm} = 2\pi \times 505\,\rm MHz\,,$$
(4.14)

whereas for  $1070\,\mathrm{nm}$  it is

$$\sigma_{1070\,\rm{nm}} = 2\pi \times 215\,\rm{MHz}\,. \tag{4.15}$$

#### Dipole matrix transition elements

The reduced dipole matrix transition element of the  $6S_{1/2}$ - $7P_{3/2}$  transition is obtained from the literature. In table 4.1 different values of  $|\langle J||er||J'\rangle| = (J||er||J')/\sqrt{2J+1}$  for  $J = 6S_{1/2}$  and  $J' = 7P_{3/2}$  are listed. Some of them have been obtained from theoretical calculations, some by experiments.

However, the crucial quantity for Rabi oscillations is not  $|\langle \mathbf{J} || er || \mathbf{J}' \rangle|$ , but the effective dipole matrix transition element  $d_{\text{eff}}$ . This is discussed in the following.

If we drive the transition resonantly, a good way is only to consider the strongest hyper-fine transition. This is  $6S_{1/2} (F = 4)$  to  $7P_{3/2} (F = 5)$ . Whereas the transition starting at the  $6S_{1/2} (F = 3)$  ground state is far off-resonant by 9.2 GHz [15, fig. 3] and thereby not involved, the remaining hyper-fine transitions  $F = 4 \rightarrow F = 3$ , 4 are much weaker due to the Clebsch-Gordan coefficients  $CG(m_F)$ . Thus, they can be neglected in first approximation and the effective dipole matrix transition element is obtained by averaging over the different  $m_F$  transitions:

$$|d_{\text{eff}}|^{2} = \frac{1}{2F' + 1} \sum_{m_{F'}} \left| CG(m_{F}) \left\langle J \| er \| J' \right\rangle \right|^{2}$$
(4.16)

$$= 0.2037 \left| \langle J \| er \| J' \rangle \right|^2 \,. \tag{4.17}$$

This is valid for any  $J = 1/2 \rightarrow J = 3/2$  transition, which is driven by  $\pi$ -polarized light.

 $<sup>^{3}</sup>$ C.f. the different definitions used in [15] and [24].

If the driving field is off-resonant, so that  $\Delta \gg \Gamma$ ,  $\Delta E_{\text{HFS}}$ ,  $\Omega$  is valid, then [15, p. 11, (45)]

$$|d_{\text{eff}}|^2 = \frac{1}{3} \left| \langle J \| er \| J' \rangle \right| \tag{4.18}$$

can be applied, in case of  $\pi$ -polarized light.

For the transition from  $7P_{3/2}$  to the Rydberg S levels the power law<sup>4</sup>

$$\left| \langle 7 \mathcal{P}_{3/2} \| er \| nS_{1/2} \rangle \right| = 9.0822 \cdot n^{*-3/2} \operatorname{ea}_{0} \tag{4.19}$$

is used. It is a fit to the result of atomic orbital calculations. For instance, the  $7P_{3/2} - 32S_{1/2}$  transition has  $|\langle 7P_{3/2} || er || 36S_{1/2} \rangle| = 3.57 \cdot 10^{-31}$  Cm, which is ten times smaller than the 6S - 7P dipole matrix transition element.

### 4.3 Simulations

By solving the master equation (4.2) numerically with usage of the Runge-Kutta method of 4<sup>th</sup> order [details described in 27], we simulated the probe response of the atoms in the vapor cell, where the response is in linear approximation given by the imaginary part of the atomic coherence  $\rho_{21}$ .

Similar to the experiment, the atomic system consisted of a probe transition corresponding to the 6S-7P level transition, and a coupling transition from 7P to nS. The former was driven at a constant Rabi frequency of  $\Omega_p = 2\pi \times 400$ MHz, whereas the latter was switched on at t=0 with a value of  $\Omega_c = 2\pi \times 2$  GHz. The probe transition was driven at a constant frequency, blue-detuned at  $2\pi \times 1.5$  GHz. The coupling transition was scanned across the two-photon resonance.

First, we considered the response of a single atom at rest, in order to obtain a basic insight of the atomic system. The result is shown in figure 4.1. Here the probe response Im ( $\rho_{21}$ ) is depicted over the time and the frequency detuning of the coupling pulse. The two photon resonance is at detuning  $\Delta_{23} = 2\pi \times -1.5$  GHz, while  $\Delta_{23} = 0$  corresponds to the resonance of the 7P - nS transition.

One can observe two-photon Rabi oscillations caused by the pulsed coupling to the Rydberg level. The slowest oscillations occur blue-detuned from the two-photon resonance. The reason for that is a near-resonant a.c. Stark shift which is included in the framework of equation (4.2).

These Rabi oscillations can also be observed in the population dynamics of the ground, excited, and Rydberg level, which are depicted in figure 4.1. The maximal population of the Rydberg level amounts to a fraction of 70%.

<sup>&</sup>lt;sup>4</sup>S. Hofferberth: private communication



Figure 4.1: Simulation of response and population dynamics of the two-photon excitation of a single cesium atom at rest. The Rabi frequencies are  $\Omega_p = 2\pi \times 400 \text{ MHz}$  and  $\Omega_c = 2\pi \times 2 \text{ GHz}$ . The probe transition is driven at  $\Delta_{12} = 2\pi \times 1.5 \text{ GHz}$ .



Figure 4.2: Simulation of response and population dynamics of the two-photon excitation of the cesium vapor at T = 110 °C. The Rabi frequencies are  $\Omega_p = 2\pi \times 400$  MHz and  $\Omega_c = 2\pi \times 2$  GHz. The probe transition is driven at  $\Delta_{12} = 2\pi \times 1.5$  GHz.

However, since the Doppler width is on the order of the probe Rabi frequency (c.f. equations (4.14) and (4.15)) it was reasonable to include Doppler broadening. Instead of only one atom at rest, we solved the equations of motion for many atoms at different velocities. Afterward the different responses were averaged and weighted by the Gaussian distribution (4.11).

The result of the simulations including Doppler broadening are shown in figure 4.2. It can be seen, that the effective Rabi oscillations are slower than the oscillations occurring at the single atom at rest and seem to wash out. The reason for this is the averaging over oscillations at different Rabi frequencies. Besides, note that, in contrast to the single atom simulation, the amplitudes of positive and negative response are not symmetric anymore.

In figure 4.2 you can observe a reduction of the maximal Rydberg population to 40% due to the Doppler broadening. Like the coherences, also the population oscillations wash out.

In these simulations Rydberg interaction was not considered. We shall discuss this in later sections.

# **5** Aspects of optics

Since we manipulate atoms in an optical way, optics plays a crucial role in our experiments. All principles of beam optics are well understood for many decades, and thus are reviewed in many textbooks [e.g. 28].

In this section, merely remarks to focal behavior of a laser beam and to imaging are mentioned.

# 5.1 General remarks

The interface between optical and atomic quantities can be expressed by the Rabi frequency

$$\hbar\Omega = d \cdot E_0 \tag{5.1}$$

while the electric field strength  $E_0$  is of purely optical and the dipole transition strength d is of purely atomic nature. Beside the electric field strength, usual experimental quantities are the intensity

$$I = \frac{1}{2}c_0\epsilon_0 E^2 \tag{5.2}$$

$$=\frac{c_0\epsilon_0\hbar^2}{2d^2}\Omega^2\tag{5.3}$$

and the light power

$$P = \int_{A} I(r)dA = 2\pi \int_{0}^{\infty} I(r)rdr$$
(5.4)

which are just listed for defining purposes.

# 5.2 Diffraction

The propagation of a light beam with a profile  $f_{d=0}(x, y)$  can be described as convolution [e.g. 28]:

$$f_d(x,y) = f_{d=0}(x,y) * h_d(x,y), \qquad (5.5)$$

where x and y are the coordinates perpendicular to the propagation direction, and d is the distance between the beam profiles  $f_{d=0}$  and  $f_d(x, y)$ .  $h_d(x, y)$  is the elemental spherical wave

$$h_d(x,y) \propto \frac{1}{r} \exp\left(-ikr\right)$$
 (5.6)

corresponding the Huygens principle, with  $r = \sqrt{x^2 + y^2 + d^2}$ , and  $k = 2\pi/\lambda$ .

#### Gaussian profile

Often in experiments it is dealt with laser beams which have a Gaussian profile in radial direction. So they can be described by

$$I(r) = I_0 \exp\left(-2\frac{r^2}{\sigma_{\rm w}^2}\right).$$
(5.7)

Here  $\sigma_{\rm w}$  is the half width at half maximum of the beam profile, and  $I_0$  is the center peak intensity in units of W/m<sup>2</sup>.

The Gaussian beam profile is special, as it is an eigensolution of the convolution with an spherical wave, i.e. a Gaussian profile remains Gaussian. If a beam of a Gaussian profile is focused at a minimum waist  $2w_0$  ( $w_0$  is the  $1/e^2$  radius), then a length can be defined in which the intensity is reduced to the half of its maximum value and the waist is  $\sqrt{2} \cdot 2w_0$ . This is the Rayleigh length

$$2z_R = 2 \cdot \frac{\pi w_0^2}{\lambda} \,. \tag{5.8}$$

The Rayleigh length is important if a region of homogeneous peak intensities is desired, which is approximately the case within this length.

#### Rectangular profile

If we consider a rectangular profile, like it happens at diffraction at a narrow aperture, then two limiting cases can be distinguished: the Fresnel limit, in which the diffracted profile is similar to the aperture form; and the Fraunhofer limit, in which the diffraction pattern can be described by the sinc function. Both regimes can be described by using different approximations in (5.5).

In order to quantify the validity of each of both limits, we can use the Fresnel number

$$N_F = \frac{a^2}{\lambda d} \,, \tag{5.9}$$

where a is the radius of the diffraction aperture. If  $N_F \approx 1$ , it will be Fresnel diffraction, if  $N_F \ll 1$ , Fraunhofer diffraction will occur in the distance d from the rectangular beam profile of radius a.





Figure 5.1: Spatial dependence of a Gaussian beam near its focal point at  $w_0 = 15 \,\mu m$ . The plotted range of the optical axis equals the Rayleigh length.

# 5.3 Discussion

If we now design an experimental setup for the excitation atoms in a thin cell by laser radiation, we often have to focus down the laser beams in order to obtain sufficiently large Rabi frequencies. However, we have to avoid spatial inhomogeneities of the intensities and equally of the Rabi frequencies, as they lead to different atomic responses. These finally are averaged on the photodiode and show a wash-out of the signal or an effective dephasing, which we want to avoid. Thereby we have to consider the Rayleigh length: The Rayleigh length has to be larger than the cell length, if we desire a nearly constant Rabi frequency distribution along the optical axis (see figure 5.1). In our case, the blue beam has a waist  $2w_0 = 35 \,\mu\text{m}$  which corresponds to a full Rayleigh length of  $2z_R = 4.0 \,\text{mm}$ , and the infrared beam of  $2w_0 = 31 \,\mu\text{m}$  has  $2z_R = 1.3 \,\text{mm}$ . Both Rayleigh lengths are much larger than the cell length and thereby homogeneity of the intensities along the optical axis is sufficiently fulfilled.

In order to obtain also almost constant intensities perpendicular to the optical axis, we have to cut off the wings of the beam profile. This is achieved by an imaging pinhole, on which the beam is focused after passing the cell. By this only the region among the beam maximum is imaged on the photodetector.

However, we have to consider which parts of the beam in the cell contribute to that part of the beam, which transmits the pinhole. This can be investigated by regarding



Figure 5.2: Spatial dependence of the aperture function, back-imaged from the imaging pinhole into the cesium cell. The image of the aperture has a width of  $10 \,\mu\text{m}$ . The plotted range of the optical axis corresponds to the length of the cell.

the back-imaging of diffraction at the pinhole into the cell. In the center of the cell, the pinhole is imaged perfectly, but before and afterwards Fresnel diffraction occurs, which can be observed in figure 5.2. Note that, while in the real experiment the beam is Gaussian, the idea of back-imaging shows us the contributions of the different spatial coordinates of the beam to the cut-off fraction. To quantify this aspect, we can use the Fresnel number. The aperture, which we imaged in the experiment, had a radius of  $a = 5 \,\mu\text{m}$ . The maximal distance, at which the atomic response influenced the electric field, was  $d = 0.5 \times 220 \,\mu\text{m}$ . A Fresnel number of  $N_F = 0.5$  belonged to the blue beam and a Fresnel number of  $N_F = 0.2$  to the infrared beam. Latter is the important quantity, as the infrared beam causes a modulation of the blue beam; and it is this modulation which is imaged on the a.c. coupled photodiode.

An alternative and more formal treatment of this issue could be provided by the use of point spread functions. These functions describe the evolution of a spatial distribution of spherical waves, which is exactly the case in the experimental situation. The discussion and the Fresnel numbers imply an occurrence of inhomogeneities of the field strengths in the cell. In the performed experiments, these influenced the precision of the measurements and therefore had to be minimized. The inhomogeneity of the Rabi frequency was guessed to be 15% at most.

# 6 Pulsed far off-resonant a.c. Stark shift

Due to the huge power of the infrared laser pulse, a.c. Stark shifts of the  $6S_{1/2}$  and  $7P_{3/2}$  states are supposed to occur, and that in a dynamical and coherent manner. To avoid confusion with Rydberg dynamics, this effect will be considered in the following.

## 6.1 Introduction

External magnetic as well as electric fields have influence on atomic levels, primarily leading to level shifts and splittings dependent on the external field. For static fields these effects are known as Zeeman effect and d.c. Stark effect. Related to these effects is the a.c. Stark effect<sup>1</sup>. It is caused by the interaction between (alternating) electromagnetic radiation leading to shifts, broadenings, and splittings of atomic levels.

Whereas in the former effects only the field strength is decisive for the behavior of the atomic levels, for the latter additionally the frequency of the electromagnetic field has to be compared to the level transition frequencies. If the frequency is close to resonance, the behavior of the levels can be described by the usual two-level optical Bloch equations [29]<sup>2</sup> including the rotating wave approximation. Here a level splitting due to Rabi oscillations occurs, which leads to Mollow triplets [31] or Autler-Townes splitting in three-level systems [32]. Due to the rotating wave approximation, this treatment cannot be correct, if the frequency is far-detuned from the transition frequency.

The latter case can be called the far-off-resonant a.c. Stark shift and is treated in more detail below.

# 6.2 Theory

The far-off-resonant a.c. Stark shift is a single-atom two-level effect, in the sense that a single isolated atom shows this shift and that only two levels are involved<sup>3</sup>.

<sup>&</sup>lt;sup>1</sup>As the electric coupling between the atomic levels overwhelms the magnetic couplings, it is reasonable to only look at the influence of the electric field and to neglect the magnetic contribution. Thereby, for systems of interest there is just one alternating field correspondence to both d.c. Stark and Zeeman effect.

 $<sup>^2\</sup>mathrm{An}$  alternative treatment [30] uses the dressed state picture.

<sup>&</sup>lt;sup>3</sup>Actually this is a simplification of the problem, as the frequency changes by a.c. Stark shifts due to any third levels are neglected. As absolute frequency separations usually are in the range of several



Figure 6.1: If the field is red-detuned, the levels will be forced apart (a). If the field is blue-detuned, they will approach each other (b).

Furthermore, the population of the involved levels does not matter for this effect. It is merely necessary for probing the shift, either by inducing a probe transition, or by spontaneous emission, both under participation of at least one of the shifted levels.

Quantitatively the far-off-resonant a.c. Stark shift can be treated by non-degenerate perturbation theory<sup>4</sup>. On making use of second order perturbation, we obtain

$$\Delta E_2 \equiv -\Delta E_1 = \frac{3\pi c^2 I_{\rm s}}{2\omega_0^3} \left( \frac{\Gamma_{2\leftrightarrow 1}}{E_2 - E_1 - \hbar\omega_{\rm s}} + \frac{\Gamma_{2\leftrightarrow 1}}{E_2 - E_1 + \hbar\omega_{\rm s}} \right) \tag{6.1}$$

following [34, p. 5, (16)] or [35, p. 212, (1)]. Here  $E_1$ ,  $E_2$  are the level energies,  $\Gamma_{2\leftrightarrow 1}$  is the decay rate directly between the levels without decays into intermediate levels;  $I_s$  and  $\omega_s$  are the intensity the frequency of the shifting light. The first order perturbation term, which is linear proportional to the electric field, vanishes for parity reasons.

Equation (6.1) is valid on the assumption that the electric dipole approximation holds, and the detuning is assumed to be large compared to the natural linewidths of the involved levels. Besides these approximations, perturbation theory is restricted to parameter regimes where the perturbation term is small, which is the case for not too large intensity of the shifting beam and a large detuning with respect to the atomic transition. In the case, that the perturbation term is not small, either the forth order perturbation has to be included [35], or it has to be switched to degenerate perturbation theory, if the level is shifted close to any neighboring level.

Perturbation theory virtually provides a standard interpretation: it can be seen as emission of a photon into a virtual state and re-absorption of the same photon, so that the initial and final state are the same. Also the inverted order is possible [35].

THz and shifts are in the GHz regime, this effect is reasonably ignorable in the case of interest.

 $<sup>^4\</sup>mathrm{A}$  dressed state treatment without using the rotating wave approximation can also be performed [c.f. 33].

In a many-level system, like a real atom, the effective level shifts are just sums of shifts between two-level sub-systems (two-level character of a.c. Stark shift), so that we obtain<sup>5</sup>

$$\Delta E_n = \frac{3\pi c^2 I_{\rm s}}{2\omega_0^3} \sum_m \left( \frac{\Gamma_{m\leftrightarrow n}}{E_n - E_m - \hbar\omega_{\rm s}} + \frac{\Gamma_{m\leftrightarrow n}}{E_n - E_m + \hbar\omega_{\rm s}} \right) \tag{6.2}$$

As in a real atomic system there is an infinite number of levels, a truncation of the sum (6.2) is necessary and mostly reasonable, as relevant shifts usually confine to a certain region of levels.

#### Discussion

In the case of interest the transition  $6S_{1/2}$ - $7P_{3/2}$  is driven by a cw field near-resonantly at 455 nm. That causes a steady state population between these two levels. The shift of both levels relatively to each other is mainly generated by the infrared laser, because of its sufficiently high power. As it is pulsed, the lower transition experiences a pulsed detuning with respect to the 455 nm probing light. Therefore, the population as well as the coherence have to oscillate into the new steady state at the different detuning. This process is depicted in figure 6.2.

These oscillations, on the one hand, are disturbing and misdirecting in some sense, that it is searched for coherent population transfer to the Rydberg state mediated by the infrared laser.

However, the a.c. Stark effect provides also this coincidence without involving the Rydberg state. A way to avoid this effect is to perform an off-resonant two-photon excitation to the Rydberg state, which means that the steady state dressing of the intermediate level is small and a change in the detuning has a merely tiny effect and hence only a little change in the signal.

On the other hand, such oscillations can be used to check the capability of the experimental system to image coherent oscillations and to prove the overlap of the two beams performing the two-photon transition. This feature becomes necessary if, as in the case on hand, a possibility to watch two-photon oscillations is not provided. The reason for this is that low-lying Rydberg states are not addressable by the laser system and oscillations to high-lying states are thereby prevented by Rydberg-Rydberg interactions.

Therefore oscillations due to the a.c. Stark effect have been investigated more closely, even though they are not strictly a matter of interest for Rydberg interactions.

 $<sup>^{5}</sup>$  referred to [34, p. 4, (10)] or [35, p. 213, (2)]



Figure 6.2: Principle of the a.c. Stark shift in the experiment: The pulsed high power beam at 1070 nm (a) disturbs the level scheme (b) resulting in a divergence of the levels and Rabi oscillations (c) of the excitation population as well as the coherence between the levels shown in (b). The parameters are adapted from the simulations (c.f. subsection 6.4) and the driving frequency is resonant to the non-shifted transition.



Figure 6.3: Shifts of  $6S_{1/2}$  and  $7P_{3/2}$  due to interaction to the neighboring levels. The dimension of the shifting beam wavelength 1070 nm is depicted in gray. The values are calculated using [20].

# 6.3 Calculations

For the calculation of the shifts of the levels  $6S_{1/2}$  and  $7P_{3/2}$ , the energy differences between the levels have been obtained by [16, table 2], and the dipole transitions were calculated via the transition rates of [20, table 10]. For the shifting beam at 1070 nm the laser power was assumed to be 14 W and the beam waist was set to 31  $\mu$ m, as it was the case in the performed experiments.

Using these parameters, the shift of  $6S_{1/2}$  sums up to  $2\pi \times 407$  MHz. Like depicted in figure 6.3, it acts in a lowering manner, as the shifting laser frequency is red-detuned to the level transition. It is mainly caused by the  $6P_{1/2,3/2}$  levels, all further P levels contribute less than the natural linewidth of the  $6S_{1/2}$ - $7P_{3/2}$  transition and thus are negligible.

For the  $7P_{3/2}$  level the situation is much more complicated. Here there are levels which are lower than  $7P_{3/2}$ , namely  $6S_{1/}$ ,  $7S_{1/2}$ , and both 5D fine structure states. Additionally, the frequencies of the level transitions have to be compared to the frequency of the



Figure 6.4: Time-resolved simulation of the atomic response dependent on the frequency detuning  $\Delta_{12}$ , assuming a pulsed a.c. Stark shift of  $2\pi \times 400$  MHz.

shifting laser. The shifts caused by the S states are quite strong, namely  $2\pi \times 173$  MHz due to  $7S_{1/2}$  and  $2\pi \times 215$  MHz due to  $8S_{1/2}$ . However, as they are shifted in opposite directions, they almost annihilate to a shift of  $2\pi \times 42$  MHz. For the shifts due to the  $D_{5/2}$  the situation is comparable, whereas the shifts of  $D_{3/2}$  are only little beyond the natural linewidth. So the total shift of  $7P_{3/2}$  sum up to about  $2\pi \times 83$  MHz to higher energies.

Both level shifts combined, a relative shift between  $6S_{1/2}$  and  $7P_{3/2}$  of  $\Delta_{a.c.} = 2\pi \times 490$  MHz is calculated. That means, due to the infrared laser both levels diverge about this effective value. As in the present excitation scheme the 455 nm light had a detuning  $\Delta = \Delta_L - \Delta_0 = 2\pi \times 1.5$  GHz, the a.c. Stark shift caused a pulsed detuning of about

$$\Delta' = \Delta_{\rm L} - (\Delta_0 + \Delta_{\rm a.c.})$$
  
=  $\Delta - \Delta_{\rm a.c.}$   
=  $2\pi \times 1.010 \,\text{GHz}$ . (6.3)

## 6.4 Simulation

Simulations of the response of a velocity distributed ensemble of atoms were performed including a time-dependent a.c. Stark shift of  $2\pi \times -400$  MHz and a Rabi frequency

of  $2\pi \times 350$  MHz in a Doppler broadened medium at T = 100 °C. Like in comparable simulations in this thesis, we used a Runge-Kutta algorithm of forth order to solve the optical Bloch equations time-dependently.

In figure 6.4 you can see the result of the simulation. As soon as the pulse enters the cell, the steady state will be perturbed and oscillations occur. The central frequency of these oscillations is at  $\Delta_{12} = 2\pi \times 400$  MHz, and corresponds to the shifted level transition resonance.

## 6.5 Measurements

As previously mentioned, we performed pulsed a.c. Stark shift measurements to obtain a feedback, how well the beams are overlapped and how well the imaging system works. Thereby, the infrared laser is turned far away from any transition to a Rydberg state, usually in the order of few Ångström. The blue cw laser is slowly scanned near-resonantly at the frequency of the  $6S_{1/2}$ - $7S_{3/2}$  transition. As soon as the infrared pulse generates the shifts, the steady state is perturbed by the change of the detuning and oscillations into the new steady state occur. This can be seen in figure 6.5. Here the probe power was 10 mW, which corresponds theoretically to a Rabi frequency of  $350 \text{ MHz}^6$ .

In figure 6.6 a simulation is fitted to a measurement trace at a single frequency detuning. To obtain any congruency, a probe Rabi frequency of  $2\pi \times 250$  MHz is assumed, which is less than the calculated value resulting from the power measurement.

Besides, the effective decoherence of the response of the atomic ensemble cannot be fully explained neither by the theoretically assumed natural linewidth of  $2\pi \times 1.2$  MHz [20, table 11] nor by Doppler-broadening. Thereby additionally a loss ("transient-like dephasing") of  $2\pi \times 20$  MHz is used to reconstruct the decay shape of the coherence signal. This fact suggests the occurrence of effects like transit time broadening [36], which is about  $2\pi \times 3$  MHz, or collisional relaxation and broadening, which is less than  $2\pi \times 10$  MHz.

To explain an enhanced effective decoherence as well as varied Rabi frequencies occurring in the oscillations, three effects can be considered. First, instead of a pure two-level system, the real situation is rather a system of three different excited states. The reason is that it consists of the  $F = 4 \Leftrightarrow F' = 3$ ,  $F = 4 \Leftrightarrow F' = 4$ , and  $F = 4 \Leftrightarrow F' = 5$ transitions, which are slightly frequency-detuned and correspond to different Rabi frequencies. As they overlap mainly due to power broadening, the Rabi oscillation at a single frequency are a superposition of Rabi oscillations at different Rabi frequencies into different hyper-fine states.

Second, each of the  $F \leftrightarrow F'$  lines consists of different  $m_F$  subsystems, each of them with different Clebsch-Gordan coefficients. This again causes an average of dynamics at different Rabi frequencies.

<sup>&</sup>lt;sup>6</sup>On making use of formula (4.17).



Figure 6.5: Time-resolved measurement of the signal, scanned across the resonance. The signal level is given with respect to the steady state signal without infrared pulse. The pulse enters the cell at t=0.



Figure 6.6: Trace of probe signal at  $2\pi \times 480$  MHz detuning. Measurement (blue) and fitted simulation (green) in comparison.

Third, the beam profile and, related to that, the imaging system causes an inhomogeneous distribution of Rabi frequencies. Actually, the set-up is done in such a way, that these effects are minimal. However, it can not be totally excluded.

# 7 Rydberg interactions I: basic principles and weak interaction regime

In order to get a basic insight into the matter of Rydberg interactions, we first use a didactically motivated simplification, considered in this section. By this we can approach the complex matter of many-body Rydberg interaction and obtain several aspects, which will also hold for more complicated systems.

The situation will be a permantently excited single Rydberg atom in a certain state nS and a probe atom. The Rydberg atom shall be in an nS Rydberg state and shall have an active character, since it generates a Rydberg potential. In contrast, the probe atom can be seen as passive receiver or sensor. Consequently in this sense, it shall not influence the active Rydberg atom. The probe atom shall be a quantum-mechanical two-level system<sup>1</sup>, which is excited into the same state nS by an external laser field. The here explained simplified situation of Rydberg interaction is sketched in figure 7.1.

In section 8, we will overcome this ambivalence by treating both atoms symmetrically.



Figure 7.1: Sketch of the situation system of Rydberg interaction.

<sup>&</sup>lt;sup>1</sup>Indeed, we are interested in three-level atoms, but the formalism of a two-level system can be adapted using adiabatic elimination of the excited state.

# 7.1 Theory

Firstly, we will introduce the simplest form of the Rydberg interaction potential: the van der Waals potential. Using this, we obtain the effect of Rydberg blockade. Finally, we expand the van der Waals potential by higher orders in order to have a more accurate description of Rydberg interactions.

#### Van der Waals Rydberg potential

As known from classical electrodynamics, the potential of an induced dipole in the field of a permanent dipole is given by the van der Waals potential<sup>2</sup> [c.f. 37, chapter 4d]

$$V(r) = \frac{C_6}{r^6},$$
(7.1)

where r is the distance between the particles and  $C_6$  is the dispersion or van der Waals coefficient. In our situation, this principle can be adapted, in the sense that the Rydberg atom is the permanent dipole and the probe atom is the induced dipole. As mentioned earlier, both atoms have to be in the nS state. Using quantum-mechanical perturbation theory, the dispersion coefficient was calculated to be [38, table 6]<sup>3</sup>

$$C_6 = n^{*11} \cdot \left( -10.64 + 6.249 \cdot 10^{-1} n^* - 2.330 \cdot 10^{-3} n^{*2} \right) , \qquad (7.2)$$

where  $n^*$  is the effective Rydberg quantum number. In equation (7.2) we observe that for small  $n^*$  the potential is attractive, whereas the sign of the potential changes for increasing  $n^*$  leading to a repulsive potential (see figure 7.3). The change of sign occurs between  $n^* = 18$  and  $n^* = 19$ .

Note, that in the picture used in this section only the probe atom experiences this potential, which leads to a shift of its Rydberg level.

#### Blockade

Now hypothetically suppose an infinitesimal narrow excitation bandwidth, which shall excite our probe atom. The level shift due to equation (7.1) will prohibit any excitation of the probe atom into its Rydberg state; no matter in which distance to the Rydberg atom it is located.

However, there is always a finite excitation bandwidth, given by the effective Rabi frequency [39]

$$\Omega_{\rm eff} \equiv \sqrt{\Omega^2 + (2\Delta)^2} \,, \tag{7.3}$$

<sup>&</sup>lt;sup>2</sup>if one atom shows a dipole moment  $p_1$  caused by fluctuations, an electric dipole field  $E_1 \propto p_1/r^3$  is provoked. This field induces a dipole moment of a neighboring nonpolar atom of  $p_2 \propto E_1 \propto p_1/r^3$ . The resulting interaction potential then is  $\phi \propto -\frac{p_1p_2}{r^3} \propto \frac{1}{r^6}$ .

<sup>&</sup>lt;sup>3</sup>Note the different choice of sign convention compared to [38, (1) and table 6].

where  $\Omega$  is the two-level Rabi frequency and  $\Delta$  is the frequency detuning. The natural linewidth and other line broadenings are neglected.

As a consequence, within a certain distance from the Rydberg atom the probe atom cannot be Rydberg-excited. By comparison of the potential (7.1) and the excitation bandwidth (7.3) we obtain the blockade radius [39]

$$r_{\rm B} \equiv \left(\frac{C_6}{\hbar\Omega_{\rm eff}}\right)^{1/6} \,. \tag{7.4}$$

The principle of Rydberg blockade is illustrated in figure 7.2.

#### Higher Rydberg potential orders

Mathematically spoken, the Rydberg potential (7.1) is a Taylor expansion of the physically real potential, and is obtained by a non-degenerate perturbation approach. Physically spoken, the potential (7.1) stands for the interaction between two dipole moments.

Both perceptions let us consider higher order interaction terms of the Rydberg potential, for instance dipole-quadrupole interaction. It turns out that for nS-nS Rydberg interaction, the potential can be extended to [38, table 6]

$$V(r) = \frac{C_6}{r^6} + \frac{C_8}{r^8} + \frac{C_{10}}{r^{10}} + O\left(\frac{1}{r^{12}}\right), \qquad (7.5)$$

where  $C_6$  is defined by (7.2) and the other dispersion coefficients are

$$C_8 = n^{*15} \cdot (30.19 - 3.777n^* + 1.581 \cdot 10^{-2}n^{*2}) \tag{7.6}$$

$$C_{10} = n^{*22} \cdot \left(-3.190 \cdot 10^{-3} + 5.920 \cdot 10^{-5} n^* - 3.022 \cdot 10^{-7} n^{*2}\right).$$
(7.7)

The higher order terms proportional to  $r^{-8}$  and  $r^{-10}$  become important for increasing Rydberg quantum number and small interatomic distances. In figure 7.3 (a) the potential terms are plotted for different effective Rydberg quantum numbers.

The Taylor-expanded potentials have been calculated using non-degenerate perturbation theory, as the pair state "nS-nS" is taken as an isolated level. As soon as the interaction energy is on the order of the energy spacing to the neighboring pair states, non-degenerate perturbation is not valid any more. Besides, the order truncation of the power series has to be reasonable, so that the influence on the potential is negligible.<sup>4</sup>

<sup>&</sup>lt;sup>4</sup> Also a minimal radius is given by the LeRoy radius, which is the sum of both root-mean-squared distances from their respective cores [40]. However, for lower Rydberg states it is on the order of few tens of nanometers [c.f. 41].



Figure 7.2: Principle of Rydberg blockade: a Rydberg atom (red) generates a van der Waals potential experienced by a laser-driven ground state atom (blue). For distances larger than the blockade radius  $r_{\rm B}$ , a finite excitation bandwidth allows excitation into the Rydberg state; for distances below this radius the excitation is prevented. Note the disparate roles of the participating atoms. Whereas one atom is totally Rydberg excited and generates a Rydberg potential, the other atom only experiences the potential in presence of exciting radiation.



Figure 7.3: Dependence of the Rydberg interaction potential orders (a) on the interatomic distance for various effective Rydberg quantum numbers. Note the change of the  $C_6$  term from attractive to repulsive potential for increasing  $n^*$ . In (b) the nearest neighbor distribution is shown for three orders of densities.

#### Partially excited Rydberg atom

Up to now we have considered a potential-generating Rydberg atom, that is fully excited, which means a population fraction  $f \equiv \rho_{33} = 1$ . If we assume partial excitation, which means f < 1, the potential experienced by the probe atom has to be weighted by this probability factor and we obtain

$$V(r) = f \frac{C_6}{r^6} \,. \tag{7.8}$$

Using the same argument, we obtain the potential energy for the probe atom

$$E_{\rm pot} = f f_{\rm p} \frac{C_6}{r^6} \,, \tag{7.9}$$

where  $f_{\rm p}$  is the excitation fraction of the probe atom.

Therefore, light interaction of the formerly permanently excited Rydberg atom is allowed, since this is connected to a varying excitation fraction. But the interaction is still allowed in one direction only. This slightly modified situation is illustrated in figure 7.4.



Figure 7.4: Sketch of the situation system of Rydberg interaction: Now, atom-light interaction of the formerly permanently excited Rydberg atom is allowed.

# 7.2 Models

The more or less didactic simplification of a fixed Rydberg atom and a probe atom can be used to describe Rydberg interaction in an atomic ensemble. In the following, we will first investigate the distance distribution model, which uses an inhomogeneous ensemble, and subsequently, we will treat the so-called dephasing model, which is related to the former, but mainly empirically motivated.

#### Distance distribution model<sup>5</sup>

The distance between the Rydberg atom and the probe atom can be described by the nearest-neighbor distribution [42, p.86, (671)]

$$w(r) = 4\pi r^2 \rho \exp\left(-\frac{4\pi}{3}r^3\rho\right), \qquad (7.10)$$

where r is the distance between two particles and  $\rho$  is the particle density. w(r)dr is the probability to find two atoms in the distance interval (r, r + dr). Related to w(r), a characteristic length

$$r_0 \equiv (4\pi/3\rho)^{1/3} \tag{7.11}$$

can be defined, which is related to the mean distance  $r_{\rm m} = 0.893 r_0$  and the most probable distance  $r_{\rm p} = 0.874 r_0$ .

The distance distribution for various densities is shown in figure 7.3.

Now let us suppose a large ensemble of probe atoms with density  $\rho$ , which shall be coupled to a Rydberg state nS. Due to their passive character, they shall not interact with each other. Furthermore, an nS-Rydberg atom is placed in the ensemble, which neither interacts with the driving light nor is influenced by the probe atoms. We now can map the distance distribution (7.10) onto a detuning distribution  $w(\Delta_{int})$  of level shifts of the probe atom on making use of the potential (7.1). The transformation of the probability distribution is

$$w(\Delta_{\rm int}) = \frac{dr}{d\Delta_{\rm int}} w(r) , \qquad (7.12)$$

which leads us to

$$w\left(\Delta_{\rm int}\right) = \frac{1}{2} \frac{\Omega_{\rm eff}}{\Delta_{\rm int}^{3/2}} \frac{\varrho}{\varrho_{\rm crit}} \exp\left(-\left(\frac{\Omega_{\rm eff}}{\Delta_{\rm int}}\right)^{1/2} \frac{\varrho}{\varrho_{\rm crit}}\right), \qquad (7.13)$$

where

$$\varrho_{\rm crit} \equiv \left(\frac{4\pi}{3}r_{\rm B}^3\right)^{-1} \tag{7.14}$$

is the critical density.

This model can be improved. For that we allow the formerly fixed Rydberg atom to interact with the external laser field. Now, Rabi oscillations cause a time-dependent Rydberg potential: since  $C_6$  is substituted by  $f(t)C_6$  and hence, the distance distribution (7.13) becomes time dependent. Thereby, the following happens: If the atom is Rydberg excited, the Rydberg population of the probe atoms in its vicinity will be small, and vice

<sup>&</sup>lt;sup>5</sup>This model was developed by B. Huber [6, supplemental material].

versa. From this perspective, the fact that ground state atoms do not screen Rydberg interaction, is included. Furthermore, the Rydberg interaction of the whole ensemble saturates above a certain density.

The nature of this model is closely related to the Doppler broadening effect, at which different velocity classes of moving atoms are connected to different detunings, so that the behavior of the ensemble is just the superposition of every velocity  $class^{6}$ .

Besides, note that the nearest neighbor distribution is valid only for interaction-free particles. Rydberg interactions modify the particle distributions. This is one reason, why the distance distribution model is restricted to the weak interaction limit.

#### Dephasing model

A different model is the dephasing model, which is empirically motivated, since a smearing-out of Rabi oscillations [6, 43] was observed. It can be understood as a mean-field theory. Therefore let us suppose an atom, which feels the Rydberg potentials of all other atoms in the ensemble. All of them cause different shifts, which can be effectively combined into a single dephasing term. Due to the mean-field treatment, all atoms are identical and we obtain a homogeneous ensemble of effectively dephased Rydberg atoms.

Experiments in the weak Rydberg interaction regime have been performed in cold atomic clouds by [43]. In measurements using atomic vapors at room temperature, an effective Rydberg dephasing

$$\Gamma_{33} \sim \Gamma_{\rho_{33}} \rho_{33}$$
 (7.15)

was experimentally observed [6], where

$$\Gamma_{\rho_{33}} \propto \frac{\varrho}{\varrho_{\rm crit}} \propto n^{*\,11/2} \,.$$
 (7.16)

Corresponding to the effective character of this model, the dephasing term is closely related to the root-mean-square variance of the distribution (7.13) of the level shifts .

## 7.3 Measurements

Measurements have been performed with the Rydberg state 32S and in a regime of low vapor densities, where the lower limit of densities was set due to the decreasing signal strength. Thereby, the lowest reasonable density was about  $5 \cdot 10^{12} \text{ cm}^{-3}$ , at which the signal almost vanished. This density corresponded to an effective atomic density of

<sup>&</sup>lt;sup>6</sup>As well as the Doppler broadening effect, this model of Rydberg interaction is collective in the sense that it cannot be described by a single atom, but only by an inhomogeneously broadened ensemble [c.f. 22, ch. 16].



Figure 7.5: Measured signals at densities  $(5, 11)9/16 \times 10^{12} \text{ cm}^{-3}$ : (a) shows frequency cuts at  $\Delta_{23} = 2\pi \times -1 \text{ GHz}$ , where the amplitudes are normalized; in (b) the dependence of the measured signals on the frequency detuning  $\Delta_{23}$  is shown. The coupling pulse enters the cell at time t = 0. The signal is broader at larger density.

 $9/16 \times 5 \cdot 10^{12} \text{ cm}^{-3}$ , as the light interacts only with the F = 4 hyper-fine ground state, whereas the F = 3 state is not involved. The fraction is due to the degeneracy of both levels with respect to its  $m_F$  levels. The powers were 5 mW for the probe and 14 W for the pulsed coupling beam, which corresponded to Rabi frequencies of  $2\pi \times 400$  MHz and  $2\pi \times 2$  GHz.

Assuming  $C_6$  interaction, these parameters corresponded to a blockade radius of  $0.5 \,\mu\text{m}$  and a critical density of  $1.6 \cdot 10^{12} \,\text{cm}^{-3}$ . The densities of the measurements are much larger. However, the critical density is a quantity which corresponds to 100% Rydberg excitation, which is here not the case. Assuming a Rydberg excitation fraction of 10%, the critical density is  $5 \cdot 10^{12} \,\text{cm}^{-3}$ .

Traces at a driving Rabi frequency  $2\pi \times -1$  GHz detuned relative to the 7*P*-32*S* transition are shown in figure 7.5 (a). As it can be observed in figure 7.5 (b), Rabi oscillations could not be observed, also not in comparable measurements at different parameters. Besides Rydberg interactions, different broadening effects give rise to the suppression of Rabi oscillations, like pressure broadening, different ionization mechanisms, plasma formation, etc. They all sum up to  $2\pi \times 20$  MHz at most.

However, Rabi oscillations were observed in atomic Rubidium vapor [5]. The most essential difference between the experiments is the excitation scheme, which corresponds to different probe signal strengths and thereby to different ensemble responses. In our case, the dipole matrix transition element is reduced by a factor of 10 compared to the non-inverted excitation scheme. So the response of the ensemble is diminished by a factor of hundred. The reduction of the ensemble response has to be compensated by increasing the vapor density, which leads to stronger Rydberg interaction between the atoms, but also to other interatomic effects (e.g. collisions).

# 7.4 Simulation

We performed simulations with the two presented models: Cesium atoms in a hot vapor are two-photon excited via a cw-driven  $2\pi \times 1.5$  GHz off-resonant transition from the  $6S_{1/2}$ to  $7P_{3/2}$  and a pulsed coupling transition to 32S. The probe light is  $2\pi \times 1.5$  GHz blue detuned, the coupling pulse is  $2\pi \times -1.0$  GHz red detuned, so that the excitation is close to the two-photon resonance. The Rabi frequencies are  $2\pi \times 400$  MHz and  $2\pi \times 2.0$  GHz for the probe transition and for the coupling transition respectively. Thereby the effective Rabi frequency (see formula (4.9)) is  $2\pi \times 267$  MHz.

#### Distance distribution model

The simulations have been done by solving the optical Bloch equations for different classes of velocities as well as Rydberg interaction shifts.

In figure 7.6 the results with a constant Rydberg shift and a dynamical Rydberg shift are compared. Whereas in former case you can observe a smearing out of the Rabi oscillations as the density increases, in the latter a different behavior occurs. As the Rydberg interaction strength oscillates at the frequency of the Rydberg population, but shifted by a phase of  $\pi$ , the dephasing of the Rabi oscillations only happens in certain parts of the time evolution. In our case, this leads to a suppression of the enhanced transmission (signal > 0).

The simulations merely are in qualitative agreement with the experimental results for small two-photon detunings (figure 7.5).

Especially at higher densities, for which neither the restriction to nearest neighbor interaction (c.f. figure 7.3) nor the choice of the interaction potential is reasonable, simulations and measurements still have similar signal shapes. As a large interaction shift causes a decrease of Rydberg atom density, a saturation of  $C_6 \cdot f(t)$  occurs, which enlarges the regime of qualitative validity of this model.

#### Dephasing model

We also simulated the atomic behavior with use of the dephasing model (section 7.2). The Rydberg dephasing was assumed to be

$$\Gamma_{33} = 2\pi \times 750 \,\mathrm{MHz} \cdot \frac{\varrho}{\varrho_{\mathrm{crit}}} \cdot \rho_{33} \,, \tag{7.17}$$

whereas the constant is a reasonable value so that  $\Gamma_{33}$  remains small compared to the effective Rabi frequency. The critical density is  $\rho_{\rm crit} = 1.5 \cdot 10^{12} \,{\rm cm}^{-3}$  for the case of  $n^* = 28$  and  $\Omega_{\rm eff} = 2\pi \times 250 \,{\rm MHz}$ .

The results of the simulations at the same parameters as in the simulations of the distance distribution model are shown in figure 7.6. By comparing the results with the measurements shown in figure 7.5, we observe quite good agreement for the higher densities. However, it is not clear that the dephasing of the measured signals is totally due to Rydberg interaction.

Additionally we simulated the response of the ensemble dependent on the detuning of the pulsed coupling beam at a particle density of  $9/16 \times 5 \cdot 10^{12}$  cm<sup>-3</sup>, shown in figure 7.8. One can observe a washing-out of the Rabi oscillations in the region of the two-photon resonance. This is not surprising, as it is this resonance which causes a high population of the Rydberg level and so large dephasing.



Figure 7.6: Simulations of the time-evolution of the blue signal from the Dopplerbroadened three-level system using the distance distribution model (inhomogeneous), done for several densities: (a) at a fixed Rydberg potential and (b) at a time-dependent potential; the coupling pulse shape is each depicted in gray. The Rydberg population fractions of the atom, which generates the Rydberg potential, are shown in (c) and (d). The parameters of the two-photon excitation are  $\Omega_{\rm p} = 2\pi \times 400$  MHz,  $\Omega_{\rm c} = 2\pi \times 2$  GHz,  $\Delta_{12} = 2\pi \times 1.5$  GHz,  $\Delta_{23} = 2\pi \times -1$  GHz.



Figure 7.7: Simulations of the time-evolution of the blue signal from the Dopplerbroadened three-level system using the dephasing model (homogeneous), done for several densities. The coupling pulse shape is depicted in gray. The parameters are  $\Omega_{\rm p} = 2\pi \times 400$  MHz,  $\Omega_{\rm c} = 2\pi \times 2$  GHz,  $\Delta_{12} = 2\pi \times 1.5$  GHz,  $\Delta_{23} = 2\pi \times -1$  GHz. The effective dephasing term is described in the text.



Figure 7.8: Simulation of the response and the atomic population dynamics using the dephasing model (homogeneous). The pulse starts at t=0.The parameters are  $\Omega_{\rm p} = 2\pi \times 400 \,\mathrm{MHz}$ ,  $\Omega_{\rm c} = 2\pi \times 2 \,\mathrm{GHz}$ , and  $\Delta_{12} = 2\pi \times 1.5 \,\mathrm{GHz}$ . The atomic density is  $9/16 \times 5 \cdot 10^{12} \,\mathrm{cm}^{-3}$ .

Looking at the populations of the several levels in figure 7.8, a decrease of the Rydberg population simultaneously to an increase of the excited level can be observed. The reason for this is that the coupling from ground to Rydberg state does not involve the excited level, but the dephasing (line broadening) leads to a coupling of the Rydberg population to the excited level. There, it is only weakly coupled the ground state.

# 7.5 Remarks

In this section Rydberg interaction was treated simplified by neglecting quantum mechanical principles such as coherence and identical treatment of the atoms. However, the comparison of measurements and simulations justifies this approach.

The parameters of these measurements are located in the weak interaction regime. This implies weak correlations between the single Rydberg atoms an atomic ensemble and the atomic behavior is only quantitatively changed with respect to vanishing Rydberg interaction. That means, Rydberg interactions merely cause slight modifications of quantities like dephasing [6, 43].

However, if we increase the particle density, the interatomic distances will decrease and we will enter a parameter regime, in which the ensemble behavior is totally different due to strong Rydberg interactions. Then the atomic behavior changes qualitatively compared to a single atom. The atomic behaviors will correlate strongly and interatomic cooperativity cannot be neglected [44].

We will investigate this transition of the atomic ensemble behavior in the following section by looking into theoretical aspects and by measurements at densities, at which strong Rydberg interaction is expected to appear.

# 8 Rydberg interaction II: strong interaction regime

The investigation of the strong Rydberg interaction regime forces us to consider Rydberg interaction in a deeper sense than it is treated in section 7. This will lead us to much more complicate potentials, which we will use to explain the results of the measurements. These might help us to explain the measurement results, which, as we will see in this section, exhibit a qualitatively different behavior.

# 8.1 Theory

If two Rydberg atoms are much closer than the blockade radius, the potentials (7.1) and (7.5) are not valid any more. So far neglected effects have to be included now and we cannot use the description of an active Rydberg atom and a passive probe atom any more.

![](_page_53_Figure_4.jpeg)

Figure 8.1: Rydberg interaction between two identical atoms. Each of them is driven by external radiation and experiences the Rydberg interaction potential of the other atom.

For this reason, we consider two identical atoms, as depicted in figure 8.1. Both are driven by an external field, which excites them into an Rydberg state nS. By this the meanings of the terms "potential" and "blockade" have to be modified: Now, the Rydberg interaction potential describes the potential of the Rydberg levels of two excited Rydberg-exited atoms. So both see the potential caused by each other, and both are identical. If they are now located closely, both strongly interact with each other and blocking further excitations, so that one excitation is coherently distributed over both atoms.

#### Pair states and pair state mixing

A further difference comes into play with the system of two identical atoms. Now they do not have to be excited into the nS-nS state. Also different combinations of Rydberg states are allowed, if it is energetically possible.

In the spectral vicinity of the nS - nS pair state, there are many other pair states  $n_1L_1 - n_2L_2$ . This can be seen as accidental occurrence<sup>1</sup> or as result of the manifold possible combinations  $n_1L_1 - n_2L_2$ . For instance the  $30S_{1/2} - 33D_{3/2}$  pair state lies just  $2\pi \times 6.0$  GHz above the  $32S_{1/2} - 32S_{1/2}$  pair excitation. This gives rise to restrict the validity of the given potential only to small level shifts. As soon as the particles approach each other, the differently shifting pair states will hit each other. Consequently, non-degenerate perturbation theory is not valid any more and degenerate perturbation theory has to be applied [41]. Then avoided crossings occur, which give rise to binding potentials and thereby may lead to molecule formation, so called macrodimers [40].

In figure 8.2 the crossing behavior of two levels is depicted. In the crossing regime, the different states are mixed. In the vicinity of such a crossing region, small admixtures  $\epsilon \ll 1$  of the other pair state level occur. Note that, if one of the pair state levels is dipole-forbidden, it still has an influence on the potential structure as nevertheless the  $\epsilon$  admixture of the allowed level appears.

By the discussion of this simple crossing of two pair state levels, it is clear that the levels structure of Rydberg pair states is extremely complicated. This can be seen in figure 8.3. Here all pair state levels in the vicinity of  $30S_{1/2} - 30S_{1/2}$  are shown. A tremendous number of crossings occur, as the interatomic distance decreases.

The potentials are calculated by D. W. Booth and J. P. Shaffer (University of Oklahoma) via diagonalization of the pair state Hamiltonian including many neighboring pair state levels [similar calculations are described in 41].

#### **Collective behavior**

In the strong Rydberg interaction regime the individual Rydberg atoms show so-called collective behavior. A central aspect of this collectivity is the enhancement of collective

<sup>&</sup>lt;sup>1</sup>Accidental occurrence in the sense, that it is not a degeneracy due to any symmetry.

![](_page_55_Figure_1.jpeg)

Figure 8.2: Crossing behaviors of two spectrally close levels: one level attractive (A) and one repulsive (R). Near the crossing regions  $\epsilon$  mixtures occur. While here a ideal situation of two neighboring levels shall demonstrate the principle of anticrossing of pair state levels, results of calculations in a real system are shown in figure 8.3.

Rabi frequency. That means, the ensemble Rabi frequency  $\Omega_{\text{coll}}$  is enhanced the squareroot of the number N of atoms within one blockade sphere:

$$\Omega_{\rm coll} = \sqrt{N\Omega}\,,\tag{8.1}$$

while  $\Omega$  is the Rabi frequency of a single isolated atom<sup>2</sup>.

This enhancement was observed by T. Pfau and coworkers [3] in an ultracold ensemble of rubidium atoms.

Beyond the enhanced ensemble dynamics, collective behavior of Rydberg excited atomic ensembles opens a wide field of interesting effects, like critical phenomena and phase transitions [45, 46]. A detailed treatment goes beyond the scope of this thesis and connections to the measurements in the here performed experiments are not entirely understood.

# 8.2 Measurements

We performed measurements at densities ranging from  $5 \cdot 10^{13} \text{ cm}^{-3}$  to  $2 \cdot 10^{14} \text{ cm}^{-3}$ . The measurements were performed at a blue power of 3 mW, which corresponds to  $2\pi \times 360 \text{ MHz}$ , while the infrared power was 14 W, which is about  $2\pi \times 2 \text{ GHz}$ .

Assuming  $C_6$  interaction, these parameters corresponded to critical densities of  $1.6 \cdot 10^{12} \text{ cm}^{-3}$  (32S),  $1.0 \cdot 10^{12} \text{ cm}^{-3}$  (34S), an ,  $0.7 \cdot 10^{12} \text{ cm}^{-3}$  (36S). Hence, the densities

 $<sup>^{2}</sup>$ A detailed description is given e.g. in [39].

![](_page_56_Figure_1.jpeg)

Figure 8.3: Potentials of the pair states of cesium in the vicinity of the  $30S_{1/2} - 30S_{1/2}$ pair state, dependent on the interatomic distance. In combination with an excitation bandwidth in the gigahertz regime and sufficient interatomic distances, the shown pair state level structure does principally not allow the application of the concept of blockade. These results are kindly provided by D. W. Booth and J. P. Shaffer (University of Oklahoma).

occurring in this measurements were much larger than the critical densities and strong interaction was expected.

In figure 8.4 (a) frequency scans of the time-dependent signals at different particle densities are shown. The densities varied from  $6.5 \cdot 10^{13} \text{ cm}^{-3}$  to  $2.37 \cdot 10^{14} \text{ cm}^{-3}$ , whereas the abundance factor 9/16 of the driven  $6S_{1/2}(F = 4)$  sublevel has to be taken into account.

The frequency scan measurements can be characterized by three different features: The fast absorption, near one- and two-photon resonance, delayed absorption at few gigahertz red detuning, and the often sharp, kink-like transition between these two regions.

#### **Fast absorption**

In the band of the two-photon and single photon resonance, the signal looked similar to the measurements at lower densities. That means, absorption was enhanced, and that instantaneously to the coupling pulse, due the population transfer into the Rydberg state.

In figure 8.4 (b) the time-dependent signal form is shown. After the sharp absorption feature, a constant signal was obtained rapidly. This new steady state showed enhanced absorption with respect to the steady state signal before the coupling pulse enters the cell (t=0).

In figure 8.6 (a) the signal from a few nano-seconds after the coupling pulse entered the cell is shown. For both low densities a absorption peak at the shifted two-photon resonance is visible. Red detuned to that line was a feature which caused a sharp absorption kink. This feature became a dominant feature as the density increased. At large densities, the lineshape looks similar to bistable lines, caused by nonlinearities.

#### **Delayed** absorption

At a certain frequency the instantaneous absorption vanishes. Instead, the absorption occurred delayed by few nanoseconds, depending on the frequency detuning and on the density. In figure 8.5 (a) the time-dependent signals at different densities are compared, each at  $2\pi \times -3400$  MHz and coupling to 32S. The absorption becomes larger, faster, and shorter in time for increasing density. Besides, oscillations seem to occur.

In figure 8.5 (b) the behavior dependent on the density and the Rydberg quantum number is analyzed. A clear dependence on the Rydberg state occurs. This gives rise for scaling the densities by the critical density assuming Rydberg van der Waals interaction. The result is shown in figure 8.5 (c). However, this seems not to be the correct critical density, which is not surprising in the face of the complicated structure of the molecular potentials.

We fitted the data assuming a power law of the form  $f(\varrho) = A \varrho^{\alpha}$  and obtained:

![](_page_58_Figure_1.jpeg)

Figure 8.4: (a): Frequency detuning scans at various densities. The Rydberg-coupling pulse enters the cell at t = 0. The density dependence of the kinks and the delayed absorptions is clearly visible. (b): Time-dependent signal at various densities. The coupling frequency is detuned by  $2\pi \times -1.5$  GHz to the one-photon resonance. The signal strength is normalized. The time offset is with respect to the coupling pulse.

![](_page_59_Figure_1.jpeg)

Figure 8.5: Time-dependent signal at various densities (a). The coupling frequency is detuned by  $2\pi \times -3400$  MHz to the one-photon resonance. The signal strength is normalized. In (b) the delays are shown dependent on the density in a logarithmic scale, comparing different Rydberg states. The dependence on the Rydberg state is accounted by scale the densities by the critical density of  $C_6$  interaction (c).

nS	A $\left(\frac{2\pi \times \text{GHz}}{(10^{12}\text{cm}^{-3})^{\alpha}}\right)$	$\alpha$
32S	$10.8 \pm 6.4$	$-1.29 \pm 0.14$
34S	$3.4 \pm 1.5$	$-1.10 \pm 0.10$
36S	$0.96 \pm 0.52$	$-0.95 \pm 0.13$

As the processes occurring in these measurements are not understood, there is no theoretical description available, to which the quite clear results can be compared.

#### Kink-like transition

The dependency of the frequency of the instantaneous red edge was investigated. In figure 8.6 (a) signals shortly ( $\approx 1 \text{ ns}$ ) after the switch-on of the coupling pulse are shown. We carried out such measurements at many different densities, as well as different Rydberg states. The results are shown in figure 8.6 (b). The frequencies of the kinks depend on the involved Rydberg state. However, a scaling behavior cannot be observed (figure 8.6 (c))

Additionally, the dependence of the kink frequency on the probe power was investigated, where the Rydberg state was 32S. The results are shown in figure 8.7.

We fitted the data assuming a power law of the form  $f(\varrho) = A \varrho^{\beta} + C$  and obtained:

nS A 
$$\left(\frac{2\pi \times \text{MHz}}{(10^{12} \text{ cm}^{-3})^{\beta}}\right)$$
  $\beta$  C  $(2\pi \times \text{MHz})$   
32S 8.6  $\cdot 10^{-3} \pm 3.2 \cdot 10^{-3}$  2.06  $\pm 0.17$  -1634  $\pm 65$ 

The fit results in a quadratic Rabi frequency dependence of the kink frequency.

The uncertainties in the performed measurements were given by the temperature in the cell, which was obtained by absorption spectroscopy, and power deviations during the measurement procedure. Especially the former was not accurate enough and might be improved.

## 8.3 Remarks

Several effects may cause the behavior which we observed.

The delayed absorption feature may appear due to small admixtures ( $\epsilon$ ) of the nSnS pair states to neighboring states. The excitation probability would decrease as the coupling frequency departs from nS-nS resonance. This could explain why the absorption delay increases in that case. However, due to the complexity of the level structures, the establishment of a connection between the experimental results and the pair state potentials in figure 8.3 is rather complicate and is beyond the scope of this work.

![](_page_61_Figure_1.jpeg)

Figure 8.6: Frequencies of the red edge of the signals in the probe transition at various densities for the 32S Rydberg state (a); the cuts are taken shortly after the arrival of the coupling pulse. The density dependence of the sharp edge at three different Rydberg quantum states are compared (b). In (c) the densities are scaled by the critical density of  $C_6$  interaction. Tendencies in the density and Rydberg state dependency can be observed; however, any clear scaling behavior is not visible.

![](_page_62_Figure_1.jpeg)

Figure 8.7: Signal cuts shortly after the entering of the pulse into the cell at different probe powers (a) (similar to the measurements shown in figure 8.6); evaluation of the frequency of the red edge dependent on the probe power (b). A quadratic dependence of the red edge frequency on the probe power is evident. The origin of this is not clear, expecially whether any Rydberg-specific nature causes this scaling. Further measurements investigating the Rydberg state dependence have to be done.

The reason for the kink structure and the fast absorption signal may be critical behavior and a phase transition into crystal-like structure of the Rydberg system.

These and additional aspects are under current theoretical as well as experimental investigation.

# 9 Conclusion and Outlook

The scope of this thesis spanned the experimental set up as well as first measurements of pulsed Rydberg excitations in thermal cesium vapors.

The purpose of this experiment was to provide an alternative solution to the experiment using rubidium, since the differing technical features were supposed to provide new experimental opportunities. It turned out that the cesium experiment provides rather a complement than a substitution of the established rubidium experiment.

The first step of this work was to combine several technical advices like the Pockels cell and the signal detection at nanosecond timescales. Moreover, the setup of the twophoton excitation was designed and realized. In order to reach sufficiently fast dynamics, we were forced to work with optics at few micrometers, which has been taking a long time until adequate control was achieved.

Besides, due to the inverted excitation scheme, the a.c. Stark effect had to be taken into account.

This setup enabled us to investigate Rydberg interactions. We performed measurements in the weak interaction regime. These were accompanied by simulations using mean field and ensemble models, which have been set up and improved during this thesis, so that qualitative agreement could be obtained.

By increasing the vapor densities we could enter the regime of strong Rydberg interactions and consequently we started to explore qualitatively new effects due to the strength of interaction between the Rydberg atoms. Finally, we characterized these features systematically, which are not fully understood theoretically up to this date.

This work, for the one hand, might contribute to future understanding of many-body interaction mechanisms, especially of quantum phase transitions into strongly correlated regime. On the other hand, the investigated Rydberg interactions are a crucial component for the setup of a single photon source, complementary to the closely related experiment of pulsed Rydberg excitation of thermal rubidium vapors.

Further investigations have to be done in the regime of strong Rydberg interactions and towards the manifestation of Rydberg crystals, which might be present in this system. Connected to this, a deeper understanding of processes occurring in our system needs to be developed.

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# Zusammenfassung

Diese Arbeit behandelt die zeitaufgelöste kohärente Anregungsdynamik von Rydbergzuständen in thermischen Gasen. Der erste Schritt dieser Arbeit war der Aufbau des Experiments. In anschließenden Messungen wurde zeitaufgelöst die Anregungsdynamik gemessen. Dabei konnten Effekte beobachtet werden, die starke Rydbergwechselwirkungen zwischen den Atomen vermuten lassen. Es wurde ein kritschen Verhalten beobachtet, das weiterführenden Untersuchungen unterzogen werden muss und den Rahmen dieser Arbeit sprengen würde.

Die Messungen wurden durch theoretische Analysen dieses Systems begleitet, wobei verschiedene Modelle herangezogen wurden, die in qualitativer Übereinstimmung mit Messungen in Parameterbereichen schwacher Rydbergwechselwirkung stehen. Das Ensembleverhalten bei starker Rydbergwechselwirkung ist bislang theoretisch noch nicht voll erschlossen. Jedoch werden in dieser Arbeit einige Aspekte und somit Ansätze zur Lösung offener Fragen angeführt.

Die Ergebnisse im Rahmen dieser Arbeit aufgenommener Messungen könnten einen wichtigen Beitrag in der theoretischen Weiterführung von kollektiven Effekten in Rydbergsystemen sowie zum Aufbau einer Einzelphotonenquelle leisten.

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