# Superradiance of Rydberg atoms in thermal vapour cells

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Hauptberichter: Prof. Dr. Tilman Pfau Mitberichter: Dr. Jürgen Weis

> Universität Stuttgart 5. Physikalisches Institut

## Abstract

In this thesis the investigation of superradiance in a thermal vapour of caesium atoms is reported. The atoms are continuously pumped to the Rydberg state with principal quantum number n = 32 via a two-photon excitation scheme. The superradiance is indirectly observed via the investigation of the optical fluorescence. It was observed that superradiance exhibits a threshold behaviour under continuous pumping. The threshold behaviour was studied for varying ground state density, Rabi frequency and excitation volume. It was shown that under certain conditions it is possible to influence the superradiant dynamics via the variation of the excitation volume. In order to describe the measurements a simple theoretical model was developed.

## Zusammenfassung

In dieser Arbeit wird die Superradianz in einem Ensemble thermischer Zäsiumatome, die durch kontinuierliches Pumpen in den Rydbergzustand angeregt werden, beobachtet. Dabei werden die Atome über eine Zwei-Photonen-Anregung in den Rydbergzustand mit der Hauptquantenzahl n = 32 gebracht und die Fluoreszenz detektiert. Anhand von Änderungen in der spektralen Zusammensetzung des Fluoreszenzlichtes wird auf die Eigenschaften der Superradianz geschlossen. Unter kontinuierlichem Pumpen der Atome zeigt die Superradianz ein Schwellenverhalten. Das Schwellenverhalten wurde in Abhängigkeit von der Grundzustandsdichte, der Rabifrequenz und des Anregungsvolumens untersucht. Es zeigte sich, dass es unter bestimmten Voraussetzungen möglich ist die Superradianz über die Änderung des Anregungsvolumen zu beeinflussen.

## Declaration

I hereby declare that this submission is my own work and that, to the best of my knowledge and belief, it contains no material previously published or written by another person, except where due acknowledgment has been made in the text.

Margarita Reschke Stuttgart, September 29, 2014

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# Chapter 1 Introduction

A wide range of the research activities in atomic physics deals with two fundamental circumstances. The first is the interaction of atoms with electric, magnetic and electromagnetic fields. In particular lasers, as a coherent electromagnetic field, are used to manipulate atoms in a controlled way making use of the atom-light interaction. The second interaction is the interaction that the atoms experience among themselves, referred to as atom-atom interaction. A possibility to investigate new effects owing to atom-atom or atom-light interaction is to investigate their influence upon already known optical properties of atoms. For example, broadening effects can be studied via the linewidth of the signal in an electromagnetically induced transparency (EIT) experiment [1]. In the last decades particularly Rydberg atoms have experienced an enormous growth of interest in atomic physics. Rydberg atoms are atoms, where at least one electron is excited to highly excited state  $(n \gg 1)$ . Rydberg atoms experience strong inter-atomic interaction strengths arising from their large electric dipole moment. Due to these interaction properties Rydberg atoms are expected to be ideal for applications in quantum information processing [2]. However, because of their large electric dipole moments Rydberg atoms strongly interact with long-wave radiation that is resonant to transitions between Rydberg states. This can result in a coherent emission of radiation by the atomic ensemble. Characteristically, the optical response is a short and intense pulse on time scales much shorter than the natural lifetime. Such an enhanced spontaneous emission is called superradiance [3]. The concept of superradiance was first introduced theoretically by Dicke in 1954 [4]. After the development of pulsed dye-laser systems there has been extensive experimental studies on this effect. The first superradiant pulses were observed in 1973 in HF vapour [5] followed by the observation of superradiant cascading effects of alkali atoms excited to Rydberg states [6, 7, 8]. In the case of superradiance the coherent emission of radiation is the result of a phase-locking of the microscopic dipoles, which interact through a light field. Laser amplifiers make use of the collective phase. A superradiant medium is in principle a mirror-less laser with the difference that the threshold for amplification is far smaller than for conventional lasers. In this context it has been demonstrated by Gross *et al.* that superradiance can be exploited to develop coherent microwave sources [8]. They demonstrated that it is possible to tune superradiance by filtering the microwave transi-

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Figure 1.1: Schematic illustration of a quantum module based on Rydberg atoms in thermal vapour cells.

tion with a cavity. In this experiment an atomic beam was excited by two pulsed lasers when it passed the cavity. With the development of cw-laser systems it is possible to excite atoms under continuous pumping with high powers. With regard to future applications cw-systems could facilitate the realization of continuous microwave sources. Recently, the first superradiant laser has been realized with cold Rubidium atoms [9]. A big problem of such systems with cold atoms, however, is that they require a enormous amount of space. This is a huge disadvantage regarding future applications in quantum information processing or atomic clocks, since the integration potential of such systems is limited. In contrast, thermal vapour cells of alkali atoms offer a promising chance towards scalability of quantum systems. In the last years a lot has been done in this field [10, 11, 12, 13]. Lately, superradiance has also been observed in a cw-pumped vapour cell of caesium atoms [14]. This discovery motivates the development of quantum networks (fig. 1.1), which are based on Rydberg atoms and are controlled via



Figure 1.2: Schematic illustration of a coherent microwave source based Rydberg atoms in a vapour cell.

customized superradiant properties. Furthermore, the superradiant properties could be exploited to design coherent microwave sources in frequency regimes, which are still not accessible with today's commercially available coherent microwave sources. A possible design is illustrated in fig. 1.2. However, only a few investigations have been done on superradiant properties in cw-pumped thermal vapour. The first focus of this thesis to demonstrate that superradiance can be observed in thermal cw-pumped caesium vapour. This will provide the basis for exploration of superradiant properties. The second focus is then to investigate possibilities to manipulate the superradiance, i.e. to suppress or amplify its influences. As superradiance strongly shortens the life time of Rydberg states it is, for example, a limiting factor for the observation of Rabi oscillations to Rydberg states [15]. Optical coherence, however, is a basic requirement for quantum information processing based on atomic systems [16].

This thesis consists of two three parts. The first chapter introduces the fundamental theory, which is required to understand and interpret the measured data. In section 1 the basic principles of the atom-light interaction are presented. Section 2 introduces a theoretical description of superradiance and its characteristics. The final section 3 presents the numeric simulation, that is used to simulate the superradiance.

The second chapter is devoted to the experimental setup. In this experiment, the superradiance is observed in thermal caesium vapour via fluorescence spectroscopy. The excitation scheme and the excitation setup, which was used to perform the measurements, are presented in section 1 to 3. In section 4 a detailed overview of the fluorescence spectroscopy setup and the relevant constituents is given.

The third chapter presents the results that were obtained during this thesis. The first section outlines the observation procedure and describes how the superradiance manifests itself in the atoms fluorescence. In the following second section the measured data is analysed, discussed and compared to the simulation results. Finally, in the third section a discussion of the simulation model is presented.

Chapter 1. Introduction

# Chapter 2

# Theoretical foundation

This chapter outlines the fundamental theory required in this thesis. The first part of this chapter deals with the atom-light interaction, where basic principles are introduced on the basis of the simplified cases of a two-level and three-level atom. In the second part the theory on superradiance, which is the optical effect that is the major subject of the studies in this thesis. The last part of this chapter is devoted to the numeric simulation, that was developed during this thesis with the purpose to explain experimental results.

### 2.1 Atom-light interaction

In this section the theory of atom-light interaction is presented in the quantum mechanical description under use of the density matrix formalism. In the quantum mechanical approach a system of an atom interacting with a light field is described by the total Hamiltonian

$$\hat{H}_{\text{tot}} = \hat{H}_{\text{atom}} + \hat{H}_{\text{light}} + \hat{H}_{\text{int}}$$
(2.1)

where, under definition of  $\omega_{\rm L}$  := laser frequency,  $\hat{a}^{\dagger}, \hat{a}$  := creation and annihilation operators of the light field,  $\hbar \omega_i$  := energies and  $|i\rangle$  := eigenstates of the atom,

$$\hat{H}_{\rm atom} = \sum_{i} \hbar \omega_i |i\rangle \langle i| \tag{2.2}$$

$$\hat{H}_{\text{light}} = \hbar \omega_{\text{L}} \left( \hat{a}^{\dagger} \hat{a} + \frac{1}{2} \right)$$
(2.3)

are the Hamiltonians for the bare atom and light field respectively.  $\hat{H}_{\text{int}}$  represents the Hamiltonian accounting for the interaction between atom and light. The quantum state of an atom can be written as a superposition of eigenstates  $|\psi\rangle = \sum_i c_i |i\rangle$ ,  $c_i \in C$ , that motivates the definition of the density matrix operator

$$\hat{\rho} = |\psi\rangle\langle\psi| = \begin{pmatrix} |c_1|^2 & c_1c_2^* & \cdots \\ c_1^*c_2 & |c_2|^2 & \\ \vdots & \ddots \end{pmatrix} = \begin{pmatrix} \rho_{11} & \rho_{12} & \cdots \\ \rho_{21} & \rho_{22} & \\ \vdots & \ddots \end{pmatrix}.$$
(2.4)

Diagonal elements  $\rho_{ii}$  represent the population of level *i*, while the off-diagonal elements  $\rho_{ij}$  contain the coherence between level *i* and *j*. The time evolution of the density matrix operator is described by the LIOUVILLE-VON-NEUMANN equation

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [\hat{H}_{\text{tot}}(t), \rho(t)] + \mathcal{L}(\rho(t)).$$
(2.5)

The LINDBLAD opterator  $\hat{\mathcal{L}}$  accounts for the decay processes in the atom.

### 2.1.1 Two-level atom

Although the two-level system is a very simplified model compared to the real nature of an atom it's well suited as a start to illustrate the basic principles of atom-light interaction. A system consisting of ground and excited state

$$|1\rangle \equiv \begin{pmatrix} 1\\ 0 \end{pmatrix} \text{ and } |2\rangle \equiv \begin{pmatrix} 0\\ 1 \end{pmatrix}, \quad (2.6)$$

which are separated energetically by  $\hbar\omega_0$  is considered (see figure (2.1)). An atom excited to  $|2\rangle$  decays back to the ground state with the rate  $\Gamma$ . The difference of the laser frequency  $\omega_{\rm L}$  compared to the transition frequency  $\omega_0$  is characterised by the detuning  $\delta = \omega_{\rm L} - \omega_0$ .



Figure 2.1: Scheme of two-level system. With the laser frequency, transitions frequency and decay rate.

For a two-level system the interaction Hamiltonian exhibits the form

$$\hat{H}_{\rm int} = -\vec{d} \cdot \vec{E} = \frac{\hbar\Omega}{2} \left( |2\rangle\langle 1| \cdot \hat{a} + |1\rangle\langle 2| \cdot \hat{a}^{\dagger} \right) \quad \text{with} \qquad \Omega = \frac{-d_{12}E_0}{\hbar}. \tag{2.7}$$

The unknown arising quantity  $\Omega$  is the Rabi frequency that is composed of the magnitude of the electric field  $E_0$  and the dipole matrix element defined as  $d_{12} = |\langle 1 | \vec{d} \vec{\epsilon} | 2 \rangle|$  with  $\vec{\epsilon}$  being the polarisation vector of the electric field and the electric dipole moment  $\vec{d}$ . In the product basis  $\{|1,n\rangle, |2,n-1\rangle\}$  with the zero level  $n\hbar\omega_{\rm L} \equiv 0$ , the total Hamiltonian can be written as

$$\hat{H}_{\text{tot}} = \frac{\hbar}{2} \begin{pmatrix} 0 & \Omega\\ \Omega^* & 2\delta \end{pmatrix}.$$
(2.8)

Introducing additionally the density matrix and LINDBLAD operator

$$\hat{\rho} = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix}, \qquad \qquad \mathcal{L}(\rho) = \begin{pmatrix} \rho_{22} & -\frac{\rho_{12}}{2} \\ -\frac{\rho_{21}}{2} & -\rho_{22} \end{pmatrix}$$
(2.9)

for the discussed two-level system one obtains with the help of equation (2.5) the differential equations for the time evolution of each density matrix component

$$\dot{\rho_{11}} = \Gamma \rho_{22} + \frac{i\Omega}{2} (\rho_{12} - \rho_{21})$$

$$\dot{\rho_{12}} = -\left(\frac{\Gamma}{2} + i\delta\right) \rho_{12} + \frac{i\Omega}{2} (\rho_{11} - \rho_{22})$$

$$\dot{\rho_{21}} = -\left(\frac{\Gamma}{2} - i\delta\right) \rho_{12} + \frac{i\Omega}{2} (\rho_{22} - \rho_{11})$$

$$\dot{\rho_{22}} = -\Gamma \rho_{22} + \frac{i\Omega}{2} (\rho_{21} - \rho_{12})$$
(2.10)

also known as the Optical Bloch equations (OBE). As the atoms are examined only under continuous pumping in this thesis, only the steady state solution of the differential equations, i.e.  $\dot{\rho} \stackrel{!}{=} 0$ , is of interest. With the condition  $\rho_{11} + \rho_{22} = 1$  the steady state solutions are

$$\rho_{11} = \frac{\Omega^2 + 4\delta^2 + \Gamma^2}{2\Omega^2 + 4\delta^2 + \Gamma^2} \\
\rho_{12} = \frac{\Omega(i\Gamma + 2\delta)}{2\Omega^2 + 4\delta^2 + \Gamma^2} \\
\rho_{21} = \frac{\Omega(i\Gamma - 2\delta)}{2\Omega^2 + 4\delta^2 + \Gamma^2} \\
\rho_{22} = \frac{\Omega^2}{2\Omega^2 + 4\delta^2 + \Gamma^2}.$$
(2.11)

#### **Optical properties**

The response of an atom to a light field E is characterized by the susceptibility  $\chi$  which in case of a weak electric field is related to the polarisation by

$$P = \epsilon_0 \chi E. \tag{2.12}$$

The real part of  $\chi$  corresponds to the dispersion, while the imaginary part is related to the absorption properties. At the same time the polarisation P of an atom is given by the product of the expectation value of the dipole moment  $\langle d \rangle$  and the atomic density n

$$P = n\langle d \rangle = n \cdot (d_{12}\rho_{12}^* + d_{12}^*\rho_{12}).$$
(2.13)

Consequently, the information on optical properties is contained in the density matrix element  $\rho_{12}$  and therefore yields

$$\chi = nd_{12} \frac{\Omega(i\Gamma + 2\delta)}{2\Omega^2 + 4\delta^2 + \Gamma^2}.$$
(2.14)

For small Rabi frequencies  $\Omega$  the imaginary part of (2.14), which is related to the absorption of the light field, is a Lorentzian function with half-value width  $\Gamma$ , the natural linewidth with respect to varying the detuning  $\delta$ .

### 2.1.2 Three-level atom

Similar to two-level approach a theoretical model for a three-level system can be constructed. This is insofar useful as one additional level gives rise to new phenomena which cannot be explained by the two-level theoretical model. In the experiment the excitation to a Rydberg level is performed via an intermediate state. Therefore, the modifications that arise when involving a further coupled transition, are shortly mentioned in the following. The three level scheme considered is presented in figure 2.2. The system is represented by the three states

$$|1\rangle \equiv \begin{pmatrix} 1\\0\\0 \end{pmatrix} \quad |2\rangle \equiv \begin{pmatrix} 0\\1\\0 \end{pmatrix} \quad |3\rangle \equiv \begin{pmatrix} 0\\0\\1 \end{pmatrix} \quad (2.1)$$

and the density matrix

5) Figure 2.2: Scheme of threelevel ladder system coupled to two light fields.

$$\begin{pmatrix} \rho_{11} & \rho_{12} & \rho_{13} \\ \rho_{21} & \rho_{22} & \rho_{23} \\ \rho_{31} & \rho_{32} & \rho_{33} \end{pmatrix} .$$
 (2.16)

The coupling of the two transitions are characterised by the Rabi frequencies  $\Omega_{12} = \frac{-d_{12}E_{0,1}}{\hbar}$  and  $\Omega_{23} = \frac{-d_{23}E_{0,2}}{\hbar}$ . The total Hamiltonian and the LINDBLAD operator are of the form

$$H_{\text{tot}} = \hbar \begin{pmatrix} 0 & \Omega_{12}/2 & 0\\ \Omega_{12}^*/2 & -\delta_{12} & \Omega_{23}/2\\ 0 & \Omega_{23}^*/2 & -\delta_{12} - \delta_{23} \end{pmatrix}$$
(2.17)

and

$$\mathcal{L}(\rho) = \begin{pmatrix} \Gamma_{12}\rho_{22} + \Gamma_{31}\rho_{33} & -\frac{1}{2}\Gamma_{21}\rho_{12} & -\frac{1}{2}\Gamma_{32}\rho_{13} \\ -\frac{1}{2}\Gamma_{21}\rho_{21} & -\Gamma_{12}\rho_{22} + \Gamma_{32}\rho_{33} & -\frac{1}{2}(\Gamma_{32}\Gamma_{21})\rho_{23} \\ -\frac{1}{2}\Gamma_{32}\rho_{31} & -\frac{1}{2}(\Gamma_{32} + \Gamma_{21})\rho_{32} & -\Gamma_{32}\rho_{33} \end{pmatrix}$$
(2.18)

and lead with the LIOUVILLE-VON NEUMANN equation (2.5) to the differential equations for the time evolution of the density matrix entries:

$$\dot{\rho_{11}} = \Gamma_{21}\rho_{22} - \operatorname{Im}(\rho_{12}\Omega_{12}^{*}) \\
\dot{\rho_{12}} = \left(-\frac{\Gamma_{21}}{2} - i\delta_{12}\right)\rho_{12} + \frac{i}{2}\left(-(\rho_{22} - \rho_{11})\Omega_{12} + \rho_{13}\Omega_{23}^{*}\right) \\
\dot{\rho_{13}} = \left(-\frac{\Gamma_{32}}{2} - i(\delta_{12} + \delta_{23})\right)\rho_{13} + \frac{i}{2}\left(\rho_{12}\Omega_{23} - \rho_{23}\Omega_{12}\right) \\
\dot{\rho_{22}} = -\Gamma_{21}\rho_{22} + \Gamma_{32}\rho_{33} + \operatorname{Im}(\rho_{12}\Omega_{12}^{*}) + \operatorname{Im}(\rho_{23}\Omega_{23}^{*}) \\
\dot{\rho_{23}} = \left(-\frac{\Gamma_{21}}{2} - \frac{\Gamma_{32}}{2} - i\delta_{23}\right)\rho_{23} + \frac{i}{2}\left(-(\rho_{33} - \rho_{22})\Omega_{23} - \rho_{13}\Omega_{12}^{*}\right) \\
\dot{\rho_{33}} = \Gamma_{32}\rho_{33} - \operatorname{Im}(\rho_{23}\Omega_{23}^{*}).$$
(2.19)

The amount of fluorescence emitted by an ensemble of atoms with the atomic density  $\mathcal{N}$  is proportional to the Rydberg population  $\rho_{33}$ :

$$\mathcal{F} \propto \Gamma_{32} \rho_{33} \mathcal{N} \tag{2.20}$$

#### Electromagnetically induced transparency

In the experiment presented in this thesis the optical properties of three-level systems are studied by observing the absorption of the light coupling the transition  $|1\rangle \rightarrow |2\rangle$ , which is therefore referred to as the probe field. The light field of the transition  $|2\rangle \rightarrow$  $|3\rangle$  on the other hand is called the coupling field. One of the new phenomena that arise in a three-level system with two light fields is namely the electromagnetically induced transparency (EIT). Hereby an atomic transition (probe field) becomes transparent in the vicinity of its resonance, i.e. the absorption is not a Lorentzian any more as for a two level atom. The width of the transparency window depends on the strength of the coupling field. A detailed discussion on this effect and various applications can be found in [17].

### 2.2 Collective atom-light interaction

The model introduced so far treats the atoms in the single-atom frame, i.e. under the assumption of each atom interacting with the light field individually. The effects created by the single atoms are simply added together when considering a system of many atoms. In contrast the theory presented in the following, however, deals with atoms interacting with light as a collective. Indeed, considering atoms as group entails besides new dynamic variables and collective states some interesting new properties. The description of superradiance presented in this section is a quite simple approach which is leaned on the description in [18, 4].

### 2.2.1 Collective atomic states

At first, we go back to the two-level system with the ground and excited states  $|1\rangle$ ,  $|2\rangle$  and introduce, in the same manner of creation and annihilation operators for the light field, rising and lowering atomic operators  $\hat{\mathbf{b}}$ ,  $\hat{\mathbf{b}}^{\dagger}$  with the properties

$$\begin{aligned}
\dot{\mathbf{b}}|1\rangle &= 0 & \dot{\mathbf{b}}^{\dagger}|1\rangle &= |2\rangle \\
\dot{\mathbf{b}}|2\rangle &= |1\rangle & \dot{\mathbf{b}}^{\dagger}|2\rangle &= 0.
\end{aligned}$$
(2.21)

In addition to that we introduce the traceless Pauli spin operators by

$$\hat{\mathbf{R}}_{1} = \frac{1}{2} \left( \hat{\mathbf{b}}^{\dagger} + \hat{\mathbf{b}} \right)$$

$$\hat{\mathbf{R}}_{2} = \frac{1}{2} \left( \hat{\mathbf{b}}^{\dagger} - \hat{\mathbf{b}} \right)$$

$$\hat{\mathbf{R}}_{3} = \frac{1}{2} \left( \hat{\mathbf{b}}^{\dagger} \hat{\mathbf{b}} - \mathbf{b} \hat{\mathbf{b}}^{\dagger} \right)$$
(2.22)

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with

$$\hat{\mathbf{R}}_{3}|2\rangle = \frac{1}{2}|2\rangle$$

$$\hat{\mathbf{R}}_{3}|1\rangle = -\frac{1}{2}|1\rangle.$$
(2.23)

States  $|1\rangle$ ,  $|2\rangle$  are eigenstates of  $\hat{\mathbf{R}}_3$ , so that this operator represents a measure of the atomic inversion. The atomic energy  $H_A$  and single atom dipole moment  $\mathbf{d}$  can then be written as

$$\hat{H}_{A} = \hbar \omega_{0} \hat{\mathbf{R}}_{3} + E_{0}$$

$$\hat{\mathbf{d}} = \mathbf{d}_{12} |1\rangle \langle 2| + \mathbf{d}_{12}^{*} |2\rangle \langle 1| = \mathbf{d}_{12} \hat{\mathbf{b}} + \mathbf{d}_{12}^{*} \hat{\mathbf{b}}^{\dagger},$$
(2.24)

with the matrix element  $\mathbf{d}_{ij} = \langle i | \hat{\mathbf{d}} | j \rangle$ . Considering now a group of N identical two-level atoms, equivalently to the single atom case, the collective atomic spin operators

$$\hat{\mathcal{R}}_{1} = \sum_{j=1}^{N} \hat{\mathbf{R}}_{1,j}$$

$$\hat{\mathcal{R}}_{2} = \sum_{j=1}^{N} \hat{\mathbf{R}}_{2,j}$$

$$\hat{\mathcal{R}}_{3} = \sum_{j=1}^{N} \hat{\mathbf{R}}_{3,j}$$

$$\hat{\mathcal{R}}^{2} = \hat{\mathcal{R}}_{1}^{2} + \hat{\mathcal{R}}_{2}^{2} + \hat{\mathcal{R}}_{3}^{2}$$
(2.25)

and lowering and raising operators

$$\hat{\mathcal{R}}^{(-)} = \sum_{j=1}^{N} \hat{\mathbf{b}}_{j}$$

$$\hat{\mathcal{R}}^{(+)} = \sum_{j=1}^{N} \hat{\mathbf{b}}_{j}^{\dagger}$$
(2.26)

are defined as the sum of the operators for each atom j. These new defined operators follow the same commutations relations as angular momentum operators, especially

$$[\hat{\mathcal{R}}_l, \hat{\mathcal{R}}^2] = 0, \quad l = 1, 2, 3.$$
 (2.27)

With respect to this new operators the total atomic energy and total dipole moment for N atoms, with the state  $|1\rangle$  considered as the ground state, are

$$\hat{H}_{A} = \hbar\omega_{0} \left(\hat{\mathcal{R}}_{3} + \frac{1}{2}N\right)$$

$$\hat{\mathbf{D}} = \mathbf{d}_{12}\hat{\mathcal{R}}^{(-)} + \mathbf{d}_{12}^{*}\hat{\mathcal{R}}^{(+)}.$$
(2.28)

#### Dicke states

Consider the product state  $\Psi$  as a representation of N atoms in which  $N_1$  atoms are in state  $|1\rangle$  and  $N_2$  atoms are in state  $|2\rangle$ , e.g.

$$\Psi = |1\rangle_1 |1\rangle_2 |2\rangle_3 |1\rangle_4 \cdot \ldots \cdot |2\rangle_N.$$
(2.29)

Defining the measure of the total atomic inversion

$$m = \frac{1}{2}(N_2 - N_1) \tag{2.30}$$

it follows that

$$\hat{\mathcal{R}}_3 |\Psi\rangle = m |\Psi\rangle, \qquad -\frac{1}{2}N \leqslant m \leqslant \frac{1}{2}N.$$
 (2.31)

 $|\Psi\rangle$  is eigenstate of  $\hat{\mathcal{R}}_3$  with eigenvalue *m* and eigenenergy  $\hbar\omega_0(m + \frac{1}{2}N)$ . Since the energy eigenvalue is independent of the manner in which the  $N_2$  excitations are distributed over the *N* atoms, *m* has a degeneracy of

$$d_m = \frac{N!}{N_1! N_2!} = \frac{N!}{\left(\frac{1}{2}N + m\right)! \left(\frac{1}{2}N - m\right)!}$$
(2.32)

which is greatest for m = 0 for  $m = \pm \frac{1}{2}$ . To reduce this degeneracy new states  $|l, m\rangle$  are defined, which are eigenstates of both  $\hat{\mathcal{R}}_3$  and  $\hat{\mathcal{R}}^2$  at the same time, are defined. By considering m as the eigenvalues of  $\hat{\mathcal{R}}_3$  and denoting the eigenvalues of  $\hat{\mathcal{R}}^2$  in analogy to angular momentum operators by l(l-1) it states

$$\hat{\mathcal{R}}_{3}|l,m\rangle = m|l,m\rangle$$

$$\hat{\mathcal{R}}^{2}|l,m\rangle = l(l-1)|l,m\rangle$$
with  $|m| \leq l \leq \frac{1}{2}N.$ 
(2.33)

It holds that

$$\hat{\mathcal{R}}^{(-)}|l,m\rangle = [(l+m)(l-m+1)]^{1/2}|l,m-1\rangle$$

$$\hat{\mathcal{R}}^{(+)}|l,m\rangle = [(l-m)(l+m+1)]^{1/2}|l,m+1\rangle,$$
(2.34)

i.e. that the lowering and rising operators increase and decrease the value of m and facilitate a generation of further Dicke states with m > -l by applying the raising operator  $\hat{\mathcal{R}}^{(+)}$  or m < l states by  $\hat{\mathcal{R}}^{(-)}$ . The states  $|l,m\rangle$  are called Dicke states and were first introduced by DICKE in 1954 in order to describe N atoms interacting through an electromagnetic field. To the number l he referred to as the *cooperation number*, that will be discussed later.

### 2.2.2 Superradiance: Cooperative spontaneous radiation

Superradiance results from a coupling of multiple atoms through an electromagnetic field. If these atoms are enclosed in a volume, whose dimensions are smaller than the fluorescence wavelength, each of the atoms is not only stimulated by his own fields but also by the fields of his neighbours. As a result, each atom radiates at an enhanced rate and loses energy faster than a single atom.

Under the assumption that each atom senses the same electromagnetic field the total atom-light interaction Hamiltonian states

$$\hat{H}_{\rm I} = -\hat{\mathbf{D}}(t) \cdot \hat{\mathbf{E}}(t) = -\mathbf{d}_{12} \cdot \hat{\mathbf{E}}(t) \hat{\mathcal{R}}^{(-)}(t) - \mathbf{d}_{12}^* \cdot \hat{\mathbf{E}}(t) \hat{\mathcal{R}}^{(+)}(t).$$
(2.35)

Suppose the atomic system to be initially prepared in the Dicke state  $|l, m\rangle$ , while the field resides in the vacuum state  $|vac\rangle$ . The probability to meet the total system in the final state  $|\Phi\rangle$  after the time t is given by the matrix element,

$$-\langle \Phi | \mathbf{d}_{12} \cdot \hat{\mathbf{E}}(t) \hat{\mathcal{R}}^{(-)}(t) - \mathbf{d}_{12}^* \cdot \hat{\mathbf{E}}(t) \hat{\mathcal{R}}^{(+)}(t) | l, m \rangle | \text{vac} \rangle.$$
(2.36)

#### Rate of photon emission

As we are interested in transitions which lead to the emission of a photon, the matrix element reduces to  $\langle \Phi | d_{12} \cdot \mathbf{E}^{(-)}(t) \hat{\mathcal{R}}^{(-)}(t) | l, m \rangle | \text{vac} \rangle$  with the negative part  $\mathbf{E}^{(-)}(t)$  of the electric field. The rate of photon emission is then given by

$$\Gamma_{\text{emission}} \propto \sum_{\text{all}\Phi} \left| \langle \Phi | \mathbf{d}_{12} \cdot \hat{\mathbf{E}}^{(-)}(t) \hat{\mathcal{R}}^{(-)}(t) | l, m \rangle | \text{vac} \rangle \right|^2, \qquad (2.37)$$

that can be separated into matrix elements for the atomic system and the light field yielding the expression

$$\Gamma_{\text{emission}} \propto \langle l, m | \hat{\mathcal{R}}^{(+)}(t) \hat{\mathcal{R}}^{(-)}(t) | l, m \rangle \times \\ \langle \text{vac} | \mathbf{d}_{12} \cdot \hat{\mathbf{E}}^{(+)}(t) \mathbf{d}_{12}^* \cdot \hat{\mathbf{E}}^{(-)}(t) | \text{vac} \rangle$$

$$\stackrel{(2.34)}{=} (l+m)(l-m+1) \cdot A.$$
(2.38)

A can be identified in the case N = 1 which leads to  $m = \frac{1}{2}$  and  $l = \frac{1}{2}$ . This yields the photon emission rate value of A which is identified as the Einstein A-coefficient in the single atom case. For a system of N atoms being all in the ground state the m and l numbers are  $-\frac{1}{2}$  and  $\frac{1}{2}$  respectively. In that case the emission rate becomes zero, as expected. On the contrary having all N atoms excited, with  $m = \frac{1}{2}N$  and  $l = \frac{1}{2}N$ , the emission is NA. This is exactly the photon emission rate for N independently radiating atoms. An more interesting case opens up when the atomic system is exactly half deexcited, i.e. when  $m = 0.^1$  Then photon emission rate for that case is

$$\Gamma_{\text{emission}} = l(l+1)A \tag{2.39}$$

<sup>&</sup>lt;sup>1</sup>For which case the m degeneracy value (2.32) is greatest.

revealing a l dependent magnitude. This is why l is referred to as the cooperation number. When l is largest, i.e.  $l = \frac{1}{2}N$  the emission rate exhibits  $N^2$  dependence

$$\Gamma_{\text{emission}} = \frac{1}{2} N \left( \frac{1}{2} N + 1 \right) A.$$
(2.40)

On the account of this characteristic N dependence of the spontaneous radiation Dicke named this cooperative atomic process *superradiance*.

#### Time development

A characteristic feature for superradiance is the appearance of a short light pulse, that is  $\propto N^2$  and occurs after a characteristic time [18]

$$t_0 = \frac{\ln(N+1)}{AN}.$$
 (2.41)



Figure 2.3: Time development of superradiance. The left graph compares the time dependence of the superradiant and spontaneous photon emission rate for a system of N = 20 atoms. The superradiant emission rate reaches its maximum after the time  $t_0$ , when half of the atoms are deexcited. On the right the time development of the energy is depicted. After the characteristic time  $t_0$  the system has lost half of its initially stored energy.

This is due to the time dependent rate of photon emission, that reaches a maximum value after the characteristic time  $t_0$ . Figure 2.3 illustrates the time development of the superradiant emission rate in comparison to the spontaneous emission rate, that decreases linearly in time. The superradiant emission rate, as already mentioned, exhibits for t = 0 the same magnitude as the spontaneous emission rate, i.e. for N independently radiating atoms. A more detailed discussion on the time development can be found in [18].

Chapter 2. Theoretical foundation

property	scaling
binding energy	$n^{*-2}$
orbital radius	$n^{*2}$
radiative life time	$n^{*3}$
energy separation between adjacent Rydberg states	$n^{*-3}$
dipole moment $\langle nl er n'l'\rangle$ between adjacent Rydberg states	$n^{*2}$

Table 2.1:  $n^*$  scaling of some properties of Rydberg atoms.

### 2.2.3 Rydberg atoms

The term Rydberg atoms denotes excited atoms which have one or more electrons in a high excited state with a large principal quantum number n. The principal quantum number n is associated with the binding energy of an electron in a hydrogen atom insofar as  $E_{\rm B} \propto -n^{-2}$ . Rydberg atoms exhibit hydrogen like behaviour since the highly excited electron experiences an effective potential of a singly positive charged configuration, which equivalently to the hydrogen. Accordingly, Rydberg atoms can be characterized with nearly the same binding energy relation except by a difference in the principal quantum number. The principal quantum number is slightly modified by a quantum defect  $\delta_{nlj}$  to  $n^* \equiv n - \delta_{nlj}$  accounting for the inner structure of the single charged ionized atom [19, 20].

As indicated by table 2.1 Rydberg atoms exhibit a couple of interesting features that intensify with larger principal quantum numbers, for instance large spatial expansion and long radiative lifetimes. However, the features of special interest with respect to superradiance are the two lower ones in table 2.1. The oscillator strengths to nearby states increase quadratically with the principal quantum number. Due to that Rydberg atoms interact very strongly with radiation that is resonant to transitions that connect nearby states. In addition to that the transition wavelengths, which might arise,

$$\lambda_{nn'} \propto n^{*3} \tag{2.42}$$

strongly increase with the principal quantum number. A larger wavelength increases the number of cooperative atoms. That means, that for higher principal quantum numbers less atomic density suffices to get superradiance. Therefore the superradiance is strongly present for Rydberg states and can be a limiting factor for the lifetime of Rydberg atoms [15].

# 2.3 Simulation: Superradiance under continuous pumping

The theory on superradiance above describes the time evolution of an excited atomic system. This approach considers that after the excitation the atomic ensemble decays into the ground state without any interruption. In the experiment, however, the atomic system is pumped continuously at a constant rate to the excited state. As soon as the atoms return to the ground state they are excited again, while the superradiant process is still in progress. Since the superradiance depends on the number of atoms in the ground and excited state the continuous pumping modifies the evolution of the superradiant atomic system. In this case the superradiance exhibits threshold behaviour, which can be demonstrated by simple deterministic and stochastic equations [21].

In this section the simulation on superradiance is introduced that was acquired in the framework of this thesis. As already pointed out in the discussion on the three-level model new effects arise as soon as a further coupled state comes



Figure 2.4: Level scheme of the simulation.

coupled state comes into play. To provide predictions on the upcoming effects the differential equations (2.19) have to be solved, where the experimental demands determine the amount of effort that has to be spent. For the steady state case analytical expressions are available, whereas time dependent solutions require numeric simulations. However, numeric simulations generally hold a speed up in any case and are therefore utilised for the steady state case as well. Since the excitation in the experiment is performed via

two continuously pumping light fields the differential equations (2.19) in the steady state case ( $\dot{\rho} = 0$ ) form the basis for simulation on superradiance.

In figure 2.2 the basic dynamics are governed by the decay rates  $\Gamma_{32}$ ,  $\Gamma_{21}$  and the Rabi frequencies  $\Omega_{12}$ ,  $\Omega_{23}$ . To account for superradiance the three-level model is augmented by one further level insofar as the population transfer from  $|3\rangle$  to  $|4\rangle$  is carried out superradiantly (red decay in figure 2.4). Furthermore, a natural fluorescence decay from  $|3\rangle$  to  $|5\rangle$  is added. This new level scheme involves the additional decay rates

•  $\Gamma_{31}$  is actually not dipole allowed, but serves here as an option to integrate broadening effects like transit time broadening

- $\Gamma_{34,\text{eff}}$  accounts for depopulation of state  $|3\rangle$  through superradiance
- $\Gamma_{35}$  accounts for depopulation of state  $|3\rangle$  through natural fluorescence
- $\Gamma_{51}$ ,  $\Gamma_{41}$  transfers the population back to ground state  $|1\rangle$  to avoid losses of population owing to accumulation in states  $|4\rangle$  and  $|5\rangle$ .

Level  $|4\rangle$  represents the collectivity of states where the population is distributed to in reality. The deviations that arise when the real complex structure is simplified by representation of one surrogate state are examined later in section 4.3.

### 2.3.1 Implementation

The basis of the whole simulation is formed by a standard linear solver method of MATLAB which solves the system of linear equations of the form (2.19). The desired populations are calculated for the given decay rates  $\Gamma_{ij}$  and Rabi frequencies  $\Omega_{ij}$ . Superradiance is implemented by treating level  $|3\rangle$  as the excited state and  $|4\rangle$  the ground state with respect to superradiant processes and setting the single atom decay rate  $\Gamma_{34}$  to the corresponding value. The value of  $\Gamma_{34}$  is determined in the following simple way

$$\Gamma_{34,\text{eff}} = \Gamma_{34} + \Gamma_{34} N_{\text{coop}} = \Gamma_{34} \left( \underbrace{\rho_{33} \cdot \mathcal{N}}_{\mathcal{N}_{\text{Ryd}}} \cdot V_{\text{coop}} + 1 \right), \qquad (2.43)$$

with the ground state density  $\mathcal{N}$ , the Rydberg density  $\mathcal{N}_{\text{Ryd}}$  and the cooperative volume  $V_{\text{coop}}$ . The cooperative volume  $V_{\text{coop}}$  is either defined by the wavelength  $\lambda_{\text{coop}}$  of the radiation that induces the superrdiance

$$V_{\rm coop} = \left(\frac{\lambda_{\rm coop}}{2}\right)^2 \tag{2.44}$$

or by a smaller volume depending on the experimental realisation, for example, a small excitation volume. All atoms enclosed by this volume contribute in the cooperative spontaneous emission. The number of cooperative atoms is in this thesis denoted by  $N_{\text{coop}}$ . The natural decay rate  $\Gamma_{34}$  is multiplied by the number of cooperative atoms  $N_{\text{coop}}$ . This leads to a decay rate that depends on the Rydberg population:  $\Gamma_{34,\text{eff}}(\rho_{33})$ .

### 2.3.2 Evaluation process

Assuming  $\Gamma_{34,\text{eff}}$  according to (2.43) results in a system of non-linear differential equations, as some equations now contain decay terms being proportional to

$$\propto \Gamma_{34,\text{eff}} \cdot \rho_{33} = \Gamma_{34} \cdot \rho_{33}^2, \qquad (2.45)$$

i.e. it is not possible anymore to treat the problem with linear solver methods. Therefore, the solutions are determined via an iterative process, that is introduced in the 2.3. Simulation: Superradiance under continuous pumping



Figure 2.5: Scheme of matching process.

following. Figure 2.5 illustrates a schematic of the iteration process. The iteration process is initiated with a start value  $\rho_{33}(0)$  from which  $\Gamma_{34,\text{eff}}$  is evaluated according to (2.43).  $\Gamma_{34,\text{eff}}$  is again linked to a corresponding Rydberg population  $\rho_{33}$  in the five-level model. If the deviation of the new calculated  $\rho_{33}(i=1)$  compared to the last  $\rho_{33}(0)$ is larger than 0.1% the process returns to a new evaluation of the effective decay rate  $\Gamma_{34,\text{eff}}$  on the basis of the latest  $\rho_{33}$ . This matching of  $\rho_{33}$  and  $\Gamma_{34,\text{eff}}$  is repeated till the condition, that  $\rho_{33}(i)$  differs less than 0.1% from the previous calculated  $\rho_{33}(i-1)$  after the *i*th iteration step, is fulfilled. The resulting value of  $\rho_{33}$  and  $\Gamma_{34,\text{eff}}$  are the wanted solutions for the system of non-linear differential equations. In fact the matching procedure is not completely as simple as presented so far. Namely the calculation of a new  $\Gamma_{34,\text{eff}}$  out of the current solution can bear problems since too large values of  $\Gamma_{34,\text{eff}}$  and a corresponding too small  $\rho_{33}$  can induce oscillations which will not fulfil the condition, even after an arbitrary long iteration time. Therefore, a damping mechanism in order to prevent arising oscillations is integrated. Thereby the next  $\Gamma_{34,\text{eff}}$  value is evaluated out of a modified  $\rho_{33mod}$  which is determined in the following way. First, an average over the last four results is performed

$$\rho_{33\text{mod}} = \frac{1}{4} \sum_{k=i-4}^{i} \rho_{33(k)}.$$
(2.46)

This rough procedure produces a reduction of the distance between the  $\rho_{33}$  value used for determination of the new effective decay rate and  $\rho_{33}(i-1)$ , in order to avoid too large jumps. If the difference between modified  $\rho_{33mod}$  and the reference value, which is the pre-last result  $\rho_{33}(i-1)$ , is still too large, namely if

$$|\rho_{33\text{mod}} - \rho_{33}(i-1)| > |\rho_{33}(i-1) - \rho_{33}(i-1-2)|, \qquad (2.47)$$

then  $\rho_{33 \text{mod}}$  is overwritten by

$$\rho_{33\text{mod}} = \rho_{33}(i-1) + sgn\left(\rho_{33}(i) - \rho_{33}(i-1)\right) \cdot \left|\rho_{33}(i-1) - \rho_{33}(i-1-2)\right|. \quad (2.48)$$

In principle, this mechanism just allows small changes of  $\rho_{33}$ , however in the right direction, so that the solution is probed till the terminating condition is fulfilled.



### 2.3.3 Stability

Figure 2.6: **Dependence of the simulation on the start values.** The figure shows the iteration process for four different initial values of  $\rho_{33}$ . The required times for the matching procedure are depicted for each case.

Actually, this method proves to be insensitive with respect to the start values and converges reliably. This is demonstrated in figure 2.6, where  $\rho_{33}$  approaches the same end value independently of the actual given start value  $\Gamma_{34,\text{eff}}(0)$  or  $\rho_{33}(0)$  respectively. The number of steps and the required iteration time is shorter the closer the initial value is to the actual solution. Furthermore, it can be seen in figure 2.6, for the case of the smallest start value, how quickly the damping mechanism eliminates the starting oscillations of  $\rho_{33}$ .

# Chapter 3 Experimental Setup

In this thesis the impact of superradiance on the decay dynamics is detected indirectly via fluorescence spectroscopy. The purpose of this Chapter is to present the experimental setup used to observe superradiance in thermal vapour of caesium atoms. It is structured in four parts. The first part presents the excitations scheme, followed by the second part which introduces the optical setup. The third part presents the laser system that was used for the excitation. introducing the excitation scheme, the optical setup and the used laser systems. At the end of this chapter the basic technical aspects of the fluorescence spectroscopy are discussed.

### 3.1 Which excitation scheme is most suitable?

A significant demand on this experiment is to guarantee a preferably large Rydberg population in order to investigate superradiance, since we know that superradiance scales with the number of excited atoms. Figure 3.1 shows, with respect to the available laser systems in our group, possible realizations of Rydberg excitations.



Figure 3.1: Overview over the technically accessible excitation schemes. The wavelengths, the magnitudes of the dipole matrix elements as well as the accessible laser powers for the particular transitions are displayed.

The first two excitation schemes illustrate two-photon transitions in Rb, where the

lower transitions from  $5S_{1/2}$  to the  $5P_{1/2}$  and  $5P_{3/2}$  states are addressed by 780 nm and 795 nm cw lasers, while the Rydberg transitions from the intermediate state are driven by 480 nm and 474 nm cw light respectively. The third one depicts a Rydberg excitation in Cs. Here the excitation to the Rydberg S state is carried out with a 455 nm cw laser coupling the states  $6S_{1/2}$  and  $7P_{3/2}$  and infrared 1070 nm cw laser exciting further to the S state. The transition in Cs differs insofar from these of Rb as the probe laser wavelength is shorter than the coupling laser wavelength:  $\lambda_c < \lambda_p$ . This configuration is referred to as an inverted excitation scheme that leads to deviant optical behavior in thermal vapour compared to usual excitation schemes. As these effects are not fundamental for this work they shall not be discussed further here. With the relevant experimental values given in figure 3.1 it's now possible to estimate the achievable Rydberg populations for each system. A three-level simulation according to section 2.1.2 yields to Rydberg populations in Cs up to  $\rho_3 = 0.0025$  being  $10^2$  times larger compared to the Rb systems. On this account the measurements in this thesis were performed with caesium.

### **3.2** Optical setup

Figure 3.2 shows a schematic view of the setup. The two laser beams pass a 5 mm wide vapour cell in counter propagating configuration. The spectroscopy cell is an at the institute commonly used vapour cell with a reservoir, that contains droplets of Cs, attached to it. The cell is placed in an electrical heater that provides separate heating of



Figure 3.2: Schematic of the setup used for the experiment.

the cell and reservoir to adjust the density of the vapour. A pinhole is used to minimize one of the beam sizes and with it the effective illuminated volume in the cell in order to afford measurements on volume dependent behaviour of the atoms that is discussed more in detail below. The fluorescence is detected perpendicular to the direction of the exciting beams. Hereby the fluorescence light is collected with a f = 50 mm A-coated lens and mapped onto the one end a commercial M43L02 Thorlabs multimode fibre with a core diameter of  $\emptyset 105 \ \mu$ m. The multimode fibre provides a wavelength range from 400 nm to 2400 nm. The other end of the fibre is attached to a spectrometer of the model Andor Shamrock SR-303i. Further technical details concerning the fluorescence detection with the Andor Shamrock SR-303i are discussed below. A further laser beam with 852 nm wavelength passes the vapour cell and is detected by a photodiode. This part of the setup is a absorption spectroscopy configuration and serves for measurements of the ground state density, and verification of the vapour temperature respectively. In principle the optical setup, where the relevant measurements on superradiance are performed, is simple and oversee-able. In fact the whole experiment moreover also consists of the preceding laser system, that facilitates the two-photon excitation in caesium, and of the successive fluorescence detection apparatus. As these both are essentially for the entire experiment they will be discussed in the following.

### 3.3 Laser system

The experiment presented in this thesis makes use of an already existing laser system, that was designed and arranged in the framework of a diploma thesis [22]. The system accomplishes the two-photon transition  $6S_{\frac{1}{2}} \rightarrow 7P_{\frac{3}{2}} \rightarrow nS, nD$  in caesium (figure 3.1). This is realized with a commercial tunable frequency doubled amplified diode laser (Toptical TA-SHG Pro) at 455 nm with an output power of ~ 220 mW and a tunable diode laser (External Cavity Diode Laser, Toptica DL100) at 1070 nm with output power of 110 mW. The whole laser set-up is composed of two parts, shortly presented below.

#### Set-up for 455 nm laser light

The first part deals with the 455 nm laser light and consists itself of two elements. The first component is an optical configuration that makes it possible to detune the frequency of the 455 nm light and the second a saturation spectroscopy set-up that affords locking of the laser. The detuning is realized via two double passes with two AOMs which select the  $\pm 1^{st}$  diffraction orders (see left hand side of figure 3.3). After that the light is guided to the laser lock set-up in saturation spectroscopy like configuration. This realisation of a laser lock is referred to as DAVLL (Dichroic Atomic Vapor Laser Lock) spectroscopy [23].

For the purposes of this experiment, i.e. to perform resonant excitation with respect to the intermediate state, the 455 nm laser set-up has been slightly modified by two removable mirrors and  $\lambda/4$ -waveplates right before the AOMs in order to bypass the frequency detuning, when it is required. During measurements the laser was locked to the hyperfine transition  $6S_{\frac{1}{2}}(F = 4) \rightarrow 7P_{\frac{3}{2}}(F' = 5)$ , the strongest (edge) of the error signal.

Chapter 3. Experimental Setup



Figure 3.3: Set-up for the 455 nm laser and 1070 nm laser light. Image adapted from [22].

### 1070 nm Rydberg laser set-up

The second part of the laser system is formed by the optical set-up for the infrared 1070 nm light, shown in the right hand side of figure 3.3 which drives the upper transition. The optical arrangement with the spectroscopy cell enables observation of EIT in Cs. Exactly this configuration can be used to implement a frequency locking procedure for the Rydberg laser [17]. As the locking procedure is required, the spectroscopy set-up was extended by a self made coil around the cell and further optical elements for EIT detection. The magnetic field leads to a frequency shift of the  $\sigma^+$  and  $\sigma^-$  components of the probe beam and produces two shifted EIT signals whose differential signal is used to lock the Rydberg laser to resonance.

Finally the 1070 nm light passes an ytterbium doped fibre, which facilitates an amplification of up to 15 W. This gives the possibility to Rabi frequencies great enough to produce large Rydberg populations. However, measurements reveal that  $\sim 1$  W completely suffices for our purpose.

### **3.4** Fluorescence spectroscopy



Figure 3.4: Spectrometer and a schematic view of its inside.

In this thesis the fluorescence light is coupled into a multimode fibre and guided to the input slit of a grating **spectrometer** from Andor Shamrock. As illustrated in figure 3.4 the light is mapped by the optics of the spectrometer onto the CCD chip of the detector camera. A motorized slit at the side input makes it possible to drive the slit width to the desired position. In order to improve the signal-to-noise ratio the camera can be cooled down up to  $-70^{\circ}$ C by the integrated thermoelectric cooler and up to  $-100^{\circ}$ C by additional use of water cooling. In the experiment the operating temperature was  $-90^{\circ}$ C. Some specifications of the spectrometer are summarized in Table 3.1.

The wavelength selective elements of the spectrometer are reflection gratings. These are located on a grating turret which provides positions for up to three gratings so that

Spectrometer Andor Shamrock SR-303i-B-9FT				
aperture	f/4			
focal length [mm]	303			
input slit width range $[\mu m]$	10 to $2500$			
grating size [mm]	$68 \times 68$			
bandpass [nm]	138.86			
Spectral resolution [nm]	0.308			
maximum attainable wavelength [nm]	2825			
maximum recommended wavelength [nm]	1730			
Detector Andor iDus DU401A-BR-DD				
active pixels	$1024 \times 127$			
pixel size	$26 \ \mu \mathrm{m}$			

Table 3.1: Some specifications of the spectrometer. The resolution values are given with respect to a reflection grating with a groove density of 600 lines/mm at the center wavelength 500 nm. Specification values from Grating Resolution Calculator [24].

the desired grating can be changed between the acquisitions by rotation of the turret. Reflection gratings exhibit a wavelength dependent efficiency which should be checked to meet as good as possible the experimental requirements. The reflection properties are hereby specified by the blaze angle  $\theta_{\rm B}$  or blaze wavelength  $\lambda_{\rm B}$ . A more detailed discussion on diffraction gratings and the efficiency characteristics is given in appendix A. The spectrometer used in the experiment has three reflections gratings mounted on the grating turret:

- grating 1:  $G = \frac{1}{d} = 235$  lines/mm at  $\lambda_{\rm B} = 750$  nm
- grating 2: G = 600 lines/mm at  $\lambda_{\rm B} = 800$  nm
- grating 3: G = 600 lines/mm at  $\lambda_{\rm B} = 500$  nm.

For the fluorescence measurements presented below grating 3 was used.

# Chapter 4

# Experimental Results and Simulation

This chapter presents and discusses the observation of superradiance in a thermal vapour of caesium atoms under continuous pumping. In the experiment the superradiance is observed via fluorescence spectroscopy. Section 4.1 introduces the experimental procedure of the measurements and the characteristics of the acquired fluorescence spectra. Section 4.2 is devoted to the measured data. In this part the dependence on the ground state density, Rabi frequency and excitation volume is investigated. The analysis of the measured data is accompanied by the comparison to the simulation from section 2.3. In section 4.3 the simulation and modifications are discussed in detail.

### 4.1 Experimental procedure

The measurements presented in the following are performed with the grating 3 (section 3.4) 600 lines/mm at  $\lambda_{\rm B} =500$  nm blaze wavelength. The exposure time of the CCD camera was kept constant at  $t_{\rm exp} = 60$  s throughout the entire measurements. The measured wavelength area ranges from 490 nm to 1050 nm and is divided into five acquisition intervals.<sup>1</sup> Typical fluorescence spectra for the five intervals are presented in figure 4.1. The CCD camera detects the fluorescence in full vertical binning (FVB) mode. Without superradiance only the spectral lines (i), (iv) and (v) (see figure 4.1) are visible. The spectral lines (i), which in the following are referred to as the Rydberg lines, correspond to Rydberg decays from the 32S state to 6P ( $32S_{1/2} \rightarrow 6P_{1/2}$  at 497.8 nm and  $32S_{1/2} \rightarrow 6P_{3/2}$  at 511.9 nm). The quite broad spectral lines (iv) at 852 nm and 895 nm result from decays from the  $6P_{3/2}$  and  $6P_{1/2}$  states to the ground state. These two lines are also visible when only the 455 nm excitation laser is switched on. They are the result of the following decay processes:  $7P \rightarrow 7S \rightarrow 6P$  and  $7P \rightarrow 5D \rightarrow 6P$ . The spectral line (v) at 911 nm is the second order grating diffraction of the 455 nm excitation light. The peaks not mentioned so far, like in blocks (ii) and (iii), result

<sup>&</sup>lt;sup>1</sup>The wavelength range of detection is basically limited by the efficiency of the grating.



Figure 4.1: **Typically measured fluorescence spectra.** The fluorescence spectra for the five wavelength intervals of acquisition are illustrated in (a)–(e). (f) Visible decays (i) and (iv) without superradiance. (g) Additional visible decays (ii) and (iii) due to superradiance. For illustration purposes the y-axis in (a)–(e) is normalised to the strongest fluorescence of the interval (b) and the background counts are subtracted. In actual counts the y-axis ranges from ~ 1040 (background counts) to 2100 counts.

transition	highest	lowest	wavelength	visible
group	n	n	range [nm]	
$nF_{7/2,5/2} \to 5D_{5/2,3/2}$	27	4	597 - 1014	Х
nD $_{5/2,3/2} \rightarrow 6P_{3/2,1/2}$	29	6	498 - 921	х
$nS_{1/2} \rightarrow 6P_{3/2,1/2}$	31	7	498 - 1470	Х
$nS_{1/2} \rightarrow 7P_{3/2,1/2}$	31	8	1051 - 4177	-
$nP_{3/2,1/2} \rightarrow 5D_{5/2,3/2}$	31	7	596 - 1380.8	х
$nP_{3/2,1/2} \to 6S_{1/2}$	31	6	319 - 895	-
$nP_{3/2,1/2} \rightarrow 7S_{1/2}$	31	7	785 - 3119	-
$nP_{3/2,1/2} \to 6D_{5/2,3/2}$	31	8	1153 - 3206	-
$nD_{5/2,3/2} \rightarrow 7P_{3/2,1/2}$	29	7	1052 - 2424	-
$nD_{5/2,3/2} \to 4F_{7/2,5/2}$	29	7	1472 - 6292	-
$nF_{7/2,5/2} \to 6D_{5/2,3/2}$	27	4	1134 - 5474	-
$nF_{3/2,1/2} \rightarrow 4G_{5/2,3/2}$	27	6	>1133	-

Table 4.1: **Overview over some groups of transitions.** The dipole allowed transitions are combined in blocks of spectral lines. The wavelength range is given for each block. The blocks that can be detected with the spectrometer are marked with an x.

from superradiant cascading. In the following these are referred to as the superradiant lines. Table 4.1 presents an overview over the dipole allowed transitions, which could be observed in the fluorescence. As indicated in the table only transitions which end up in the  $5D_{5/2,3/2}$  and  $6P_{3/2,1/2}$  states can be detected by the spectrometer, when grating 3 is used. Other transitions are not detected as the transition wavelengths are either too large or too small. The assignment of the superradiant lines to the transition groups listed in table 4.1 is presented in the figures B.1 to B.5 in appendix chapter B. The superradiant lines will be used for the analysis to conclude on the superradiant properties. From the Rydberg lines only the 498 nm Rydberg ( $32S \rightarrow 6P_{1/2}$ ) line is used for the analysis below. The Rydberg line at  $512 \text{ nm} (32S \rightarrow 6P_{3/2})$  is not suited for analysis as there are superradiant lines nearby. When these superradiant lines become large they cannot be distinguished from the  $32S \rightarrow 6P_{3/2}$  transition any more.

The fluorescence was measured in dependence of the ground state density  $\mathcal{N}$ , the Rabi frequency  $\Omega_{\rm P}$  of the 455 nm exciting light and the excitation volume. The excitation volume is defined as

$$V_{\text{excitation}} = \frac{\lambda_{\text{coop}}}{2} \cdot \pi \left(\frac{d_{\text{beam}}}{2}\right)^2.$$
(4.1)

In the experiment the excitation volume is changed by changing the beam size  $d_{\text{beam}}$  of the 455 nm laser beam with the aperture of a pinhole, i.e. by changing the diameter of the pinhole opening.  $\lambda_{\text{coop}}$  is the wavelength that corresponds to the transition  $32S \rightarrow 31P$  and is of the size  $\lambda_{\text{coop}} \approx 1.8$  mm. Spontaneous radiation resonant to this transition is most likely to induce a cooperative spontaneous emission in the ensemble

of excited atoms. The cooperative volume  $V_{\text{coop}}(\lambda_{\text{coop}} = 1.8 \text{ mm})$  according to equation (2.44) is in the following referred to as the maximum cooperative volume.

During the measurements the Rabi frequency of the coupling beam was kept constant at  $\Omega_c/2\pi = 10.9$  MHz. Only the probe Rabi frequency  $\Omega_p$  was varied.

The ground state density was varied via the reservoir temperature (see section 3.2). For the analysis the ground state density was calculated from the temperature dependent atomic density function of caesium in gas phase [25, 22]

$$n(T) = \frac{1.0133 \cdot 10^{9.165 - \frac{3830}{T}}}{k_B}.$$
(4.2)

T denotes the absolute temperature and  $k_B$  is the Boltzmann constant. The ground state density in the measurement ranges from 2.0 to  $7.5 \times 10^{11} \text{cm}^{-3}$ .

In general, the excitation of atoms to the Rydberg state is limited by the Rydberg blockade [26]. For the conditions of the experiment presented in this thesis the Rydberg blockade is expected to be present for Rydberg densities  $N_{\rm Ryd} \gtrsim 10^{12} {\rm cm}^{-3}$ . Hence, the Rydberg blockade has not to be taken into account in the analysis.

# 4.2 Effects of the superradiance on the decay dynamics

Superradiant cascading causes a distribution of the initially excited population over various states, which gives rise to several additional fluorescence lines (see figure 4.1 and section 4.1 above). Figure 4.2 shows an example of how the fluorescence in the vicinity of the Rydberg lines changes when the Rabi frequency is increased. As can be seen from (b) to (c), some kind of phase transition takes place [14, 21]. The initially quite large Rydberg line shrinks in relation to the superradiant lines until it gets comparatively insignificant.

In the analysis procedure the intensity of the spectral lines is evaluated via determination of the maximum value of each fluorescence peak.

### 4.2.1 Dependence on ground state density

Figure 4.3 illustrates the density dependence of the Rydberg line and the superradiant lines for different beam diameters. For completeness also the intensities of the transition groups from table 4.1 are shown to illustrate their contribution.

To provide a basis for interpretation of the data one should shortly recapitulate the optical response for independent atoms. When increasing the number of atoms the intensity of the Rydberg line is expected to grow linearly with the atomic density  $\mathcal{N}$ . The fluorescence from the excited state is given by (2.20) and  $\rho_{33}$  is constant with respect to  $\mathcal{N}$ . Figure 4.3, however, shows that the Rydberg line intensity is rather decreasing when the ground state density is increased. This clearly indicates that  $\rho_{33}$ , which refers



Figure 4.2: Characteristic transformation of detected fluorescence. Fluorescence spectra in the vicinity of the Rydberg line (highlighted area) under systematic change of  $\Omega_{\rm P}$ . The Rabi frequency  $\Omega_{\rm P}$  is increased from top to bottom: (a)  $\Omega_{\rm P}/2\pi = 2.1$  MHz (b)  $\Omega_{\rm P}/2\pi = 2.97$  MHz (c)  $\Omega_{\rm P}/2\pi = 3.64$  MHz and (d)  $\Omega_{\rm P}/2\pi = 4.7$  MHz. Experimental parameters: pinhole truncated beam size diameter  $d_{\rm beam} = 1.0$  mm and ground state density  $\mathcal{N} \sim 3 \times 10^{11} {\rm cm}^{-3}$ .

to the population of the 32S state, no longer remains constant but changes with the total number of atoms, as is expected for superradiance (see section 2.2.2). Furthermore the data shows that the intensity of the superradiant lines increases strongly. As the decrease of the Rydberg line intensity starts when the intensity of all superradiant lines increases these two processes seem to be correlated. The experimental data in figure 4.3 clearly show a threshold behaviour around  $4-5 \times 10^{11} \text{cm}^{-3}$ .

In figure 4.4 the intensity of the Rydberg line and the superradiant lines for beam size  $d_{\text{beam}} = 1 \text{ mm}$  from figure 4.3 are illustrated again. The data is normalized and rescaled with  $\mathcal{N}$  in order to extract the density dependence of the Rydberg population  $\rho_{33}(\mathcal{N})$ . This data is compared to the theoretical simulation from section 2.3. As can be seen, the simulation on density dependence also exhibits a phase transition from higher to lower Rydberg population of the 32S state. The population  $\rho_{33}$  is transferred to level 4 so that the population  $\rho_{44}$  has a phase transition from lower to higher population. Although the simulation also exhibits a phase transition it does not reproduce the

threshold behaviour from the measurements. The phase transition of the simulation is broader than in the measurement. Possible reasons for the discrepancy between simulation and reality are discussed in section 4.3.



Figure 4.3: **Density**  $\mathcal{N}$  dependence of detected fluorescence. Fluorescence is measured for three different beam sizes: 1 mm, 1.5 mm and 2 mm. The upper graph (a) shows the intensity of the Rydberg line and the lower one (b) the intensity of all superradiant lines against the ground state density. From (c) to (e) the intensity of the fluorescence blocks (table 4.1) is illustrated for each beam size. The Rabi frequency of the probe beam is  $\Omega_p/2\pi = 2.1$  MHz.



Figure 4.4: Comparison of the dependence of the measurement and simulation on the ground state density  $\mathcal{N}$ . Measurement and simulation for a probe Rabi frequency of  $\Omega_p/2\pi = 2.1$  MHz and a beam diameter of  $d_{\text{beam}}=1$  mm. The measurement data is rescaled with  $\mathcal{N}$  to clarify the superradiant modification on the Rydberg population. For illustration purposes both datasets are normalized to 1. The blue highlighted area in the simulated graph indicates the density range of the measurements.

### 4.2.2 Dependence on the Rabi frequency

The measured intensities are plotted against the Rabi frequency in figure 4.5. For small Rabi frequencies the intensity of the Rydberg line increases linearly while the intensity of the superradiant lines stays almost at zero. As the Rabi frequency reaches  $\sim 3$  MHz the spontaneous emission from the 32S state collapses, while the intensity of the superradiant lines increases. Similar to the density measurement the decrease of the Rydberg line intensity and the increase of the superradiant lines intensity and the increase of the superradiant lines intensity are very likely to be correlated and exhibit a threshold behaviour.



Figure 4.5: Dependence of the detected fluorescence on the Rabi frequency  $\Omega_{\mathbf{p}}$ . (a) illustrates the measured intensity of the Rydberg line and the superradiant lines against the Rabi frequency  $\Omega_{\mathbf{p}}$ . (b) shows the decomposition of the intensity of the superradiant lines into the transitions groups from table 4.1. Experimental parameters are: beam size diameter  $d_{\text{beam}} = 1.0$  mm and ground state density  $\mathcal{N} \sim 3.1 \times 10^{11} \text{cm}^{-3}$ .

Figure 4.6(a) presents the results of the simulation from 2.3 for varying Rabi frequency. Obviously, the simulation does not reproduce the measured results.  $\rho_{44}$  and  $\rho_{33}$  rise and decrease nearly for the same Rabi frequency. The measured Rydberg population decreases clearly faster than the simulation. For comparison, the results of the 5-level model without superradiance are depicted in figure 4.6(b). In the model without superradiance  $\rho_{44}$  and  $\rho_{33}$  also increase and decrease at the same Rabi frequency. The difference to the simulation with superradiance is that  $\rho_{44}$  is 10<sup>4</sup> times smaller in magnitude than  $\rho_{33}$ . On the contrary, in the simulation with superradiance  $\rho_{44}$  is two times larger than  $\rho_{33}$ .

In the experiment the rise of emission from neighbouring states, and therefore the rise of their population, is shifted to higher Rabi frequencies with respect to the Rydberg line whereas in the simulated  $\rho_{44}$  rises right from the beginning on and is larger than  $\rho_{33}$  in magnitude almost all the time. Besides that, the superradiant curves in Figure 4.6 are much broader than the curves of the non-superradiant five-level simulation. Possible reasons for the discrepancy between the simulation and the measurement are discussed in section 4.3.



Figure 4.6: Simulated dependence on Rabi frequency  $\Omega_{\mathbf{p}}$ . The graph (a) shows the results of the simulation with superradiance approximation for the beam diameter  $d_{\text{beam}} = 1.0 \text{ mm}$  and a ground state density of  $\mathcal{N} \sim 3.1 \times 10^{11} \text{ cm}^{-3}$ . For comparison (b) shows the simulation without superradiance.

### 4.2.3 Dependence on the excitation volume

The purpose of varying the excitation volume is to examine whether it has an influence on the superradiant dynamics. The number of cooperative atoms decreases when the excitation volume (4.1) is smaller than the maximum cooperative volume. On the assumption that two beam sizes  $d_1$  and  $d_2$  with  $d_2 > d_1$  produce excitation volumes  $V_1$ and  $V_2$  according to (4.1) which are smaller than the maximum cooperative volume, the critical threshold density is higher for  $V_1$  than for  $V_2$ . The critical densities for different excitation volumes are related to each other by

$$\mathcal{N}_{\text{crit},d_1} \cdot V_1 = N_{\text{crit}} = \mathcal{N}_{\text{crit},d_2} \cdot V_2 \tag{4.3}$$

with  $N_{\rm crit}$  being the critical number of atoms, or with equation (4.1) by the ratio

$$\frac{\mathcal{N}_{d_1}}{\mathcal{N}_{d_2}} = \frac{d_2^2}{d_1^2}.$$
(4.4)

If  $V_2$  is larger than the maximum cooperative volume  $V_{\text{coop}}$  then the ratio is

$$\frac{\mathcal{N}_{d_1}}{\mathcal{N}_{d_2}} = \frac{V_{\text{coop}}}{V_1} \stackrel{(2.44)}{=} \frac{\lambda_{\text{coop}}^2}{d_1^2 \pi}.$$
(4.5)

The fluorescence is measured from 1 mm to 2 mm in steps of 0.1 mm and an additional measurement at 0.6 mm. The experimental results are depicted in figure 4.7. Figure



Figure 4.7: Beam size  $d_{\text{beam}}$  dependence of the fluorescence. (a) illustrates the dependence of the Rydberg line intensity and the intensity of the superradiant lines on the beam size. (b) shows the decomposition of the intensity of the superradiant lines into the transitions groups from table 4.1. The experimental parameters are: Rabi frequency  $\Omega_{\rm P}/2\pi = 2.1$  MHz and ground state density  $\mathcal{N} \sim 3.65 \times 10^{11} {\rm cm}^{-3}$ .

4.7(a) shows that the Rydberg line intensity first increases linearly and then remains constant from 1.2 mm to 2 mm. In parallel the intensity of the superradiant lines increases rapidly with increasing beam size. The intensity of the superradiant lines shows a threshold behaviour which is similar to the behaviour for varying ground state density and Rabi frequency. The Rydberg line intensity, however, does not show the decrease that is observed in the previous measurements at threshold.

The increase of the signal in figure 4.7(a) results from the fact that varying the beam size can change the experimental conditions. This is demonstrated in figure 4.8(a). The size of the detection area is approximately of the size of a few hundred micrometers  $\sim 100 - 200 \ \mu \text{m.}^2$  In the case that the fluorescence detection area is located at the center of the beam cross-section, an increasing beam does not change the detected fluorescence intensity. If, however, the fluorescence detection area is located at the edge of the beam cross-section then a larger beam size causes more illuminated atoms in the detection area. The detected fluorescence increases with the beam size. As a result the changes for different beam sizes in figure 4.8(a) are a mixture of both the contribution of the superradiance and of the experimental conditions.

<sup>&</sup>lt;sup>2</sup>This value was estimated via calculating the image size of the multimode fibre core for a f = 50 mm lens.

4.2. Effects of the superradiance on the decay dynamics



Figure 4.8: Fluorescence detection and demonstration of the properties of the intensity curves. In (a) a schematic of two possible configurations of the detection volume relative to the laser beams is illustrated. (b) Illustration of the fluorescence intensity in dependence of ground state density for two different beam diameters. For illustration purposes the fluorescence signal is represented by error functions. (c) depicts the ratio between the two curves from (b).

As mentioned above the critical threshold density is expected to be higher for a smaller beam sizes. As a result the intensity curves for varying density, which in the following are denoted by the expression density curves, are shifted with respect to each other, as demonstrated in figure 4.8(b). An additional contribution due to experimental conditions results in a constant factor of the curve for the larger beam size with respect to the curve with the smaller beam size. Indeed, figure 4.3(b) reveals that there seems to be such a factor between the 1 mm density curve and the 1.5 mm and 2 mm density curves. This leads to the conclusion that in this measurements the detection area is located near the edge of the beam cross-section. A shift, which indicates an influence of the excitation volume on the superradiant dynamics, however, is not really visible in figure 4.3(b). To get the information, whether there is a shift, one has to calculate the ratio between the density curves for two different beam sizes. If there is a shift the resulting curve has a form like the one presented in figure 4.8(c). If there is no shift the ratio is constant for all ground state densities.

In order to extract more information from the density curves in figure 4.1 a fit of the form

$$I_{\rm fl}(\mathcal{N}) = a \cdot \operatorname{erf}\left(\frac{(\mathcal{N}-b)}{c}\right) + d \tag{4.6}$$

is applied for each beam size. The fitted density curves are presented in figure 4.9(a). The fitting parameters b and d with the fitting errors are displayed in table 4.2. The

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beam size	$b \ [10^{11} \mathrm{cm}^{-3}]$	$d \ [10^5 \ \text{counts}]$
1 mm	$4.85 (\pm 0.11)$	$0.42 \ (\pm 0.02)$
$1.5 \mathrm{~mm}$	$4.61 \ (\pm 0.23)$	$0.74~(\pm 0.07)$
2  mm	$4.89 (\pm 0.31)$	$0.99~(\pm 0.11)$

Table 4.2: Fitting parameters according to equation (4.6). The table presents the values of the fitted parameters b and d for the curves in figure 4.9(a) with the fit errors.

parameter d describes the offset due to experimental conditions and parameter b holds the information on the shift, i.e. it corresponds to the critical density. The ratio, which was calculated from the data points and from the fitted curves, is illustrated in figure 4.9(b). The ratios  $I_{1.5mm}/I_{1mm}$  and  $I_{2mm}/I_{1mm}$  have a form that is similar to the curve in 4.8(c). This reveals what cannot be seen clearly in figure 4.3(b), namely that the threshold for 1 mm density curve is shifted with respect to the 1.5 mm and 2 mm density curves. The shift proves that the decrease of the beam size from 1.5 mm to 1 mm has influence on the superradiant dynamics. The ratio  $I_{2mm}/I_{1.5mm}$ , however, is nearly constant for all densities. The increase from  $d_{beam} = 1.5$  mm to  $d_{beam} = 2$ mm does not influence the superradiant dynamics. This implies, that with the beam



Figure 4.9: Modification of detected fluorescence intensity with different beam sizes. (a) shows the data from figure 4.3(b) with the fitted error functions. (b) illustrates the ratios, which were calculated from the data points and from the fitted curves.

size  $d_{\text{beam}} = 1.5$  mm the excitation volume is larger than the maximum cooperative volume.

As there is a significant difference in the superradiant dynamics between the beam sizes  $d_{\text{beam}} = 1 \text{ mm}$  and  $d_{\text{beam}} = 1.5 \text{ mm}$  beam sizes, one could check relation (4.4) for the two beam sizes. The fitted *b* parameters for these beam sizes correspond to the critical threshold density. The ratio of the critical densities is

$$\frac{\mathcal{N}_{\text{crit,1 mm}}}{\mathcal{N}_{\text{crit,1.5 mm}}} = \frac{4.85}{4.61} = 1.05.$$
(4.7)

In comparison the ratio of the beam sizes is

$$\frac{d_2^2}{d_1^2} = \frac{1.5^2}{1} = 2.25. \tag{4.8}$$

The ratio of the beam sizes (4.8) is two times larger than the ratio of the densities (4.7). The relation (4.4) is not true for these beam sizes. However, as recognized above the excitation volume that corresponds to the beam size  $d_{\text{beam}} = 1.5$  mm is larger than the maximum cooperative volume  $(I_{2\text{mm}}/I_{1.5\text{mm}} = const.$  in figure 4.9(b)). Therefore the calculated ratio of densities (4.7) should fulfill the equation (4.5)

$$\frac{\lambda_{\rm coop}^2}{d_1^2 \pi} = \frac{1.8^2}{1^2 \cdot \pi} = 1.03. \tag{4.9}$$

This value is very close to the ratio (4.7), which confirms the assumption that the maximum cooperative volume is completely illuminated.

Furthermore the analysis procedure, as performed above, allows to calculate the actual maximum cooperative volume. From the calculated maximum cooperative volume it could be possible to draw conclusions on the actual cooperative wavelength  $\lambda_{\text{coop}}$  and the corresponding transition that stimulates the superradiant process.

The beam size dependent measurements were mainly performed for beam sizes from 1 mm to 2 mm. The ratios (4.8) and (4.9), however, show that these beam sizes are almost at the truncation point, which is characterized by the maximum cooperative volume. If one is interested in imaging the modification of superradiant dynamics through the beam size, one has to repeat these measurements for smaller beam sizes.

Figure 4.10 shows the simulated dependence on the beam size. It can be seen that the simulated  $\rho_{33}$  decreases until the truncation point (indicated by vertical line) and reaches a constant value. Similarly  $\rho_{44}$  first increases and then reaches a constant value at the truncation point. At the truncation point the excitation volume in the simulation becomes larger than the maximum cooperative volume.

As discussed above, the measured beam size dependence is a mixture of the contribution from superradiance and the experimental conditions. Although the d parameters contain some information about the experimental parameters, three points are too

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Figure 4.10: Comparison of the simulated  $d_{\text{beam}}$  dependence and rescaled measurement data. Depicted are measurement and simulation for a probe Rabi frequency of  $\Omega_p/2\pi = 2.1$  MHz and ground state density  $\mathcal{N} \sim 3.65 \times 10^{11} \text{cm}^{-3}$ . For comparison, the measurement data is rescaled with  $1/d_{\text{beam}}$ .

unprecise to determine the modification due to experimental conditions. However, in order to compare the simulation to the measured data, the measured beam size dependence is rescaled with  $(d_{\text{beam}})^{-1}$ . This corresponds to the assumption, that the modification due to experimental conditions goes linearly with the beam size. When the measured data is rescaled the Rydberg line intensity shows a decrease, as is also predicted by the simulation and the previous measurements at threshold.

In figure 4.11 on the contrary the simulated data is rescaled with  $d_{\text{beam}}$  and compared to the measured data. The rescaled  $\rho_{33}$  then also increases at the beginning like the measured data.

The comparison of measured and simulated data after rescaling one of them confirms again, that the measured dependence of the fluorescence on the beam size results from a mixture of superradiance and the experimental conditions.

### 4.2.4 Disucussion of the problems

During the measurements several problems occurred, which limited the accuracy of measurements. These are discussed in the following.



Figure 4.11: Comparison of the measured  $d_{\text{beam}}$  dependence and the rescaled simulation data. Depicted are measurement and simulation for a probe Rabi frequency of  $\Omega_{\rm p}/2\pi = 2.1$  MHz and ground state density  $\mathcal{N} \sim 3.65 \times 10^{11} {\rm cm}^{-3}$ . For comparison, the simulated results are rescaled with  $d_{\text{beam}}$ .

#### Stable laser powers

For the Rabi frequency dependence the laser power of the 1070 nm Rydberg laser was kept constant at  $\sim 900$ mW while the power of the 455 nm ground state laser was varied from 1 mW to 60 mW. From figure 4.5 is visible that the interesting region, i.e. the region of phase transition, lies between 2 and 5 MHz of Rabi frequencies of the probe beam. This corresponds approximately to laser powers from 2-10 mW. The drifts of the laser power, however, could be from time to time of the magnitude of 2-5 mW within a few tens of seconds. This is a quite large drift, if one intends to measure to an accuracy of 0.5 mW, which obviously is required to resolve the phase transition. Hence the laser power had to be continuously checked and readjusted during the running measurements. This could result in inaccuracy of the data.

#### Locking of the Rydberg laser

Although the EIT spectroscopy set-up of the 1070 nm light was accomplished for a locking procedure of the Rydberg laser, a locking of the laser actually failed due to peculiar problems with the used PID controllers. This causes a detuning with respect to Chapter 4. Experimental Results and Simulation

the 32S state, which can drift during a fluorescence measurement. A varying detuning, however, changes the Rydberg population. As the fluorescence is very sensitive to the Rydberg population at the threshold this could also result in inaccuracy of the data. Due to the detuning drifts is has been observed that the Rydberg population is larger when the 1070 nm laser is red-detuned ( $\delta_{1070,nm} < 0$ ) with respect to the resonance ( $\delta = 0$ ) and smaller when the laser is blue-detuned ( $\delta_{1070,nm} > 0$ ). This asymmetry has also been observed by [14, 27].

### Fluorescence detection

As explained above, the fluorescence detection area seemed to be located at the edge of the exciting beams during the experiments. This circumstance causes an annoying additional influence, when measuring the dependence on the excitation volume (see section 4.2.3). For future measurements the fluorescence detection should be readjusted, so that the detection area is located at the center of the beam cross-section. This also leads to a larger signal in the detected spectra and enable analysis of the spectra in the regime, where the detection signal is at the moment too small for analysis.

## 4.3 Simulation

As discussed above the 5-level simulation with superradiance proves to be stable in terms of sensitivity to initial conditions and exhibits a threshold behaviour similar to the one discovered in the measurements. However, there is still a noticeable discrepancy to the behaviour observed in the experiment. This section is intended to discuss the assumptions that were investigated in order to explain these disagreements.

### 4.3.1 Additional levels and superradiant decays

The 5-level simulation is a very simplified model that considers superradiant effects with barely one superradiant decay, which affects the population of the initially excited state 32S. In reality, superradiant cascading occurs. States that are populated by superradiance are depopulated again by superradiance. This procedure continues for the next populated state until the superradiance condition breaks down. In order to analyze whether and how successive superradiant decays affect the Rydberg population of 32S state the 5-level simulation is extended by two further superradiant decays in two steps. The resulting 5-, 7- and 8-level simulations consist of the levels described in 4.12. Note that the fixed decay rates have to be matched among the single simulations in order to keep them comparable. For instance, the fixed decay rate from state 4 in the 5-level simulation should contain the sum of all decay paths after level 4 in the 8-level simulation.

Figure 4.13 demonstrates for each population the modification of the dependence on the ground state density. As can be seen the Rydberg population  $\rho_{33}$ , which represents

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Figure 4.12: **Overview on the simulated level scheme.** The primary 5-level includes levels 1 to 5. In the 7-level scheme one superradiant and one natural fluorescence decay, which depopulate level 4, are added. The 8-level simulation contains one additional superradiant decay, which depopulates level 7. Curly arrows with a red S denote superradiant decays. Curly arrows without a red S denote natural decays.

the population of the 32S state, exhibits the same behaviour in the 5-, 7- and 8-level simulation. Successive superradiant decays do not influence  $\rho_{33}$ . In contrast,  $\rho_{44}$  differs from 5- to 7-level simulation but not from 7- to 8-level simulation and  $\rho_{77}$  differs from 7- to 8-level simulation.  $\rho_{44}$  reveals a different behaviour in the 7-level simulation in comparison to the 5-level simulation because level 4 no longer suffers from a constant depopulation but from a density dependent depopulation. As a result of superradiant depopulation  $\rho_{44}$  is "compressed" in density frame. The same happens with  $\rho_{77}$ . The simulation data also show that the increase in population of level 7 occurs later than the increase of level 4. The results of fig. 4.13 lead to the conclusion that the consideration of successive superradiant depopulation does not affect the density behaviour of the initially excited Rydberg population when the relevant decay rates are matched. This means that the simulation with superradiance can be reduced to a few states.

Up to this point we just discussed the influence of succesive superradiant cascading on the populations but did not discuss the fact that the 32S state could also radiate superradiantly to P-states other than 31P. To investigate the magnitude of this effect it is not necessary to expand the simulation by further P-states. This is due to the fact that decays to additional P-states only change the effective decay rate of the 32S state. The next subsection will discuss the influence of modified decay rates on the populations.



Figure 4.13: Simulation for different number of levels. The figure illustrates the results of the primary 5-level simulation in comparison to the extension to 7and 8-level simulation in accordance with the level scheme presented above (fig. 4.12). Depicted is the dependence on ground state density  $\mathcal{N}$  of the populations  $\rho_{33}$ ,  $\rho_{44}$  and  $\rho_{77}$ .

### 4.3.2 Modified decay rates

To imply the influence of additional decays to P-states the primarily effective decay rate (2.43) with  $\Gamma_{34,\text{eff}} = \Gamma_{32S \rightarrow 31P}$  for transition  $32S \rightarrow 31P$ 

$$\Gamma_{\text{eff}} = \Gamma_{32S \to 31P}(\rho_{33} \cdot N_{\text{coop}} + 1) = \Gamma_{32S \to 31P}(\rho_{33} \cdot (\mathcal{N} \cdot V_{\text{coop}}) + 1)$$
(4.10)

with the ground state density  $\mathcal{N}$  and the cooperative volume  $V_{\text{coop}}$  has to be expanded by the sum decay rates to other P-states

$$\Gamma_{\text{eff,new}} = \Gamma_{\text{eff}} + \sum_{10 < n < 31} \Gamma_{32S \to nP} \left( \rho_{33} \cdot \left( \mathcal{N} \cdot \left( \frac{\lambda_{32S \to nP}}{2} \right)^3 \right) + 1 \right)$$
(4.11)

$$=\Gamma_{\text{eff}} + \sum_{10 < n < 31} \Gamma_{32S \to nP} \rho_{33} \cdot \mathcal{N} \cdot \left(\frac{\lambda_{32S \to nP}}{2}\right)^3 + \sum_{10 < n < 31} \Gamma_{32S \to nP} \qquad (4.12)$$

$$\approx \Gamma_{\text{eff}} + \Gamma_{32S \to 31P} \rho_{33} \cdot \mathcal{N} \cdot 0.037 + \Gamma_{32S \to 31P} \cdot 27.3.$$
(4.13)

The values 0.037 and 27.3 in equation (4.13) has been determined from the values for the decay rates  $\Gamma_{32S\to nP}$ . The radial part of the decay rates  $\Gamma_{32S\to nP}$  has been calculated with the semiclassical approximation from [28]. When assuming superradiant effects down to state 10P then approximately 27.3 times the natural linewidth of  $\Gamma_{32S\to 31P}^{3}$ 

<sup>&</sup>lt;sup>3</sup>With the radial part calculated with [28]:  $\Gamma_{32S\to31P} < 10$  kHz.



Figure 4.14: Comparison of simulation results for different decay rates. (a) shows the density dependence of  $\rho_{33}$  (upper graph) and  $\rho_{44}$  (lower graph) calculated with the 5-level simulation for constant  $\Gamma_{31}$  and varying  $\Gamma_{41}$  rate. In (b)  $\Gamma_{41}$  is kept constant while  $\Gamma_{31}$  is varied. Each curve is normalised to [0,1] to provide direct comparison in density dependence for different decay rates.

and 4% of the amount coming from superradiance has to be added. Executing the simulation under assumption of these values the difference is insignificantly small. In the 5-level simulation superradiance to further P-states is negligible.

In order to demonstrate to what extent different decay rates affect the decay dynamics, figure 4.13 shows the populations  $\rho_{33}$  and  $\rho_{44}$  for varying values of  $\Gamma_{31}$  and  $\Gamma_{41}$ . While an altering  $\Gamma_{41}$  rate does not affect density dependence of both  $\rho_{33}$  and  $\rho_{44}$ , except the magnitude of  $\rho_{44}$ , figure 4.13(b) shows that larger  $\Gamma_{31}$  rates shift the threshold to higher densities. Furthermore, though not clearly visible in 4.13(b), a larger  $\Gamma_{31}$  rate slightly flattens the drop of  $\rho_{33}$  and the rise of  $\rho_{44}$ .

In all 5-level simulations presented above a constant rate of  $\Gamma_{31} = 6.9$  MHz was assumed. This rate is composed of two basic thoughts. A decay rate of ~ 0.9 MHz for  $\Gamma_{31}$  takes into account that the atoms have a finite transit time after which they leave the region illuminated by the beam (equation (3.29) in [17]). That means that after this time an excited atom is substituted by an atom in the ground state. In this experiment we assume additionally a broadening of ~ 6 MHz for Rydberg states, which was observed in another experiment in the same lab. Similarly to  $\Gamma_{31}$   $\Gamma_{41}$  was fixed at 9.3 MHz.  $\Gamma_{41}$ , however, does not change the density behaviour that much and is therefore not that important (see figure 4.13). To sum up, the measurement results reveal that the phase transitions starts later and the threshold exhibits a sharper drop as predicted by the simulations. Altering the decay rates however does not reproduce the threshold slope of the experiment.

### 4.3.3 Effective ground state density

During measurements the 455 nm laser is locked to the  $6S_{\frac{1}{2}}(F=4) \rightarrow 7P_{\frac{3}{2}}(F=5)$ transition, i.e. atoms in the  $6S_{\frac{1}{2}}(F=3)$  are not addressed by the laser and remain in the ground state. As the initial  $^26S_{\frac{1}{2}}$  population is equally distributed over the 16  $m_{\rm F}$ levels about half of the ground state atoms are effectively accessible for the excitation process. Figure 4.15(a) presents the changes of the five-level simulation results when only half of the ground state density is considered. Similar to the discussion of figure 4.14 the density dependent curve is both shifted to higher densities and exhibits a slight flattening of the slope. The Rabi frequency dependent curves, depicted in figure 4.15(b), on the other hand are "compressed" with less effective ground state density. Additionally, the maximum value of  $\rho_{33}$  and  $\rho_{44}$  increases and decreases respectively. Another mechanism causing a reduction of the effective ground state density could be optical pumping to the  $6S_{\frac{1}{2}}(F=3)$  state, which is a dark state with respect to the exciting lasers. One could argue that the  $6S_{\frac{1}{2}}(F=4) \rightarrow 7P_{\frac{3}{2}}(F=5) \rightarrow 32S_{\frac{1}{2}}$  is not a closed system leading to decays via intermediate state to the F = 3 ground state. Actually, observations showed that this excitation scheme exhibits almost cyclic character [22] so that optical pumping effects do not significantly influence the ground state density.



Figure 4.15: Comparison of simulation results for different effective ground state densities  $\mathcal{N}_{\text{eff}}$ . (a) shows the five-level simulation results of the ground state density dependence  $\mathcal{N}$  of  $\rho_{33}$  (upper graph) and  $\rho_{44}$  (lower graph) for different fractions of addressable ground state atoms: 1,  $\frac{1}{2}$ ,  $\frac{1}{5}$ ,  $\frac{1}{10}$ ,  $\frac{1}{20}$ . Similarly (b) presents the simulation results for Rabi frequency dependence of  $\rho_{33}$  and  $\rho_{44}$ .



Figure 4.16: Comparison of simulation results for different effective Rydberg densities  $\mathcal{N}_{\mathbf{Ryd,eff}}$ . (a) shows the five-level simulation results for the dependence of  $\mathcal{N}$  of  $\rho_{33}$  (upper graph) and  $\rho_{44}$  (lower graph) on ground state density for different fractions of excited atoms: 1,  $\frac{1}{2}$ ,  $\frac{1}{10}$ ,  $\frac{1}{100}$ . Similarly (b) presents the simulation results for the dependence of  $\rho_{33}$  and  $\rho_{44}$  on the Rabi frequency.

### 4.3.4 Effective Rydberg population

The five-level simulation on superradiance, as it is designed, assumes that all excited atoms, that reside in the effective volume, participate in the cooperative process. This basically requires that all atomic dipoles stay in phase throughout the process. Actually, this assumption is not always true, since there are several mechanisms which can cause a dephasing of the atomic dipoles. Moreover, the simulation assumes the simplified case where the photon, which initiates the superradiant process, always originates from the center of the excitation volume, so that all or most of the excited atoms are involved. However, if the excitation volume is slightly smaller than the emitted wavelength and the initiating photon emerges at the edge of this volume, then the number of atoms contributing to the enhanced emission is only a fraction of the excited. Accordingly, one has to examine modifications of the simulation when only a part of the excited atoms are considered for superradiance. This is presented in figure 4.16. The dependence on ground state density and Rabi frequency is simulated for contribution of all, 50%, 10% and 1% of the excited atoms. Similar to the discussions in the sections 4.3.2 and 4.3.3 the density dependent curves are shifted to higher densities and experience a flattening, while the Rabi frequency dependent curves are compressed.

### 4.3.5 Conclusion

The comparison of the simulation to the experimental results in section 4.2 showed that the simulation does not reproduce the threshold behaviour that was observed in the experiment. In this section we discuss possible reason for this discrepancy.

### Chapter 4. Experimental Results and Simulation

The five-level model was extended by further states with superradiant decays. It was demonstrated that further superradiant decays do not affect the density dependence of the Rydberg population (see section 4.3.1). The simulation can therefore be reduced to a few states. Further, modified decay rates, different effective ground state densities and different effective Rydberg populations were considered. Changing these parameters resulted in a shift of the density curves to higher densities and a "compression" of the Rabi frequency curves (see sections 4.3.2, 4.3.3 and 4.3.4). It was not possible to reproduce the threshold behaviour by changing these parameters.

It is not surprising that the model presented here does not exactly account for the superradiance effects. Indeed, superradiance is a many-body effect and the model is in the single-atom frame, with the superradiance embedded as a mean-field effect. h

# Chapter 5

# Summary and Outlook

### Summary

Within the frame of this thesis it has been demonstrated that it is possible to generate superradiance in a caesium vapour cell when the atoms are continuously pumped to the Rydberg state via the excitation scheme  $6S_{1/2} \rightarrow 7P_{3/2} \rightarrow 32S_{1/2}$ . In the experiment the superradiance was investigated indirectly via the detected fluorescence of the atoms. Nearby states, which are not adressed by the excitation lasers, are populated through superradiant cascading. As a result, additional spectral lines which correspond to spontaneous emission from these states appear in the fluorescence spectra. Intense fluorescence from nF and nD states shows that superradiant cascading strongly populates higher *L*-states.

The fluorescence was measured under the variation of ground state density, the Rabi frequency and the excitation volume. The measurement results presented above reveal that under continuous pumping the superradiance exhibits threshold behaviour as expected from the theory [21]. When the number of cooperative atoms reaches a critical value the spontaneous emission from nearby states rapidly increases while the direct optical fluorescence from the  $32S_{1/2}$  state decrease.

Furthermore, it was demonstrated that it is possible to influence the threshold behaviour of the superradiance by varying the excitation volume. On the basis of the ground state density measurements it was verified that in case of an excitation volume which is smaller than the maximum cooperative volume the critical threshold density is higher. In addition to that the maximum cooperative volume can be determined from density measurements for two different excitation volumes. The condition for this measurement is that the one excitation volume is larger and the other one smaller than the maximum cooperative volume. In the experiment it turned out that more than half of the adjusted beam sizes produced excitation volumes larger than the maximum cooperative volume. If one is interested in a clear imaging of the influence of the excitation volume on the threshold behaviour measurements should also be performed for beam sizes smaller than 1 mm. Chapter 5. Summary and Outlook

## Outlook

The results achieved in this thesis form a first basis for the understanding of superradiant dynamics from S-states in caesium. Now, as the relevant regimes are known further investigations can focus on particular properties of the superradiance. With regard to arrays of vapour cells as quantum modules one could perform measurements on a experimental configuration of two cells that can be addressed by laser beams individually. Such a configuration allows to investigate the interaction between the microwave, that was generated in the first cell, and the Rydberg atoms of the second cell.

Based on the realization of cavity tuned superradiance by Gross *et al* [8] a future idea is to integrate the vapour cells in a microwave-guide. The purpose of this design is to provide selective suppression or amplification of particular superradiant modes. Such a design makes it, for instance, possible to avoid limitations on the Rydberg lifetime due to superradiance. Although the superradiance is quite long known effect it still holds a lot of interesting physics to be investigated.

# Appendix A Diffraction gratings

### A.1 Reflection gratings

An overview on the basic theory of spectrographs and diffraction gratings can be found in [29]. More detailed information on diffraction gratings is given in [30]. Reflection gratings consist of many straight grooves, which have been ruled onto an optically smooth glass substrate or produced by holographic techniques, [30] chapter 3.-4. . The grooves, which have a width that is comparable to the wavelength  $d \sim \lambda$ , act as many small radiation sources. The total reflected light results then in an interference pattern. For a parallel light beam (figure A.1), with the incidence angle  $\alpha$  to the grating normal<sup>1</sup> and wavelength  $\lambda$ , constructive interference is observable for those angles  $\beta$  of the reflected beam for which the grating equation

$$d(\sin\alpha \pm \sin\beta) = m\lambda \tag{A.1}$$

is satisfied, i.e. the path difference  $\Delta s = \Delta s_1 - \Delta s_2$  is a multiple of  $\lambda$ .<sup>2</sup> *m* denotes the diffraction order with  $m = 0, \pm 1, \pm 2, ...$  and is according to eq. (A.1) limited by a finite maximum value  $|m| < 2\frac{d}{\lambda}$ .

From equation (A.1) is obvious that for m = 0, when  $\alpha = -\beta$  and  $\Delta s = 0$ , all components of the zero order light are radiated in the same direction. That means, there is no separation of wavelengths in the zero order. Hence it is called specular reflection. In contrast all other orders can be used to separate light into its constituent wavelengths. However note that the diffraction angle  $\beta$  depends on the incidence angle  $\alpha$  and the groove density G = 1/d.

When the angles of the incident light and the mth-order of the diffracted light are equally with respect to the groove normal, i.e. when incident light and mth-order light

<sup>&</sup>lt;sup>1</sup>Not necessarily normal to the grooves.

<sup>&</sup>lt;sup>2</sup>The plus sign is for the case, when  $\beta$  and  $\alpha$  are on the same side of the gratin normal.





Figure A.1: Diffraction on a reflection grating. a) Illustration of the grating equation (A.1). b) Littrow configuration of a grating with  $\beta = \alpha$ .

describe mirror reflection on the groove facet and

$$\theta_{\rm B} = \frac{\alpha + \beta(m)}{2},\tag{A.2}$$

most of the diffracted energy is gathered in the mth order. The blaze angle  $\theta_{\rm B}$  is the gratings characteristic angle between groove normal and grating normal. The favoured wavelength, called the blaze-wavelength  $\lambda_{\rm B}$ , then is

$$\lambda_{\rm B} = \frac{2d}{m} \sin(\theta_{\rm B}) \cos(\alpha - \theta_{\rm B}), \tag{A.3}$$

which varies with  $\theta_{\rm B}$  and  $\alpha$ . Technically a reflection grating properties are specified by the indication of (A.3) for the case  $\alpha = \theta_{\rm B}$ :

$$\lambda_{\rm B,Litt} = 2d\sin(\theta_{\rm B}).\tag{A.4}$$

This is when  $\alpha = \beta$  and the diffracted light is reflected back into the direction of incident light. Such configuration is called *Littrow mount* (see figure A.1), which is relevant in laser optics. Accordingly, in which wavelength and order most of the diffracted energy is concentrated is determined via the blaze angle  $\theta_{\rm B}$ .

## A.2 Efficiency characteristics

The diffraction efficiency is a quantity that denotes the extent of diffracted energy with respect to the incident energy. Each reflection grating has a wavelength dependent diffraction efficiency which should be accounted for in the designing process of the experiment. The grating efficiency is either specified in relative or absolute efficiency. The absolute efficiency is designated by the ratio of diffracted power P to the incident power  $P_0$ 

$$\eta_{\rm abs} = \frac{P}{P_0},\tag{A.5}$$

while the relative efficiency expresses the diffraction efficiency with respect to the reflectance of the coating material i.e.

$$\eta_{\rm rel} = \frac{P}{P_{\rm coat}} = \frac{P}{\eta_{\rm coat}P_0} = \frac{\eta_{\rm abs}}{\eta_{\rm coat}} \tag{A.6}$$

with  $\eta_{\text{coat}}$  denoting the reflectivity of the coating material. The diffraction efficiency curves are usually given or measured for standard conditions which is the Littrow mount configuration. The characteristic blaze wavelength  $\lambda_{\text{B,Litt}}$  is then the wavelength of maximum efficiency or the peak wavelength. The actual grating efficiency depends on the usage, that means it depends on the incidence angle  $\alpha$  (see eq.(A.3)). Figures A.2 to A.4 illustrate the diffraction efficiencies for the gratings that are available in the experiment.



Figure A.2: Diffraction efficiency for reflection grating 235 lines/mm at 750 nm blaze wavelength. Depicted is the relative grating efficiency with respect to aluminium. Figure from [31].





1575, 600 g/mm, 800 nm, 13.9 deg., M=1, Cat# 53-\*-351, Plane ruled, Max RA 102 x 102 mm

Figure A.3: Diffraction efficiency for reflection grating 600 lines/mm at 800 nm blaze wavelength. Depicted is the relative grating efficiency with respect to aluminium. Figure from [31].



Figure A.4: Diffraction efficiency for reflection grating 600 lines/mm at 500 nm blaze wavelength. Depicted is the absolute grating efficiency. Figure from [31].

# Appendix B Identification of fluorescence lines

Chapter B. Identification of fluorescence lines

Figure B.1: Identification for wavelength area: 490 nm - 590 nm. Vertical lines indicate dipole allowed transitions and each column represents a set of possible transitions. Experimental parameters of spectrum: beam size diameter  $d_{\text{beam},455 \text{ nm}} = 1.0 \text{ mm}$ , ground state density  $\mathcal{N} \sim 3 \times 10^{11} \text{cm}^{-3}$  and rabi frequency  $\Omega_{\rm P}/2\pi = 2.97$  MHz.



Figure B.2: Identification for wavelength area: 590 nm - 710 nm. Vertical lines indicate dipole allowed transitions and each column represents a set of possible transitions. Experimental parameters of spectrum: beam size diameter  $d_{\text{beam},455 \text{ nm}} = 1.0 \text{ mm}$ , ground state density  $\mathcal{N} \sim 3 \times 10^{11} \text{cm}^{-3}$  and rabi frequency  $\Omega_{\rm P}/2\pi = 2.97$  MHz.



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Chapter B. Identification of fluorescence lines

Figure B.3: Identification for wavelength area: 710 nm - 840 nm.Vertical lines indicate dipole allowed transitions and each column represents a set of possible transitions. Experimental parameters of spectrum: beam size diameter  $d_{\text{beam},455 \text{ nm}} = 1.0 \text{ mm}$ , ground state density  $\mathcal{N} \sim 3 \times 10^{11} \text{cm}^{-3}$  and rabi frequency  $\Omega_{\rm P}/2\pi = 2.97$  MHz.



Figure B.4: Identification for wavelength area: 820 nm - 945 nm Vertical lines indicate dipole allowed transitions and each column represents a set of possible transitions. Experimental parameters of spectrum: beam size diameter  $d_{\text{beam},455 \text{ nm}} =$ 1.0 mm, ground state density  $\mathcal{N} \sim 3 \times 10^{11} \text{cm}^{-3}$  and rabi frequency  $\Omega_{\rm P}/2\pi = 2.97$  MHz.



Chapter B. Identification of fluorescence lines

Figure B.5: Identification for wavelength area: 930 nm - 1050 nm.Vertical lines indicate dipole allowed transitions and each column represents a set of possible transitions. Experimental parameters of spectrum: beam size diameter  $d_{\text{beam},455 \text{ nm}} =$ 1.0 mm, ground state density  $\mathcal{N} \sim 3 \times 10^{11} \text{cm}^{-3}$  and rabi frequency  $\Omega_{\rm P}/2\pi = 2.97$  MHz.

Note: The last two peaks in the spectrum are ghost peaks caused by errors occuring from time to time on the CCD chip.



normalized signal

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