Fluorescence spectroscopy of Rydberg atoms in thermal vapor cells in a collision dominated regime

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Stuttgart, den 24. Oktober 2017

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Zusammenfassung

In dieser Abschlussarbeit wird Fluoreszenzspektroskopie von thermischen Cäsiumdampf auf zwei verschiedene Arten realisiert. Dabei wird die Tatsache ausgenutzt, dass der Dampf entlang der Laserpfade in der Dampfzelle leuchtet wenn der Couplinglaser genügend hohe Rabifrequenzen besitzt.

Im ersten Experiment wird der Dampf über einen Zwei-Photonen-Übergang in den $42D_{5/2}$ Rydbergzustand angeregt, wobei der Couplinglaser im Gegensatz zum Probelaser nicht auf Resonanz festgehalten, sondern um diese herum gescannt wird. Dadurch lässt sich der Grenzübergang zwischen leuchtendem und nicht leuchtendem Teil des Cäsiumdampfes räumlich kontrollieren. Zusätzlich wird mittels Mikrowellenstrahlung der Übergang von $42D_{5/2}$ zu $40F_{5/2}$ gescannt, womit sich zeigen lässt, dass lonen in der Zelle während des Experimentes präsent sein müssen, da die Resonanz bei niedrigeren Frequenzen in der Fluoreszenz beobachtet wird, je höher die Rabifrequenz des Probelasers ist. Die Verschiebung wird durch den Stark-Effekt erklärt.

Der Experimente des zweiten Teils dieser Arbeit beschäftigen sich mit dem Prozess der für die Erscheinung von geladenen Teilchen verantwortlich ist. Für diese Experimente werden nun beide Laser auf Resonanz festgehalten (der Probelaser ist nun resonant auf $6S_{1/2} \rightarrow 7P_{3/2}$, der Couplinglaser hingegen auf $7P_{3/2} \rightarrow nD_{5/2}$ mit n = 30, 42, 50), was dazu führt, dass der gesamte Laserpfad in der Zelle leuchtet. Die aufgesammelte Fluoreszenz wird dann mit theoretischen Modellen, von denen das Modell das Ionisation des Dampfes durch Kollisionen mit Elektronen voraussetzt am vielversprechendsten ist, verglichen.

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1. Introduction

During the last decades, there was a constant growth of interest in the research of quantum systems, promising an amplification of computational capacity and efficiency as well as communication using quantum cryptography[Eke91] or sensors for electric, magnetic fields or other quantities like temperature. There are many systems with different approaches like Nitrogen-Vacancy centers in diamond [RTH⁺14], superconducting[USP17] or semi-conducting materials like quantum dots[MM17]. Besides these research fields, atomic physics also has approaches for realizing applications such as quantum gates[HY08].

New effects based on atom-light or atom-atom interactions for example can be investigated with electromagnetically induced transparency (EIT) in coherent media, which can be interpreted as quantum interference in the amplitudes of optical transitions and achieved by exciting atoms with a laser where the coupling exceeds the line width of the bare atom[MF05]. Based on EIT, optical properties in media can be changed and new possibilities for nonlinear optics and quantum information can be explored, particularly in the for ultracold atoms. In case for hot atomic vapors, this kind of research is hardly realized.

The extraordinary properties of highly excited atoms, so called Rydberg atoms, provide a widespread playground in applications for such quantum systems. Especially the field of hot atomic Rydberg atoms in gas phase is showing promising applications like microwave electrometry [JAS2s], since its implementation is quite easy compared to systems using ultracold atoms. Recent studies[WML⁺17] show that cesium atoms in vapor cells can be used as a room-temperature terahertz detector exploiting the effect of optical bistability that is induced by charges[WUR⁺16] instead of the previously assumed dipole-dipole interaction of Rydberg atoms[LHC12]. Additionally, [CRW⁺13] reports on a change in fluorescence that not explicable with the aid of Stark shifted Rydberg states or dipole-dipole interaction alone. Since the emphasis of the research in [WUR⁺16] and [WML⁺17] is concentrated on bistability and not the fluorescence they also mention, this thesis deals with this topic. The fluorescent effect is examined via fluorescence spectroscopy partly using microwave excitation, taking advantage of the fact that Rydberg atoms have large dipole moments which enables them to interact with long-wave radiation resonant to transitions between Rydberg states.

2. Theory

2.1. Rydberg atoms

A major part of this thesis are the experiments on energetically highly excited atoms, so-called Rydberg atoms. Their most significant property is the extremely large orbit of at least one outer electron which results in a large electric dipole moment, whose scaling is proportional to n^2 , where n is describing the principal quantum number of the state. Due to this scaling Rydberg atoms have a high polarizability and as a consequence are highly sensitive to electric fields and are easily ionized. Since the polarizability scales with the sum of the squared dipole matrix elements and the energy difference between to adjacent levels scales with n^{-3} , the polarizability scales with $(n^2)^2/n^{-3} = n^7$.

2.1.1. Energy levels of Alkali atoms

The importance of high values of n did not become clear until Bohr proposed his model of the hydrogen atom back in 1913. Bohr's basis was that an electron orbits its nucleus in a classical circular motion, the quantization of its angular momentum in units of the Planck constant (divided by 2π) \hbar and the fact that an electron only gives off part of its energy in form of radiation when making transitions between two states of defined energy. Radiated energy thus can be calculated as their energy difference, which is dictated by their principal quantum numbers, where for one atom the energy can be written as[Gal05]:

$$E_n = -\frac{e^4 m_{\rm e}}{32\pi^2 \epsilon_0^2 \hbar^2} \frac{1}{n^2}$$
(2.1.1)

The prefactor, containing elementary charge e and its mass m_e , vacuum permittivity ϵ_0 and also the Planck constant \hbar , is summed up as the Rydberg energy:

$$R_{\infty} = \frac{e^4 m_{\rm e}}{32\pi^2 \epsilon_0^2 \hbar^2} \approx 13.6 \,{\rm eV} \tag{2.1.2}$$

According to Bohr's model, the radius of the orbit can be calculated to

$$r = \frac{4\pi\epsilon_0\hbar^2}{e^2m_{\rm e}}n^2 = a_0n^2,$$
(2.1.3)

with $a_0 = 0.529$ Å as the Bohr radius. Going beyond Bohr's model and considering that the electron behaves like a wave, the expectation value of the radius is $\langle r \rangle = 3/2a_0n^2$ after integration over the radial probability density.

2.1.2. The quantum defect

Equation (2.1.1) describes the ideal case for the hydrogen atom that has no inner electrons and only one outer electron. Since there is no dependency on the orbital angular momentum l (3s, 3p and 3d configurations have the same energies) the energy states in hydrogen are degenerate. However, in case of a different alkali atom the inner electrons rescind this degeneracy since the 1/r-potential becomes modified for smaller distances to the nucleus[Foo11], which basically means that the wave function of the outer electron penetrates and polarizes the inner electron shell. This effect is most significant for s-electrons and can be neglected for states with l > 3. As a consequence, Bohr's formula for the energy levels has to be modified for Alkali atoms in terms of an effective principal quantum number:

$$E_{nlj} = -\frac{R}{\left(n^*\right)^2} = -\frac{R}{\left(n - \delta_{nlj}\right)^2}$$
(2.1.4)

In equation (2.1.4) $R = R_{\infty} \cdot M/(M + m_e)$ is the mass-corrected Rydberg constant with M as the atomic mass of the nucleus and δ_{nlj} is the quantum defect (n, l, j are quantum numbers) which describes the increase in binding energy for an Alkali atom relatively to hydrogen. The quantum defect is calculated as

$$\delta_{nlj} = \sum_{i=0,2,..} \frac{\delta_i}{(n-\delta_0)^i},$$
(2.1.5)

while the δ_i itself have to be measured experimentally. Table 2.1.1 shows values of these coefficients



Figure 2.1.1.: Energy level scheme of the $nS_{1/2}$, $nP_{3/2}$ and $nD_{5/2}$ states in cesium according to equation (2.1.4).

Table 2.1.1.: Coefficients δ_i for cesium of the nS, nP, nD and nF states found in [LN84] and [WS87]. During the calculation of the levels of cesium depicted in figure 2.1.1 the presented coefficients and the mass-correction factor $M/(M + m_e) = 0.999\,995\,87$ are used.

State	$nS_{1/2}$	$nP_{1/2}$	$nP_{3/2}$	$nD_{3/2}$	$nD_{5/2}$	$nF_{5/2}$
δ_0	4.04935665	3.59158950	3.5589599	2.4754562	2.46631524	0.03341424
δ_2	0.2377037	0.360926	0.392469	0.009320	0.013577	-0.198674
δ_4	0.255401	0.41905	-0.67431	-0.43498	-0.37457	0.28953
δ_6	0.00378	0.64388	22.353	-0.76358	-2.1867	-0.2601
δ_8	0.25486	1.45035	-92.289	-18.0061	-1.5532	
δ_{10}					-56.6739	

for the nS, nP, nD and nF states in cesium. For l > 2, the quantum defect δ_{nlj} does not have an huge effect on the energy levels of the Rydberg atoms anymore. Moreover, it can be seen according to the formula for δ_{nlj} (2.1.5) that the influence of n on δ_{nlj} is only of minor nature. Following, with the coefficient in table 2.1.1 depicted, the energy levels of alkali atoms can be calculated in respect to the ionization continuum which is shown in figure 2.1.1 for the energies of $nS_{1/2}$, $nP_{3/2}$ and $nD_{5/2}$ in cesium.

2.1.3. Stark effect

If an atom is surrounded by an electric field, the coupling between field and atom has to be considered. For an atom in a DC electric field \mathcal{E} defined along an axis (commonly the *z*-axis) this sort of coupling is expressed by the Hamiltonian

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + e\mathcal{E}z\boldsymbol{e}_z, \qquad (2.1.6)$$

with $\hat{\mathcal{H}}_0$ as the Hamiltonian for unperturbed states. Diagonalization of $\hat{\mathcal{H}}$ yields energy eigenvalues shifted relatively to the unperturbed states, called the Stark effect. In the vicinity of low electric fields pertubation theory is a useful tool for calculating energy shifts. Considering the quantum defects of the Alkali atoms, it has to be noted that for l < 3, the first order Stark shift vanishes leaving a second order Stark shift

$$\Delta_{\mathsf{Stark}} = -\frac{1}{2}\alpha_0 \mathcal{E}^2, \tag{2.1.7}$$

with α_0 as polarizability for the state $|nljm_j\rangle$:

$$\alpha_0 = 2e^2 \sum_{n'l'j' \neq nlj} \frac{\left| \langle nljm_j | r | n'l'j'm_j' \rangle \right|^2}{E_{n'l'j'} - E_{nlj}}$$
(2.1.8)

From that expression the n^7 scaling of the polarizability can be derived since r scales with n^2 and $E_{n'l'j'} - E_{nlj}$ scales with $\propto 1/(n+1)^2 - 1/n^2 \propto 1/n^3$.

2.2. Atom-Light Interactions

This chapter deals with interactions of an atom with a light field in a three-level atom. First, a model for the three-level atom is introduced including its Hamiltonian in an approximated form. Further, density matrix formalism is used together with the Liouville-von-Neumann equation to describe the time-evolution of the system. As a last point of this section effects coupled to the motion of the atom are explained shortly.

2.2.1. Three-Level System

For the underlying discussion about atom-light interactions the three-level system is chosen as an example. Figure 2.2.1 depicts such a three-level system schematically. It consists of a ground, an intermediate and excited state separated by $\hbar\omega_{0_{g,i}}$ and $\hbar\omega_{0_{i,e}}$ in energy space. By defining the basis [Ste]

$$|\mathbf{g}\rangle \equiv \begin{pmatrix} 1\\0\\0 \end{pmatrix}, |\mathbf{i}\rangle \equiv \begin{pmatrix} 0\\1\\0 \end{pmatrix}, |\mathbf{e}\rangle \equiv \begin{pmatrix} 0\\0\\1 \end{pmatrix}, \qquad (2.2.1)$$

the atomic Hamiltonian then can be written as

$$\hat{\mathcal{H}}_{a} = \hbar \left(\omega_{0_{g,i}} \left| i \right\rangle \left\langle i \right| + \omega_{0_{i,e}} \left| e \right\rangle \left\langle e \right| \right).$$
(2.2.2)

Considering the interaction with an electromagnetic light field (here: plane wave), where



Figure 2.2.1.: Schematic for the three-level atom. Separations of the ground $|g\rangle$, intermediate $|i\rangle$ and excited $|e\rangle$ state are denoted by $\omega_{0_{g,e}}$ and $\omega_{0_{i,e}}$. Also shown are the detunings $\Delta_{g,i}$ and $\Delta_{i,e}$ induced by the angular frequencies for the probe $\omega_{L_{g,i}}$ and the coupling $\omega_{L_{i,e}}$ laser.

$$\boldsymbol{E}_{g,i}(t) = \frac{1}{2} \left(E_{0_{g,i}} e^{i\omega_{L_{g,i}}t} + E_{0_{g,i}}^* e^{-i\omega_{L_{g,i}}t} \right)$$
(2.2.3)

$$\boldsymbol{E}_{i,e}(t) = \frac{1}{2} \left(E_{0_{i,e}} e^{i\omega_{L_{i,e}}t} + E_{0_{i,e}}^* e^{-i\omega_{L_{i,e}}t} \right)$$
(2.2.4)

denote electric fields driving $|g\rangle \rightarrow |i\rangle$ (further denoted as probe transition) and $|i\rangle \rightarrow |e\rangle$ (further denoted as coupling transition), the interaction Hamiltonian reads

$$\mathcal{H}_{int} = -d_{g,i} \boldsymbol{E}_{g,i}(t) |g\rangle \langle i| - d_{i,g} \boldsymbol{E}_{g,i}(t) |i\rangle \langle g| - d_{i,e} \boldsymbol{E}_{i,e}(t) |e\rangle \langle i| - d_{e,i} \boldsymbol{E}_{i,e}(t) |i\rangle \langle e|,$$

$$(2.2.5)$$

with

$$\boldsymbol{d}_{j,k} = e\left\langle j \,|\, r \,| k \right\rangle \tag{2.2.6}$$

as the electric dipole matrix element of the respective transition. Consequently, using equations (2.2.2) and (2.2.5), the total Hamiltonian for the three-level system is

$$\hat{\mathcal{H}}_{3L} = \hat{\mathcal{H}}_{a} + \hat{\mathcal{H}}_{int}.$$
 (2.2.7)

2.2.2. Time-evolution of the three-level system: Rotating-wave approximation and density matrix

For further use, the total Hamiltonian is simplified by transformation into a rotating frame oscillating with the same frequency as the light field. This is done by

$$\hat{\mathcal{H}}_{3L,rot} = \hat{U}^{\dagger} \hat{\mathcal{H}}_{3L} \hat{U} - i\hbar \hat{U}^{\dagger} \frac{\partial \hat{U}}{\partial t}, \qquad (2.2.8)$$

with

$$\hat{\boldsymbol{U}} = \mathrm{e}^{-i\hat{\boldsymbol{A}}t/\hbar} \tag{2.2.9}$$

as the transformation matrix and

$$\hat{A} = \hbar(\omega_{\mathsf{L}_{\mathsf{g},\mathsf{i}}} |\mathsf{i}\rangle \langle \mathsf{i}| + (\omega_{\mathsf{L}_{\mathsf{g},\mathsf{i}}} + \omega_{\mathsf{L}_{\mathsf{i},\mathsf{e}}}) |\mathsf{e}\rangle \langle \mathsf{e}|).$$
(2.2.10)

In the rotating frame, fast oscillating terms are neglected, which is called the Rotating-Wave approximation and justified by the fact that their mean value is zero. Following the Rotating-Wave approximated Hamiltonian is obtained:

$$\hat{\mathcal{H}}_{3\mathsf{L},\mathsf{RWA}} = \hbar \left(\frac{\Omega_{\mathsf{g},\mathsf{i}}^{*}}{2} |\mathsf{i}\rangle \langle \mathsf{g}| + \frac{\Omega_{\mathsf{g},\mathsf{i}}}{2} |\mathsf{g}\rangle \langle \mathsf{i}| + \frac{\Omega_{\mathsf{i},\mathsf{e}}^{*}}{2} |\mathsf{e}\rangle \langle \mathsf{i}| + \frac{\Omega_{\mathsf{i},\mathsf{e}}}{2} |\mathsf{i}\rangle \langle \mathsf{e}| \right) \\ + \hbar \left(-\Delta_{\mathsf{g},\mathsf{i}} |\mathsf{i}\rangle \langle \mathsf{i}| - \left(\Delta_{\mathsf{g},\mathsf{i}} + \Delta_{\mathsf{i},\mathsf{e}} \right) |\mathsf{e}\rangle \langle \mathsf{e}| \right)$$
(2.2.11)

In this expression $\Omega_{j,k} \equiv -d_{j,k}E_{0_{j,k}}/\hbar$ and $\Omega_{j,k}^* \equiv -d_{k,j}E_{0_{j,k}}/\hbar$ denote the Rabi frequencies and $\Delta_{g,i} \equiv \omega_{L_{g,i}} - \omega_{0_{g,i}}$ and $\Delta_{i,e} \equiv \omega_{L_{i,e}} - \omega_{0_{i,e}}$ are the detunings of the driving fields. By defining the saturation intensity

$$I_{\mathsf{sat}} \equiv \frac{2\pi^2 c \Gamma \hbar}{3\lambda^3},\tag{2.2.12}$$

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the Rabi frequency of a certain transition can be rewritten in terms of I_{sat} :

$$\Omega = \Gamma \sqrt{\frac{I}{2I_{\mathsf{sat}}}} \tag{2.2.13}$$

In this expression $I=c\epsilon_0E_0^2/2$ is the light intensity with c as the speed of light in vacuum. With the ansatz for the wave function

$$|\Psi\rangle = \sum_{j=\mathsf{g},\mathsf{i},\mathsf{e}} c_j(t) |j\rangle, \, c_j(t) \in \mathbb{C},$$
(2.2.14)

the Schrödinger equation for the three-level system can be solved. However, when it comes to describing the time-evolving population of the three levels including decays, it is useful to use the density matrix which is defined as

$$\hat{oldsymbol{
ho}} = \ket{\Psi}ig\langle\Psi|$$
. (2.2.15)

An explicit elaboration on the density matrix formalism can be found in [MvdS99]. For the three-level system, diagonal elements of $\hat{\rho}$ describe the population, whereas the off-diagonal elements characterize coherences. In the further discussion, the density matrix also has to be transformed into the same rotating frame: $\tilde{\rho} = \hat{U}^{\dagger} \hat{\rho} \hat{U}$. The time-evolution is now calculated by solving the Liouville-von-Neumann equation:

$$\frac{\partial \tilde{\boldsymbol{\rho}}}{\partial t} = -\frac{i}{\hbar} \left[\hat{\boldsymbol{\mathcal{H}}}_{3L,RWA}(t), \tilde{\boldsymbol{\rho}}(t) \right] + \hat{\boldsymbol{\mathcal{L}}} \left(\tilde{\boldsymbol{\rho}}(t) \right)$$
(2.2.16)

The Lindblad operator[Lin76]

$$\hat{\boldsymbol{\mathcal{L}}} = \sum_{j,k=\mathsf{g},\mathsf{i},\mathsf{e}} \Gamma_{j,k} \left(C_{j,k} \tilde{\boldsymbol{\rho}} C_{j,k}^{\dagger} - \frac{1}{2} \left[C_{j,k} C_{j,k}^{\dagger}, \tilde{\boldsymbol{\rho}} \right] \right)$$
(2.2.17)

in equation (2.2.16) additionally accounts for the decay processes from the intermediate and excited state with the decay rates $\Gamma_{i,g}$ and $\Gamma_{e,i}$ with the operators

$$C_{i,g} = \sqrt{\Gamma_{i,g}} |g\rangle \langle i|, \qquad (2.2.18)$$

$$C_{\mathsf{e},\mathsf{i}} = \sqrt{\Gamma_{\mathsf{e},\mathsf{i}}} \left| i \right\rangle \left\langle e \right|. \tag{2.2.19}$$

Now, in the written out form

$$\frac{\partial}{\partial t}\tilde{\rho}_{\mathsf{g},\mathsf{g}} = -\operatorname{Im}(\tilde{\rho}_{\mathsf{g},\mathsf{i}}\Omega_{\mathsf{g},\mathsf{i}}^{*}) + \tilde{\rho}_{\mathsf{i},\mathsf{i}}\Gamma_{\mathsf{i},\mathsf{g}}
\frac{\partial}{\partial t}\tilde{\rho}_{\mathsf{g},\mathsf{i}} = \frac{i}{2}\left(\tilde{\rho}_{\mathsf{g},\mathsf{e}}\Omega_{\mathsf{i},\mathsf{e}}^{*} - (\tilde{\rho}_{\mathsf{i},\mathsf{i}} - \tilde{\rho}_{\mathsf{g},\mathsf{g}})\Omega_{\mathsf{g},\mathsf{i}}\right) - \tilde{\rho}_{\mathsf{g},\mathsf{i}}\left(\frac{\Gamma_{\mathsf{i},\mathsf{g}}}{2} + i\Delta_{\mathsf{g},\mathsf{i}}\right)
\frac{\partial}{\partial t}\tilde{\rho}_{\mathsf{g},\mathsf{e}} = \frac{i}{2}\left(\tilde{\rho}_{\mathsf{g},\mathsf{i}}\Omega_{\mathsf{i},\mathsf{e}} - \tilde{\rho}_{\mathsf{i},\mathsf{e}}\Gamma_{\mathsf{g},\mathsf{i}}\right) - \tilde{\rho}_{\mathsf{g},\mathsf{e}}\left(\frac{\Gamma_{\mathsf{e},\mathsf{i}}}{2} + i\left(\Delta_{\mathsf{g},\mathsf{i}} + \Delta_{\mathsf{i},\mathsf{e}}\right)\right)
\frac{\partial}{\partial t}\tilde{\rho}_{\mathsf{i},\mathsf{i}} = \operatorname{Im}(\tilde{\rho}_{\mathsf{g},\mathsf{i}}\Omega_{\mathsf{g},\mathsf{i}}^{*}) - \operatorname{Im}(\tilde{\rho}_{\mathsf{i},\mathsf{e}}\Omega_{\mathsf{i},\mathsf{e}}^{*}) - \tilde{\rho}_{\mathsf{i},\mathsf{i}}\Gamma_{\mathsf{i},\mathsf{g}} + \tilde{\rho}_{\mathsf{e},\mathsf{e}}\Gamma_{\mathsf{e},\mathsf{i}}
\frac{\partial}{\partial t}\tilde{\rho}_{\mathsf{i},\mathsf{e}} = -\frac{i}{2}\left((\tilde{\rho}_{\mathsf{e},\mathsf{e}} - \tilde{\rho}_{\mathsf{i},\mathsf{i}})\Omega_{\mathsf{i},\mathsf{e}} + \tilde{\rho}_{\mathsf{g},\mathsf{e}}\Omega_{\mathsf{g},\mathsf{i}}^{*}\right) - \tilde{\rho}_{\mathsf{i},\mathsf{e}}\left(\frac{\Gamma_{\mathsf{i},\mathsf{g}}}{2} + \frac{\Gamma_{\mathsf{e},\mathsf{i}}}{2} + i\Delta_{\mathsf{i},\mathsf{e}}\right)
\frac{\partial}{\partial t}\tilde{\rho}_{\mathsf{e},\mathsf{e}} = \operatorname{Im}(\tilde{\rho}_{\mathsf{i},\mathsf{e}}\Omega_{\mathsf{i},\mathsf{e}}^{*}) - \tilde{\rho}_{\mathsf{e},\mathsf{e}}\Gamma_{\mathsf{e},\mathsf{i}}, \tag{2.2.20}$$

the optical Bloch-equations are obtained. In regard of an external electric field applied to the whole system, the Stark effect has to be taken into account also. Fortunately, this objective is achieved rather easily by adding the Stark shift to the detunings already created by off-resonant driving, so that the optical Bloch equations maintain their structure.

2.2.3. Doppler effect

Up until now, the discussion of atom-light interaction did not include motional effects. By assuming an atom in an ideal gas with Maxwell-Boltzmann distributed velocity v, it has to be considered that the atoms see a shifted frequency ω'

$$\omega' = \omega_{\mathsf{L}} + \omega_{\mathsf{L},\mathsf{d}} = \omega_{\mathsf{L}} \pm \mathbf{k}_{\mathsf{L}} \cdot \mathbf{v} = \omega_{\mathsf{L}} \mp k_{\mathsf{L}} v_{||} \tag{2.2.21}$$

when excited with by light of frequency $\omega_{\rm L}$ and wave vector $\mathbf{k}_{\rm L}$, which is due to the Doppler effect. The atom sees a blue shift if it is moving in the opposite direction of the beam. In equation (2.2.21) the scalar product $\mathbf{k}_{\rm L} \cdot \mathbf{v} = k_{\rm L} v_{||}$ since only velocity components $v_{||}$ parallel to $\mathbf{k}_{\rm L}$ contribute to this effect. As a consequence only the Maxwell-Boltzmann distribution in one dimension is needed:

$$N(v_{||},T) = \sqrt{\frac{m}{2\pi k_{\mathsf{B}}T}} e^{-\frac{mv_{||}^2}{2k_{\mathsf{B}}T}}$$
(2.2.22)

By defining the Doppler detuning $\delta_d = -k_L v_{\parallel}$ and substituting it into equation (2.2.22) the Doppler width (full width at half maximum: FWHM) can be calculated to

$$\Gamma_{\rm d} = |k_{\rm L}| \sqrt{\frac{8\ln(2)k_{\rm B}T}{m}}.$$
(2.2.23)

In case for a three-level atom both detunings have to be rewritten in terms of the Doppler effect:

$$\Delta'_{g,i} = \Delta_{g,i} - \Delta_{g,i_d} = \Delta_{g,i} - k_{\mathsf{L}_{g,i}} v_{||}$$
(2.2.24)

$$\Delta'_{i,e} = \Delta_{i,e} - \Delta_{i,e_d} = \Delta_{i,e} - k_{\mathsf{L}_{i,e}} v_{||}$$
(2.2.25)

Residual Doppler effect With the assumption that both lasers are driving the three-level system resonantly, the maximal velocity, an atom is allowed to have for successful excitation, can be determined. For the underlying three-level atom this velocity can be calculated by the Doppler shift:

$$k_{g,i}v_{\max} \stackrel{!}{=} \Gamma_{i,g} \tag{2.2.26}$$

$$\Rightarrow v_{\max} = \frac{k_{g,i}}{\Gamma_{i,g}}$$
(2.2.27)

The resulting line width for |e
angle
ightarrow |i
angle is then

$$\Gamma_{e,i} = \frac{k_{i,e}}{k_{g,i}} \Gamma_{i,g}.$$
(2.2.28)

In case for two counter propagating light fields (2.2.28) becomes

$$\Gamma_{\mathbf{e},\mathbf{i}} = \left(1 - \frac{k_{\mathbf{i},\mathbf{e}}}{k_{\mathbf{g},\mathbf{i}}}\right)\Gamma_{\mathbf{i},\mathbf{g}}.$$
(2.2.29)

As a consequence for resonant excitation into the excited state $|e\rangle$ the condition $\Gamma_{\rm e,i} > \Delta_{\rm i,e}$ has to be fulfilled, meaning that even for $\Delta_{\rm i,e} > 0$ the atom gets resonantly excited due to Doppler broadening. Since this thesis revolves around experiment on hot atomic gases, this fact should be considered

2.3. Bistability

In an open quantum system of atoms which is collectively driven and is emitting radiation spontaneously, the system itself occasionally jumps from a state with low Rydberg population to a high population state and vice versa. Thus one can use the term bistability to describe such a system. Essential for bistable behavior are a large number of atoms which means that these sudden changes in Rydberg population are inherently collective and do not emerge otherwise[LHC12].

2.3.1. Theoretical model

In the theoretical model a system of N two-level atoms continuously driven to a Rydberg state $|\mathbf{r}\rangle_i$ from the ground state $|\mathbf{g}\rangle_i$ (*i* denotes the *i*-th atom of the system) is considered. The resulting rotating-wave approximated Hamiltonian is

$$\hat{\mathcal{H}}_{\mathsf{bi}} = \hbar \sum_{i} \left(-\Delta \left| \mathsf{r} \right\rangle \left\langle \mathsf{r} \right|_{i} + \frac{\Omega_{\mathsf{R}}}{2} \left(\left| \mathsf{r} \right\rangle \left\langle \mathsf{g} \right|_{i} + \left| \mathsf{g} \right\rangle \left\langle \mathsf{r} \right|_{i} \right) \right) + \frac{\hbar \Delta_{\mathsf{Stark}}}{N-1} \sum_{i < k} \left| \mathsf{r} \right\rangle \left\langle \mathsf{r} \right|_{i} \otimes \left| \mathsf{r} \right\rangle \left\langle \mathsf{r} \right|_{k}, \qquad (2.3.1)$$

where Ω_R is the Rabi frequency resulting from non-resonant driving with detuning Δ . Δ_{Stark} considers a Stark shift as the cause of the energy shift of the atoms [WUR⁺16] instead of the previously assumed dipole-dipole interaction[dMWv⁺16]. Since there are no correlations between the atoms in this model[LHC12], the density matrix for the system factorizes by atom and as a result the Lindblad master equation describes the time-evolution of the population and coherences as[dMWv⁺16],

$$\frac{\partial}{\partial t}\tilde{\rho}_{\mathsf{r},\mathsf{g}} = i\Omega_{\mathsf{R}}\left(\tilde{\rho}_{\mathsf{r},\mathsf{r}} - \frac{1}{2}\right) + i\Delta_{\mathsf{eff}}\tilde{\rho}_{\mathsf{g},\mathsf{r}} - \frac{\Gamma_{\mathsf{r},\mathsf{g}}}{2}\tilde{\rho}_{\mathsf{g},\mathsf{r}}$$
(2.3.2)

$$\frac{\partial}{\partial t}\tilde{\rho}_{\mathbf{r},\mathbf{r}} = -\Omega_{\mathsf{R}}\operatorname{Im}\left(\tilde{\rho}_{\mathsf{g},\mathsf{r}}\right) - \Gamma_{\mathsf{r},\mathsf{g}}\tilde{\rho}_{\mathsf{r},\mathsf{r}}$$
(2.3.3)

where $\Gamma_{r,g}$ is the line width of the transition from the ground to the excited Rydberg state. In equation (2.3.2) the effective detuning includes the Stark shift Δ_{Stark} and $\tilde{\rho}_{r,r}$ amounts for the fraction of atomic population in the Rydberg state:

$$\Delta_{\rm eff} = \Delta + \Delta_{\rm Stark} \tilde{\rho}_{\rm r,r}, \qquad (2.3.4)$$



Figure 2.3.1.: Stable (solid lines) and unstable (dashed, non-vertical) solutions for the Rydberg population $\tilde{\rho}_{r,r}$ plotted against the detuning of the driving light field. In the bistable regime $\tilde{\rho}_{r,r}$ depends on the scan direction of the detuning. The curve was obtained by plotting the solutions of equation 2.3.5 with $\Omega_{\rm R} = 3000$, $\Gamma_{r,g} = 3 \cdot 10^{-5}$ and $\Delta_{\rm Stark} = 20000$ (arbitrarily chosen).

The underlying Stark shift in this system, is said to be caused by ions created through ionization processes of the excited Rydberg atoms[WUR⁺16]. By solving the OBEs (2.3.2) and (2.3.3) in the steady-state a cubic equation for the population of the Rydberg state is obtained[dMWv⁺16]

$$\frac{\Omega_{\mathsf{R}}^2}{4} - \left(\frac{\Omega_{\mathsf{R}}^2}{2} + \frac{\Gamma_{\mathsf{r},\mathsf{g}}^2}{4} + \Delta^2\right)\tilde{\rho}_{\mathsf{r},\mathsf{r}} - 2\Delta\Delta_{\mathsf{Stark}}\tilde{\rho}_{\mathsf{r},\mathsf{r}}^2 - \Delta_{\mathsf{Stark}}^2\tilde{\rho}_{\mathsf{r},\mathsf{r}}^3 = 0,$$
(2.3.5)

which points out the non-linear behavior of the population in form of two additional terms of quadratic and cubic order of $\tilde{\rho}_{r,r}$ compared to the case when no energy shift is considered.

2.3.2. Bistable behavior of the Rydberg population

Solving (2.3.5) for $\tilde{\rho}_{r,r}$ provides three solutions of which one is unstable and two are stable corresponding to the bistable behavior of the system. In figure 2.3.1 these three solutions are shown graphically. In case the transition is scanned from higher to lower frequencies, the population of the Rydberg state builds up which in turn sustains the ability to excite atoms into the Rydberg state even in the off-resonant regime. However, if the coupling detuning is far too off-resonant the decay mechanics outweigh the nonlinear energy shift and as a result the population drops down instantly

(see fig. 2.3.1 vertical blue dashed line). By scanning in the other direction from lower to higher frequencies, $\tilde{\rho}_{r,r}$ maintains a low value even in the bistable regime. Only when the coupling detuning reaches a point at which a sufficient excitation into the Rydberg state secured, a sudden increase in population is triggered (see fig. 2.3.1 vertical yellow dashed line). As a result the population has a hysteresis-like behavior.

2.4. Plasma physics

Since it is assumed that optical bistability is induced by charges, it is of necessity to also cover the aspect of ionized gases, i.e. a plasma. Plasmas mainly consist of free electrons and positive ions. They behave like a charge conducting fluids interacting with electromagnetic fields, and as a consequence showing collective behavior. When viewed as a macroscopic system the net charge is zero, where the term 'quasi-neutrality' then can be used (although on microscopic length scales this is not the case). In this chapter basic terms and criteria, which describe an ionized gas, and should be fulfilled in order that the system can be called a plasma, are introduced together with the Holtsmark distribution which comes in handy when describing the electric microfield emerging from ions of an ionized gas.

2.4.1. Properties of a plasma

Debye-shielding In plasma physics it is useful to look at the charge distributions/densities within the ionized gas. Inside of an ideal plasma, the number densities of positive ions n_i and electrons n_e are approximately the same since plasmas have to be electrically neutral as a whole. Even within the plasma large deviations between n_i and n_e are less likely since each separation of charges, which is induced by relative displacement of a group of electrons from a group of positive ions, produces high electric fields which then on the other hand act as an attractive force between these two groups. By going a step further and looking upon the system in a microscopic regime, it has to be mentioned that there are small deviations from charge neutrality. This is caused by the fact that for example positive ions act repulsively on other positively charged ions, but on the other hand attract electrons. As a result, positive ions amass electrons around themselves like it is depicted in figure 2.4.1a. In statistical equilibrium the number densities are $n_{i,e} = n \cdot exp (\mp e\varphi/k_B T_{i,e})$, where n is the density at large distances from the charge where the potential φ vanishes, $k_{\rm B}$ is the Boltzmann constant and $T_{\rm i}$ and $T_{\rm e}$ are the temperatures of positive ions and electrons. By assuming that the mean potential energy between particles of a plasma and their nearest neighbor is small when compared with their kinetic energy $(e\varphi/k_{\rm B}T_{\rm i,e}\ll 1)$ the number densities can be evolved geometrically. With that the Poisson equation in spherical coordinates then can be written as[Sch93]

$$\Delta \varphi = \nabla^2 \varphi = \frac{1}{r^2} \frac{\mathrm{d}}{\mathrm{d}r} \left(r^2 \frac{\mathrm{d}\varphi}{\mathrm{d}r} \right)$$
$$= \frac{e \left(n_{\mathsf{e}} - n_{\mathsf{i}} \right)}{\epsilon_0} = \frac{n e^2 \varphi}{\epsilon_0 k_{\mathsf{B}}} \left(\frac{1}{T_{\mathsf{e}}} + \frac{1}{T_{\mathsf{i}}} \right). \tag{2.4.1}$$



Figure 2.4.1.: a) Positive charge surrounded by a cloud of electrons. b) Potentials of a positive charge in vacuum (dark blue dashed curve) and in a plasma (red curve). In a plasma the potential drops much faster due to Debye-shielding.

Now, by defining the Debye lengths $\lambda_{d_{i,e}}$

$$\lambda_{\mathsf{d}_{\mathsf{i},\mathsf{e}}} = \sqrt{\frac{\epsilon_0 k_{\mathsf{B}} T_{\mathsf{i},\mathsf{e}}}{ne^2}} \tag{2.4.2}$$

and the total Debye length $\lambda_d^{-2} = \lambda_{d_e}^{-2} + \lambda_{d_i}^{-2}$ the Poisson equation (2.4.1) can be rewritten into

$$\frac{1}{r^2} \frac{\mathrm{d}}{\mathrm{d}r} \left(r^2 \frac{\mathrm{d}\varphi}{\mathrm{d}r} \right) = \lambda_{\mathsf{d}}^{-2} \varphi, \qquad (2.4.3)$$

with the Debye-Hückel potential[Str11]

$$\varphi(r) = \frac{q}{4\pi\epsilon_0} \frac{1}{r} e^{-\frac{r}{\lambda_d}}$$
(2.4.4)

as its solution. In detail the potential (2.4.4) explains how the potential of a charge q drops within the plasma. Such behavior can be seen in figure 2.4.1b which shows that the potential has a faster drop in the plasma than in the vacuum. After λ_d , the potential has dropped down to a value of $q/4\pi\epsilon_0 e$ (here e refers to Euler's number and not to the elementary charge), meaning that it is almost completely shielded. This process, in which a charge q is surrounded by a cloud of opposite charged particles and that has a radius of the size of the Debye length (also referred to as the Debye sphere), is called Debye shielding[Sch93]. A noteworthy conclusion from the Debye-Hückel potential is that the Debye shielding of denser plasmas is more effective compared to the ones with a lower density, which means that λ_d decreases. Further, a decreasing temperature also promotes a decreasing Debye length since trajectories of slow particles can be easier deflected by charges[Str11]. Now it has to be considered that the particles within a plasma have finite velocities, and because of that positive ions are not able to amass a cloud of electrons around themselves all the time. At this point quasi-neutrality is defined as follows: If the dimensions of a plasma L become much larger than λ_d , meaning local accumulation of charges, said charges become shielded on length scale much shorter than L. As a result the plasma is quasi-neutral and the approximation $n_i \simeq n_e \simeq n$ (n is now the density of the plasma itself) becomes effective. This leads to the first criterion for an ionized gas to be a plasma: $\lambda_d \ll L$.

Plasma parameter Another important physical quantity is the number of particles within the Debye sphere N_d :

$$N_{\rm d} = n \frac{4\pi}{3} \lambda_{\rm d} = \frac{4\pi}{3} \frac{\left(\frac{\epsilon_0 k_{\rm B} T}{e^2}\right)^{3/2}}{n^{1/2}}$$
(2.4.5)

By using the mean distance r_0 between particles of density n, where

$$\frac{4\pi}{3}r_0^3 = n^{-1},\tag{2.4.6}$$

 N_{d} can be rewritten in terms of λ_{d} and r_{0} [Sch93]:

$$N_{\rm d} = \left(\frac{\lambda_{\rm d}}{r_0}\right)^3 \tag{2.4.7}$$

This provides the second criterion for an ionized gas to be a plasma: The collective behavior, which is characteristic to a plasma, requires that the number of particles within the Debye sphere have to be much higher than 1 [Str11]:

$$N_{\rm d} \gg 1 \tag{2.4.8}$$

Plasma frequency While deriving the Debye length, the restricted conditions where of stationary nature. But considering the finite velocity of the plasma particles, the next step is to examine how fast the particles have to be in order to guarantee efficient shielding, or in other words: Up to which frequency are external time varying electric fields shielded by the plasma?

This is done by introducing the plasma frequency ω_p which emerges from deviations of quasineutrality in the microscopic regime. When electrons are shifted spatially relative to positive ions an electric field emerges[Sch93]:

$$E \simeq \frac{nex}{\epsilon_0} \tag{2.4.9}$$

Due to this field, the electrons experience an attractive force towards the positive ions. For such a system the ordinary differential equation (ODE) is

$$m_{\rm e} \frac{{\rm d}^2 x}{{\rm d}t^2} = -eE = -\frac{ne^2 x}{\epsilon_0},$$
 (2.4.10)

of which the solution also consists of the plasma frequency for electrons

$$\omega_{\mathsf{p}_{\mathsf{e}}} = \sqrt{\frac{ne^2}{\epsilon_0 m_{\mathsf{e}}}}.$$
(2.4.11)

By using the approximation $v_{\text{th,e}} \simeq \sqrt{k_{\text{B}}T_{\text{e}}/m_{\text{e}}}$, the thermal velocity of the electrons $v_{\text{th,e}}$ also can be calculated with λ_{d} and $\omega_{\text{p}_{e}}$:

$$v_{\rm th,e} \simeq \lambda_{\rm d} \omega_{\rm p_e}$$
 (2.4.12)

In the case for heavier positive ions, it has to be considered, that due to their inertia, the positive ions almost do not contribute to the total plasma frequency. Nonetheless, in analogy to equation (2.4.11) the plasma frequency for ions can be defined as[Sch93]

$$\omega_{p_{\rm i}} = \sqrt{\frac{n_{\rm i}Z^2e^2}{\epsilon_0 m_{\rm i}}},\tag{2.4.13}$$

where n_i is the number density of the positive ions of net charge Ze and mass m_i . Together with ω_{p_a} the total plasma frequency is

$$\omega_{\mathbf{p}}^{2} = \omega_{p_{e}}^{2} + \omega_{\mathbf{p}_{i}}^{2}.$$
(2.4.14)

With the help of ω_p the third criterion for an ionized gas to be a plasma can be formulated, namely the product of plasma frequency and collision time of plasma particles with neutral ones has to be higher than 1 [Sch93]:

$$\omega_{\mathsf{p}}\tau > 1 \tag{2.4.15}$$

2.4.2. Ideal plasmas and their boundaries

In analogy to ideal gases where neutral atoms bump elastically into each other, ions and electrons collide the same way in an ideal plasma. Meaning that for distances between two particles in the range of their mean inter atomic distance the potential energy produced by one another can be neglected when compared to their kinetic energy. For single charged ions the boundary for such an ideal plasma is given by[Str11]

$$\frac{3}{2}k_{\mathsf{B}}T \gg \frac{e^2}{4\pi\epsilon_0}n^{1/3}.$$
 (2.4.16)

The boundary between ideal to non-ideal plasmas then is reached when the interaction energy becomes comparable to the thermal energy of the plasma particles. At this boundary $N_d = 1$. In this region of N_d , for very high densities, the Pauli principle also has to be incorporated when dealing with plasmas, particularly because quantum mechanical effects become important when the Temperature of the plasma reaches values that are in the same region as its Fermi energy E_F . As a consequence, ideal plasmas have to suffice the condition that their thermal energy has to be smaller than their Fermi energy[Str11]

$$k_{\rm B}T \le E_{\rm F} = \frac{\hbar^2}{2m_{\rm e}} \left(3\pi^2 n\right)^{2/3},$$
 (2.4.17)



Figure 2.4.2.: n, T-diagram depicting the boundaries of an ideal plasma (marked as the colored area). The scheme is based on the diagram shown in [Sch93].

otherwise the electrons of a plasma underlie Fermi-Dirac instead of Boltzmann statistics. In this case the de Broglie length of the electrons are in the same scale as their mean inter-electronic distance. Another boundary of an ideal plasma is hit when particle velocities become high enough which is most probable for electrons as they are the species of charges with the lowest mass within an plasma. If that is the case, relativistic effects have to be considered. The boundary for an ideal plasma in this case is therefore defined as such that its kinetic energy has to be lower as the electron rest mass[Str11]:

$$k_{\rm B}T \le m_{\rm e}c^2 = 511 \,{\rm keV}$$
 (2.4.18)

Figure 2.4.2 depicts these mentioned boundaries graphically in a n, T-diagram which also shows the lower boundary at which hydrogen ionizes.

2.4.3. Holtsmark distribution

Since a plasma consists of free charge carriers, each with their own electric microfield E at position r, it is of interest to discuss the probability distribution describing the electric field composed of the

sum of these microfields

$$E(r) \equiv E(r; r_1, ...) = \sum_{i=1}^{N} E_i(r, r_i).$$
 (2.4.19)

For such a case this distribution is the so-called Holtsmark probability distribution[Spa12]

$$H(\beta) = \frac{2}{\pi\beta} \int_0^\infty x \sin(x) e^{-\left(\frac{x}{\beta}\right)^{3/2}} dx.$$
 (2.4.20)

In equation (2.4.20), H is expressed in units of the reduced field strength

$$\beta = \frac{E}{E_0},\tag{2.4.21}$$

with $E_0 = e/4\pi\epsilon_0 r_0^2$ as the field strength arising from an ion at the mean inter-ionic distance r_0 [HM88]. For later analysis the rational approximation of equation (2.4.20)[Hum86] is used where the asymptotic behaviors

$$H(\beta) \sim \begin{cases} \frac{4}{3\pi} \beta^2 & \text{for } \beta \to 0\\ \frac{3}{2\pi} \beta^{-5/2} & \text{for } \beta \to \infty, \end{cases}$$
(2.4.22)

play the dominant part.

3. Experimental setup

Following the experimental setup is presented. This concerns the setup for lasers used to excite the cesium vapor, the cell itself and the oven for keeping the cesium vapor heated. Additionally, the devices for the used microwave excitation and the fluorescence analysis are shown. At the end, the program for the edge lock mechanism control is shown.

3.1. Excitation scheme and Laser sources

The experiments can be divided into two categories: ones including microwave excitation but a nonlocked coupling laser, and ones without microwave excitation for which probe and coupling laser have to be locked. Either way, the cesium vapor gets excited from the ground state $6S_{1/2}$ to the intermediate state $7P_{3/2}$ and from that level to the excited $nD_{5/2}$ Rydberg state. For the microwave experiments, the excitation scheme depicted in figure 3.1.1a) is used, whereas for the case without the microwave, the same scheme can be used. However, after excitation to the $nD_{5/2}$ state no further excitation is needed.

A scheme for laser setup can be seen in figure 3.1.1b). The probe laser is a commercial TA SHG pro system from Toptica (locked on $\lambda_p = 455.66$ nm), where as the excitation from the intermediate to the excited Rydberg state is done with a commercial DL100 pro design system (also from Toptica) seeding a tapered amplifier (TA) system seeding a commercial fiber amplifier from Keopsys (KPS-CUS-BT-YFA-42-SLM-PM-HIP-111-FA-CO). That way, laser powers up to 15 W, and therefore high Rabi frequencies (in this case $\Omega_c = 2\pi \times 48.84$ MHz) even though of a relatively high beam diameter (0.7 mm), are possible. This laser system was previously described in [Urv16] and [Urv11], as well as the standard stabilization techniques for the lasers itself. Additionally, the power of the probe laser is stabilized with an AOM. That way Rabi frequencies in the range of $\Omega_p = 2\pi \times 9.41$ MHz ($\propto 10$ mW) are achieved.

3.2. Experimental setup

In this section the vapor cell and oven, that keep the cesium vapor in a heated gas phase state, are presented. The reason, that the vapor needs to stay heated is that high enough densities are inevitable for optical bistability, which in addition is accompanied by some fluorescent effect, to occur. Since the main goal of this thesis is to analyze this fluorescent effect, that appears as dim glow of a part of the cesium vapor along the laser beam path, tools are introduced in order to analyze the system and to get a better understanding for the edge lock mechanism (the term 'edge' refers to the border between glowing and non glowing part of the vapor).



Figure 3.1.1.: a) Excitation scheme for the experiments on ¹³³Cs. b) Laser sources for the experimental setup that are driving the probe and coupling transitions. Also included are the tapered amplifier, the fiber amplifier and the EIT lock scheme.

3.2.1. Vapor cell and oven

Considering the examined system, the cesium vapor is enclosed in a vapor cell made out of fused silica designed with a rectangular profile (see fig. 3.2.1a)). Height and width of the cell are 1 cm and the length is 5 cm. The purpose of this design is to get a better look on the fluorescence within the cell itself. Therefore, the oven also is designed in a rectangular manner, where the cell is embedded in a heatable copper frame (acting as a reservoir when heated for further control of the ground state density) that again is embedded in a TECAPEEK frame (see fig. 3.2.1b)). In turn, the sides of the oven are closed with fused silica plates to ensure proper observation of the processes within the cell. With that construction, cell temperatures of about 100 °C can be achieved, while ground densities of about $10^{18} m^{-3}$ are realized with a reservoir temperature of 83 °C.

Since the measurements are done outside the weak probe regime, meaning that the Rabi frequencies are much higher than what is usually applied in atomic spectroscopy, a series of phenomena is



Figure 3.2.1.: a) Cesium vapor cell with $1 \text{ cm} \times 1 \text{ cm} \times 5 \text{ cm}$ dimensions. The area marked in orange implies the glowing part of the beam path of the two counter propagating lasers $(\lambda_p \text{ from left}, \lambda_c \text{ from right})$. b) Schematic assembly of the oven and vapor cell. c) Schematic overview of the setup showing that the vapor gets excited by counter propagating probe and coupling laser. Also shown is the relative position of the microwave horn, fluorescence detection and the camera monitoring the insides of the cell.

triggered: On the one hand optical bistability can be measured and on the other hand a dim glow of the atomic vapor which is coincidently even visible to the bare eye through laser safety goggles (see fig. 3.2.1a)). It has to be noted, that without the laser safety goggles the dim glow would have been out-shined by the scattered fluorescence of the blue laser. The fluorescence is of particular interest in this thesis since proficient treatment enables full control over the edge of the glowing part.

Figure 3.2.1c) shows how the basic experimental setup looks like. The cesium vapor gets excited by the probe and coupling laser in a counter propagating manner, whereas a USB camera is constantly monitoring the inside of the cell which is helpful when trying to control the position of the edge of the glowing vapor (further insight on specifications of the camera and how the Edge lock is working in detail is given in chapter 3.3). Also schematically depicted are the microwave horn (further described in chapter 3.2.3) with which further transitions between Rydberg states are excited. Also depicted is the way how the fluorescence is analyzed (see chapter 3.2.4).

3.2.2. Control of the coupling laser

The interesting part for this thesis is the fluorescence emerging from the glowing part of the vapor along the laser beam path. As shown in figure 3.2.1a), the fluorescence has sharp transition between its bright state and the non-glowing domain. Since the effect heavily depends on the laser power and detuning, it is obvious to exploit this in order to manipulate the transition position, further only called 'edge', and study its behavior.

Since the coupling laser is a tunable grating diode laser (external cavity diode laser), its detuning can be controlled by giving a ramp signal on a piezo crystal controlling the grating of the diode laser. Appropriate control over the edge requires control over the offset parameter of the ramp signal. In case the edge needs to be spatially locked, an algorithm is needed (see chapter 3.3) which constantly requests the position of the edge and accordingly alters the offset in such a way that the edge seems to be spatially locked. The frequency and amplitude of the signal in this case are set to its minimum possible values since they do not affect the edge position.



Figure 3.2.2.: Pictures of a) the E8257D PSG Microwave Analog Signal Generator from Keysight Technologies[MW] and b) the SAR-2013-28-S2 microwave horn from SAGE Millimeter[hor].

3.2.3. Microwave setup

In order to drive transitions between two Rydberg states, long-wave radiation in form of microwaves is needed. The microwave source of choice is the E8257D PSG Microwave Analog Signal Generator from Keysight Technologies (fig. 3.2.2a)), making it possible to drive transitions in the range between 100 kHz to 67 GHz. Combined with a SAR-2013-28-S2 microwave horn from SAGE Millimeter (fig. 3.2.2b)) (with a gain of $20 \pm 1 \, dBi$ in a frequency range of 26.5 GHz to 40 GHz) and a PE3C0666-60 radio frequency cable from Pasternack[®][cab] with a length of 1.542 m, the microwave is shined into the cell from the side as depicted in figure 3.2.1c).

3.2.4. Fluorescence analysis equipment

In order to characterize the fluorescence, the collected light is coupled into a grating spectrometer (Andor, model Shamrock SR-303i-B-9FT). The camera (also from Andor, model iDus DU401A-BR-DD) connected to the Shamrock can be cooled down to -75 °C via peltier cooling in order to improve the signal-to-noise ratio. A few more specifications can be seen in table 3.2.1. Concerning the wavelength elements of the Shamrock, which has a triple grating turret mounted in its inside

Table 3.2.1.: Specifications of the grating spectrometer[Sha] and the CCD camera[IDU] from Andor.

Shamrock SR-303i-B-9FT					
aperture:	f/4				
focal length:	303 mm				
grating size:	$68\mathrm{mm} imes 68\mathrm{mm}$				
wavelength accuracy:	± 0.2 nm				
wavelength reproducibility:	± 0.05 nm				
wavelength range:	190 nm to $10\mu\text{m}$ (dependent on detector)				
Andor iDus DU401A-BR-DD					
active pixels:	1024×128				
pixel size:	$26\mu m imes 26\mu m$				

providing three different gratings, the only grating used is one with a groove density of $G = 600 \frac{1}{\text{mm}}$ with a blazing wavelength of $\lambda_{\rm B} = 800 \text{ nm}$. If desired the used grating can be swapped with two others. One with $G = 235 \frac{1}{\text{mm}}$ and $\lambda_{\rm B} = 750 \text{ nm}$ and the other one with $G = 600 \frac{1}{\text{mm}}$ and $\lambda_{\rm B} = 500 \text{ nm}$. The fiber to couple the collected fluorescence into the spectrometer is a FT1000EMT hard clad multimode fiber from Thorlabs with a length of 13 m and a core diameter of 1000 µm[Fib]. Further, specifications for the laser safety goggles, with which it was possible to see only the dim glow within the vapor cell, can be found in [Saf].

A further discussion on the accuracy of the spectrometer can be found in the appendix A.1.

3.3. Edge lock

This section deals with the spatial locking of the edge and basically can be split into two parts. The first one deals with the tracking problem since the final program constantly is monitoring the spatial position of the edge, whereas the second part deals with the regulation of prior mentioned parameters controlling the edge. For monitoring the edge position a DCC1545M CMOS camera with 1280×1024 pixel from Thorlabs[DCC] is controlled via Matlab. By filming the side of the previously shown vapor cell (fig. 3.2.1b) shows the assembly of camera, vapor cell and the microwave horn in the experiment), the recorded frame then can be analyzed. The used algorithm for the analysis of the recorded frame is depicted in figure 3.3.1 and executed in the following order:

- Record frame The recorded frame is prepared for image processing with Matlab.
- Smooth data In this process the data is smoothed to omit noise in further analysis.
- Get profile A straight line is fitted through the intensity profile of the glowing part of the beam path. This is done by using a Gaussian fit routine for every vertical array of the matrix containing the frame data.
- Calculate second derivative This is done along the prior obtained profile in order to obtain the vertical positioning of the edge.
- Submit data & repeat The obtained position is now passed onto the part of the program controlling the edge position.



Figure 3.3.1.: Schematic view of the five step tracking algorithm.

After each frame is analyzed, the coordinates of the edge are submitted to the part of the program responsible for spatially controlling the edge, which itself is achieved through an PI-loop controlling the offset of the scan of the coupling laser (how this is done is already described in chapter 3.2.2).

3. Experimental setup

That way, the edge is confined at a previously specified horizontal position. It has to be noted that the edge still performs some negligible chaotic oscillations around the specified position since the microwave also has an effect on the edge.

The final result of the program can be seen in the screenshot depicted in figure 3.3.2. It shows a snapshot of a graphical user interface (GUI) containing a frame of a live video during a measurement besides its features that count in: edge-locking, separate control of the scan parameters and features to control camera parameters such as exposure time, gain and frame rate. The chosen scale corresponds to the pixel scale of the used CMOS camera.



Figure 3.3.2.: Edge-lock program with its features. The GUI enables users to control the edge-lock as well as the camera parameter freely as he pleases. With a red dot in the snapshot of the live video, the current calculated position of the controlled edge is marked.

4. Experimental Results

In this chapter the observations during the experiments on gas phase cesium within a thermal vapor cell are presented and discussed. Via fluorescence spectroscopy, a Stark shift is observed of the transition from $42D_{5/2}$ to $40F_{5/2}$, which is shown in section 4.1. Moreover, a theoretical model to explain this behavior is evaluated.

Section 4.2 is devoted on explaining the occurrence of charges within the thermal vapor. Theoretical models are evaluated together with experimental data retrieved from fluorescence measurements.

4.1. CW-Microwave experiments

4.1.1. Experimental procedure

In the experiments concerning microwave excitation of cesium, atoms get excited from the ground state $6S_{1/2}$ to the $7P_{3/2}$ state with the probe laser locked on resonance. Then the vapor gets further excited from that intermediate level to the Rydberg state $42D_{5/2}$. The detuning of the coupling laser is controlled with the edge lock program (see chapter 3.3) in such a way that the edge is spatially centered within the cell. On the other hand the Rabi frequency is fixed at $\Omega_{R,c} = 2\pi \times 17.8$ MHz which corresponds to an output power of 2W averaged over a beam waist of 0.7 mm).

Considering the microwave excitation, the $40F_{5/2}$ is chosen as the destined Rydberg state. The transition from $42D_{5/2}$ to $40F_{5/2}$ is scanned from 41.75 GHz to 45.75 GHz in 0.25 GHz steps, expecting to have a resonance frequency of $\nu_0 = 45.37$ GHz[vPWA17]. The related Rabi frequency is estimated to $\Omega_{R,MW} \approx 2\pi \times 60$ MHz since the profile of the microwave horn is rectangular (42.4 mm × 33.5 mm) and the output power of the generator during this measurement is 0 dBm (= 1 mW).

For each frequency, five fluorescence spectra are recorded in a region between 600 nm 700 nm. The cell temperature during the experiment is $T = 100 \,^{\circ}\text{C}$ and the exposure time of the spectrometer is 1.5 s (the way this dataset is analyzed is described in chapter 4.1.2).

The Rabi frequency of the probe laser is varied in such a way that for each Rabi frequency an averaged spectrum is recorded (note: the position from which the fluorescence gets measured, is shifted roughly 1 cm to the left (camera perspective as a reference) in respect to the fixed edge (see fig. 3.3.2)). The variation is achieved by tweaking the beam power before it enters the cell in a range from 0.5 mW to 8 mW which corresponds to Rabi frequencies in the range from $\Omega_{R,p} = 2\pi \times 2.11 \text{ MHz}$ to $\Omega_{R,p} = 2\pi \times 8.42 \text{ MHz}$ by a given beam waist of 0.7 mm.

4.1.2. Total fluorescence in dependence of the microwave frequency

In this subsection the way, how the spectra of the total fluorescence in dependence of the microwave frequency are obtained, is explained. As already mentioned in the experimental procedure 4.1.1, five fluorescence spectra at each microwave frequency are recored. After averaging those spectra, which

4. Experimental Results



Figure 4.1.1.: a) Averaged fluorescence spectra for each microwave frequency which was used to excite the cesium vapor. b) The same as in figure 4.1.1, but background corrected.



Figure 4.1.2.: Total fluorescence obtained by summing the intensity of figure 4.1.1b) up along the wavelength axis.

is done in order to reduce noise, they are plotted for each microwave frequency and put beneath each other. Figure 4.1.1a) shows an example how this looks like for a measurement series at a probe Rabi frequency of $\Omega_{R,p} = 2\pi \times 3.33 \text{ MHz}$. After background correction the spectrum shows

dips around 43 GHz (see fig. 4.1.1b). In the last step to obtain the total fluorescence, which then can be relatively easy analyzed and understood, the intensity of the averaged spectra for different microwave frequencies is summed up along the wavelength axis and afterwards normalized. The final result can be seen in figure 4.1.2.

4.1.3. The effect of ions on the fluorescence in the vapor cell

In the following discussion, the total fluorescence depicted in figure 4.1.2 is further used as an example, and therefore plotted again in figure 4.1.3 that also shows some other curves obtained from theoretical model presented in the next passage. It has to be noted, that the noise appearing in this fluorescence spectrum is due to the fact that the 13 m fiber was exposed to external light sources during the measurement (the spectrometer is located in another room).

Figure 4.1.3 shows a drop in intensity at around 43.51 GHz, with a line width of about $\sigma_{exp} \approx 0.3$ GHz, instead of showing a dip at $\nu_0 = 45.37$ GHz. An explanation for the 1.86 GHz shift could be that ions within the vapor cell cause a dc Stark shift due to the electric field they produce.

To investigate this issue, a theoretical model is needed. Build upon the assumption of ions present in the vapor cell, the Holtsmark distribution $H(\beta) = H(E/E_0)$ presented in chapter 2.4.3 is used $(\beta = E_0/E_{\text{ext}})$ is the dimensionless electric field parameter in the Holtsmark model). Furthermore, a Gaussian profile of the unperturbed signal is assumed. In case this signal experiences a dc Stark shift, the Gaussian curve has the following form:

$$F\left(\nu - \nu_0 + \frac{1}{2}\alpha(E + E_{\text{ext}})^2\right) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{1}{2\sigma^2}\left(\nu - \nu_0 + \frac{1}{2}\alpha(E + E_{\text{ext}})^2\right)}$$
(4.1.1)

 $\alpha = \alpha (40F_{j=5/2,m_j=\pm 1/2}) - \alpha (42D_{j=5/2,m_j=\pm 1/2}) = (1.57+0.26) \cdot 10^3 \text{ MHz} \cdot \text{cm}^2/\text{V}^2 \text{ is the coefficient}$ for the relative shift due to the polarizability for the $42D_{5/2} \rightarrow 40F_{5/2}$ transition[vPWA17]. Also included is the quantity E_{ext} , which is important in order for the theoretical model to fit the total fluorescence depicted in figure 4.1.3. Its explanation is given after the full model is presented.

In the following discussion of the model the Holtsmark distribution is convolved with the Stark shifted Gaussian in (4.1.1). That way, a signal for modeling the absorption in dependence of the electric field E_0 created by ions within the cell is obtained:

$$S_{\mathsf{abs}}(\nu) = \int_0^\infty H(\beta) F\left(\nu - \nu_0 + \frac{1}{2}\alpha(E + E_{\mathsf{ext}})^2\right) \mathrm{d}\beta.$$
(4.1.2)

Afterwards, the transmission signal then can be calculated easily as

$$S_{tr}(\nu) = 1 - S_{abs}(\nu).$$
 (4.1.3)

By plotting the transmission signal (4.1.3), where $E_0 = 0.08 \text{ V/cm}$ and $E_{\text{ext}} = 1.3 \text{ V/cm}$, together with the measured data (see fig. 4.1.3) it can be shown that one can achieve quite good agreement between the theory curve of equation (4.1.3) and the experimental data. However, this was only possible with the electric field parameter E_{ext} . Without this extra quantity (in this case $E_{\text{ext}} = 0$), the computed signal is extremely broadened (purple curve in fig. 4.1.3) when the curve would have



Figure 4.1.3.: Measurement series at $\Omega_{R,p} = 2\pi \times 3.33 \text{ MHz}$: (blue) total fluorescence (normalized), (orange, dashed) Gaussian profile $F(\nu - \nu_0)$ of the transmission signal at its expected position without a Stark shift, (yellow) convolution of H and F ($E_0 = 0.08 \text{ V/cm}$, $E_{\text{ext}} = 1.3 \text{ V/cm}$), (purple, dashed) convolution of H and F ($E_0 = 1.38 \text{ V/cm}$, $E_{\text{ext}} = 0 \text{ V/cm}$).

its center of mass at the same position as the measured data.

Figure 4.1.4 depicts this behavior in the case for $E_{\text{ext}} = 0$. In a range from 0 V/cm and 2 V/cm the parameter E_0 , which describes the electric field produced by ions within a system, is varied while calculating the absorption profile according to equation (4.1.2) (the absorption is chosen over the transmission only because of portrayal reasons). It can be seen for increasing parameter E_0 the position of the resonance shifts to lower frequencies, which is the expected effect. However, associated with that behavior is also a broadening of the signal requiring the external field parameter E_{ext} in order to suppress this effect while the resonance shifts to the red. The value for E_{ext} which provides a relatively good agreement with the measurement depicted in figure 4.1.3 can be justified by the fact that charges created within the cell stick to the walls of the vapor cell creating an external field lateral to the beam path since the cell width/height is about 1 cm. Annotating, the total fluorescence in dependence of the microwave frequency for the other Rabi frequencies can be found in the appendix.

Besides that discrepancy, a few more quantities like ion density and Debye length, leading to a discussion whether the examined system is an ideal plasma or not, can be calculated. The following


discussion of these calculated quantities, besides some other occurring dissent, is given next.

Figure 4.1.4.: Signal form of the absorption for values of E_0 between 0 V/cm and 2 V/cm and $E_{\text{ext}} = 0 \text{ V/cm}$. The blue line traces the maximal values.

4.1.4. Ion density, Debye length, plasma frequency and plasma parameter of the examined system

Due to the presence of charges within the vapor, it is discussed whether the examined system is an ideal plasma or not. This is done by calculating quantities like ion density, Debye length, plasma frequency and the plasma parameter from the mean inter ionic distance (introduced in chapter 2.4.1) calculated from E_0 .

Following in table 4.1.1, the values for these quantities are listed. Looking at the values for the ion densities another fact stands out. By using the parameter $E_{\rm ext}$, a density which is close to the predicted value of [WUR⁺16] (that is $n \leq 10^{16} {\rm m}^{-3}$), can be calculated using $E_{\rm ext} = e/4\pi\epsilon_0 r_0^2$ (with r_0 as the mean inter-ionic distance). The other way around, by using only the electric field E_0 created by the ions, the calculated value deviates by a factor of approximately 100 from the predicted value, raising the question if the underlying theoretical model is suited.

Now considering, that the measurement shown in 4.1.3 only shows the case for one Rabi frequency,

	$E_0=1.38\mathrm{V/cm},~E_\mathrm{ext}=0\mathrm{V/cm}$	$E_0=$ 0.08 V/cm, $E_{ m ext}=$ 1.3 V/cm					
$n (m^{-3})$	$7.08 \cdot 10^{15}$	$9.89 \cdot 10^{13}$					
λ_{d} (µm)	15.76	133.44					
ω_{p} (Hz)	$4.75 \cdot 10^{9}$	$5.61 \cdot 10^{8}$					
$N_{\rm d}$ (1)	116.25	983.98					

Table 4.1.1.: Ion density, Debye length, plasma frequency and plasma parameter in dependence of the fit parameter E_0 .



Figure 4.1.5.: Electric fields E_{Stark} , E_{ext} and E_0 in dependence of the Rabi frequency of the probe laser. Also shown is the sum of E_{ext} and E_0 .

it is of interest how the drop in the transmission behaves for other Rabi frequencies. Therefore, the resonance frequencies for the measurements done at other Rabi frequencies are determined and according to a dc Stark shift (eq. (2.1.7)) the corresponding electric fields E_{Stark} are calculated. Also determined are the parameters E_0 and E_{ext} which are important for the calculation of (4.1.3). The results can be seen in figure 4.1.5. Noticeable is that the electric field E_{Stark} , calculated from the determined frequency shift, has the same magnitude as the sum of the electric field parameters E_0 and E_{ext} obtained only from optimizing the transmission signal (4.1.3) onto the measurement (since this is not done with a fit routine error bars are not included). Moreover, the electric field parameter E_0 seems to stay constant while increasing the Rabi frequency, whereas the parameter



Figure 4.1.6.: Ion density calculated with the electric fields E_{Stark} and the the electric field parameters E_0 and E_{ext} .

 E_{ext} also is not differing much from E_{Stark} . The increase of E_{Stark} could be explained in such a way that with increasing Rabi frequency more cesium atoms get excited into the Rydberg state, and as a result more Rydberg atoms become ionized by some other process (which is discussed in chapter 4.2), which can be seen in figure 4.1.6 that shows the increase in ion density on the same Rabi frequency scale.

Again, disagreement with the theoretical model emerges. A higher ion density would result in a higher electric field E_0 produced by the ions, which can not be seen in figure 4.1.6 that instead shows a rather constant behavior for the ion density only calculated with E_0 . Furthermore it shows that the density calculated with the sum of field parameters $E_0 + E_{\text{ext}}$ is closer to the value retrieved from the Stark shifted values. Another discrepancy is that in case the density is only calculated with E_0 the values of it are far too low (about a factor of 100) in order to fit the predictions from [WUR⁺16], whereas the values calculated with the sum of the field parameters are quite near to that predicted value.

Further, figure 4.1.7 shows the Debye lengths calculated from the measurement and the assumed theoretical model. Again there is good agreement between the measurement and the theoretical model in case for the the sum of the field parameters. The values for the case, in which only E_0



Figure 4.1.7.: Debye lengths calculated with the electric fields E_{Stark} and Debye lengths calculated with the sum of the electric field parameters E_0 and E_{ext} .

is considered, is not shown in this graph since it only would be a rather stagnating value in the $\sim 10^{-4} {\rm m}$ region which in turn would again show disagreement with the theoretical model. Whether the model for assuming a Holtsmark distribution convolved with a Gaussian function to describe the signal form is completely wrong or the interpretation of $E_{\rm ext}$ as an offset field is not appropriate remains unclear and has to be studied further. However, the measurements show that in some way charges are involved that in some, until now, unclear way shift the resonance frequency of the $42 {\rm D}_{5/2} \rightarrow 40 {\rm F}_{5/2}$ transition to lower frequencies supporting the assumption that a Stark shift is involved.

A reason why the Holtsmark model might not be suitable is, that it does not considers electrons within the system.

4.2. Ionization process of the Rydberg atoms

This section deals with the ionization processes during two photon excitation of the heated vapor. It is investigated, whether Penning ionization of two cesium atoms in the Rydberg state or collisional processes are the cause.

4.2.1. Experimental procedure

In contrast to section 4.1, the atoms are only excited with the inverted ladder scheme to $42D_{5/2}$ ($30D_{5/2}$, $50D_{5/2}$) and no sub-sequential microwave excitation is applied. Moreover both, probe and coupling transition are locked on resonance. As a result the whole beam path within the cell is now fluorescing, so that there is not edge any more which must be confined spatially.

The Rabi frequency of the laser driving the $7P_{3/2} \rightarrow 42D_{5/2}$ transition is $\Omega_c = 2\pi \times 12.6$ MHz (1W with a 0.7 mm beam waist at $\lambda_c = 1064.98$ nm). Then two other states are chosen as the destined Rydberg state in which the cesium vapor gets excited to, namely the $30D_{5/2}$ and $50D_{5/2}$ Rydberg state. In case for $30D_{5/2}$ the power of the coupling laser has to be adjusted to 0.33 W in order to maintain the Rabi frequency of $\Omega_c = 2\pi \times 12.61$ MHz now corresponding to a coupling wavelength of $\lambda_c = 1073.50$ nm, whereas in case for the $50D_{5/2}$ the power has to be tweaked to 1.75 W in order to reach the same goal. Additionally, the coupling wavelength, for excitation to the $50D_{5/2}$ Rydberg state is 1062.53 nm. Apart from that, the Rabi frequency for the probe laser is now $2\pi \times 4.71$ MHz during all these experiments and the temperature within the cell is around T = 100 °C (T = 83 °C as reservoir temperature).

During those excitations two fluorescence spectra are recorded covering a range from \sim 500 nm to \sim 700 nm in total. The exposure time of the spectrometer was varied according to the purposes of the experiment. In case for investigating Penning ionization as the cause for why ions are present within the cell, the exposure time is rather long (900 s). As a result some peaks show saturating behavior (see fig. 4.2.2) which nevertheless can be neglected since other peaks are examined.

For the discussion of collisional processes, the same measurement procedure is applied and additionally experiments at a reservoir temperature of $T = 92 \,^{\circ}\text{C}$ were done.

4.2.2. Penning ionization

The first assumption on how the ions within the cell are created is Penning ionization. During this process, which involves two atoms in a certain electronic state, one atoms gets ionized, whereas the electron of the other atom drops down to an energy level that is as far away in energy space as the amount of energy needed to ionized the other atom.



Figure 4.2.1.: Scheme for Penning ionization process involving two Rydberg atoms. One Rydberg atom gets ionized and the other one drops down the same amount of energy

One requirement for this process is that the sum of the energy levels of the two particles is higher than the ionization energy of one of these atoms[Gal05]. In our case the species of the two atoms would be the same as well as their energy levels. The process, in which a free electron and an ion are created, can be described with

$$Ry_{Cs} + Ry_{Cs} \rightarrow Ry_{Cs}^{+} + Ry_{Cs}^{*} + e^{-}$$
 (4.2.1)

and is schematically shown in figure 4.2.1.

For excitation into the $42D_{5/2}$ Rydberg state, the energy (after the ionization process) of the cesium atom that does not get ionized and instead drops to a lower energy level (due to energy conservation) can be determined to 0.01741 eV (which is the doubled ionization energy). Following from that, it is most probable that the atom dropped down to the $28F_{5/2}$ state since it has an ionization energy of 0.01740 eV. Then from that state, the Rydberg state should decay into lower levels like the $5D_{3/2}$ or $5D_{5/2}$ and the fluorescence lines should be detectable with a spectrometer.



Figure 4.2.2.: Fluorescence from a) 490 nm to 585 nm and b) 585 nm to 685 nm recorded with an exposure time of 900 s while exciting the cesium atoms into the $42D_{5/2}$ Rydberg state. The red line implies the position at which the cascade of peaks for the decays beginning at the $33S_{1/2}$ state, whereas the green line fulfills the same purpose in case for the $28F_{5/2}$ state.

In fact, if the $28F_{5/2}$ decays into the $5D_{3/2}$ state, the transition wavelength should be in the visible range ($\sim 596.41 \text{ nm}$). Based upon this fact, the spectra should show a cascade of fluorescence peaks beginning at this wavelength, since the $28F_{5/2}$ also decays into other states. The spectra, recorded with an exposure time of 900 s, can be seen in figure 4.2.2, which shows the measured fluorescence between 490 nm to 685 nm.

In spectrum 4.2.2b) it can be seen that a cascade of peaks actually begins around the calculated \sim 596.41 nm, and no decays from higher lying states are visible (green vertical line in fig. 4.2.2b)), although the intensity compared to other parts of the spectrum is quite low even after an 900 s exposure time. Moreover, spectrum 4.2.2a) additionally shows peaks at lower wavelength.

These could be explained by the fact that not only the $28F_{5/2}$ state is addressed by Penning ionization but also states like $33S_{1/2}$ (ionization energy: $0.016\,23\,\text{eV}$) and $31D_{3/2}$ (ionization energy: $0.016\,72\,\text{eV}$) which then decay into the $6P_{1/2}$ and $6P_{3/2}$ state with respective transition wavelengths of 497.58 nm (red vertical line in fig. 4.2.2a)) for $33S_{1/2} \rightarrow 6P_{1/2}$ and 497.68 nm for $31D_{3/2} \rightarrow 6P_{1/2}$. And indeed, spectrum 4.2.2 appears to show such features at around 596.41 nm, again with a very low intensity of the fluorescence. Another thing to mention is that some of the peaks in figure 4.2.2b) show saturating behavior due to the 900 s exposure time.

A last thing that should be mentioned is that not only decays from $nF_{5/2}$ states are visible in this spectrum but also from $nP_{3/2}$ states. The calculated cap for the principal quantum number due to Penning ionization is n = 32 which then corresponds to an ionization energy of 0.01682 eV. Transitions into the $5D_{3/2}$ and $5D_{5/2}$ state then should begin around 596.25 nm

Summarizing, the assumption for Penning ionization appears to fit the measured data. However, the same measurement is repeated twice in order for a sanity check.

In the first repetition excitation now happens to the $30D_{5/2}$ state, whereas in the second repetition the $50D_{5/2}$ state is addressed. In both cases, one would expect that the beginning of one of the mentioned peak cascades has to shift. A shift to lower wavelengths for excitation to $50D_{5/2}$ and to higher wavelengths (since less energy is needed for ionization) in case the vapor is driven to the $30D_{5/2}$ (since more energy is needed) state. Decays to $5D_{3/2}$ from F states then would begin at $34F_{5/2}$, with a respective wavelengths of 594.81 nm when exciting the $50D_{5/2}$ state, and $20F_{5/2}$, with the cascade beginning at a wavelength of 601.25 nm.

At this point it has to be mentioned that the same argumentation applies to other cascades as well, but are not treated further since only one example is sufficient as we see in figure 4.2.3, which shows three almost identical fluorescence spectra despite that three different Rydberg states are addressed with the coupling laser. In case for exciting to $30D_{5/2}$ the intensity is overall a little bit higher resulting in higher peaks around 595 nm to 600 nm. Consequently, the model concerning Penning lonization failed the sanity check and therefore is invalid since it could not meet the expectation of shifted cascades within the fluorescence spectrum.

The fact that the predicted shift of the cascades did not occur, shows that the assumption that the ions in the cesium vapor are created by Penning ionization is not right. Therefore, another model is investigated concerning on ionization through collisions of charged and uncharged particles with the heated cesium atoms in the Rydberg state.



Figure 4.2.3.: Fluorescence spectra for excitation to $42D_{5/2}$, $50D_{5/2}$ and $30D_{5/2}$. The red lines mark the position at which the cascade of $nF \rightarrow 5D_{3/2}$ transitions should begin. The spectra were recorded over an exposure time of 900s and afterwards normalized.

4.2.3. Collisional processes within the cesium vapor

In the following discussion it is assumed that the Rydberg atoms get ionized by collision. Two cases are examined: collisions with ground state atoms and with electrons created during the process. For both cases a Maxwell-Boltzmann probability distribution[Den93]

$$f_{\mathsf{MB}}(v,T) = 4\pi N \sqrt{\frac{m}{2\pi k_{\mathsf{B}}T}} v^2 \mathrm{e}^{-\frac{mv^2}{2k_{\mathsf{B}}T}}$$
(4.2.2)

is assumed, where N is the density and m is the mass of respective particles. The model starts with the ionization process of the previously mentioned $nS_{1/2}$, $nP_{3/2}$, $nD_{3/2}$ and $nF_{5/2}$ states with n = 7...50 (for the model assumed range). At the beginning it is assumed that they are all equally populated. How this is achieved however is not included in the further discussion as this process still is unknown (see fig. 4.2.4a)). Following, the model is explained with the aid of the ionization of the $nF_{5/2}$ states. Now, the mentioned states get ionized by collisions with a probability that corresponds to the area under the Maxwell-Boltzmann distribution from a velocity corresponding to the ionization energy of the respective $nF_{5/2}$ state to infinity: $P = \int_{v_{min}}^{\infty} f_{MB}(v, T) dv$. As a consequence, states with high principal quantum number have a higher chance of ionization than states with lower n



Figure 4.2.4.: a) Schematic picture of transitions within the discussed model with the ansatz of collisional processes. b) Maxwell-Boltzmann probability distribution for an electron at a temperature of T = 100 °C.

which can be seen in figure 4.2.4b) with the aid of the Maxwell-Boltzmann distribution for electrons at T = 100 °C. Whether it is justified to assume such low electron temperatures or not is discussed later. To model the ionization rate, quantities like the cross section σ and the mean particle velocity \overline{v} have to be included in the model. Parallel to that, the Einstein A coefficients of for the respective transitions (the values are calculated with the Atomic Rydberg Calculator[vPWA17]) which are examined with this model parallel to the life time of the ionizing Rydberg $\Gamma(nF_{5/2})^{-1}$ have to considered. So now, the full model to describe the intensity distribution within the fluorescence spectra is described as

$$\gamma = F \frac{A}{\sigma \overline{v} P + \Gamma(n \mathsf{F}_{5/2})},\tag{4.2.3}$$

where the overall scaling is denoted by the fit parameter F (γ is randomly chosen variable). For the particle density, $N = 10^{16} \text{m}^{-3}$ which is the predicted value from [WUR⁺16], is chosen since this example is explained by the aid of electrons (this density is then the electron density). With the model for γ , cross sections for the collisional process can be determined. Since only σ is an unknown in (4.2.3), it can be roughly estimated.

Figure 4.2.5 and 4.2.6 show the resulting curves of γ under the assumption that the atoms get ionized through electron collisions together with the measured spectra (1.5 s exposure time) in case this procedure is done when exciting the cesium vapor into the 50D_{5/2} state. The marked peaks are identified by calculating the wavelengths with a module, called Atomic Rydberg Calculator[vPWA17], of the scripting language Python. There seems to be relatively good agreement between theory and experiment since the curves for γ coincide with the marked (colored) fluorescence peaks, especially for higher quantum numbers. Resulting values for the estimated mean ionization cross sections through electron collisions are presented in table 4.2.1. Here it has to be pointed out that these values only depict the mean value of the cross section. A scaling with a power law of the principal quantum number is not explicitly considered but discussed at the end of this subsection. More



Figure 4.2.5.: Fluorescence spectrum depicting peaks, resulting from decays from the $nS_{1/2}$ $nD_{3/2}$ states, whose maximal values are marked with dots for better comparison between the measurement and the theoretical model. Also shown are the respective curves for γ .

plots where the same analysis routine is applied for excitation into the $30D_{5/2}$ state and for another reservoir temperature are listed in appendix A.3 since they show the same behavior even though the ground state densities in both cases are different ($N_{83} = 2.9 \cdot 10^{18} \text{m}^{-3}$ and $N_{92} = 5.4 \cdot 10^{18} \text{m}^{-3}$). In

Table 4.2.1.: Mean ionization cross sections σ of the $nD_{3/2}$, $nS_{1/2}$, $nF_{5/2}$ and $nP_{3/2}$ states through electron collisions. The reservoir temperature was T = 83 °C while the cesium vapor was excited to the $50D_{5/2}$ Rydberg state.

$\gamma(nD_{3/2}\to 6P_{1/2})$	0.40	$\gamma(nF_{5/2}\to 5D_{5/2})$	0.14
$\gamma(nD_{3/2}\to 6P_{3/2})$	0.35	$\gamma(nF_{5/2}\to 5D_{3/2})$	0.20
$\gamma(nS_{1/2}\to 6P_{3/2})$	1.60	$\gamma(nP_{3/2}\to 5D_{5/2})$	5.00
$\gamma(nS_{1/2}\to 6P_{1/2})$	8.00	$\gamma(nP_{3/2}\to 5D_{3/2})$	5.00

mean ionization cross sections $\overline{\sigma}~(10^{-14}{
m m}^2)$

-

the work of Deutsch et. al. [DMB⁺06] calculated values for the ionization cross section of Na(nD) Rydberg atoms vary between $4.0 \cdot 10^{-15}$ m² and $1.6 \cdot 10^{-14}$ m² and experimental values [NM03], measured in a crossed-beam collision chamber, also range in that order. Since the estimated mean values in table 4.2.1 are of the same order, the assumption for ionization through electron impacts does not seem to be unlikely.

When doing the same routine, but instead assuming the atoms get ionized by collisions with ground state atoms, the calculated cross sections can be easily estimated. Since [WUR⁺16] estimates that the ion density is around 1% of the ground state density ($N_{\rm ion} \leq 10^{16} {\rm m}^3$), the values for the mean



Figure 4.2.6.: Fluorescence spectrum depicting peaks, resulting from decays from the $nF_{5/2}$ $nP_{3/2}$ states, whose maximal values are marked with dots for better comparison between the measurement and the theoretical model. Also shown are the respective curves for γ .



Figure 4.2.7.: Fluorescence spectrum depicting peaks, resulting from decays from the $nF_{5/2}$ $nP_{3/2}$ states, whose maximal values are marked with dots for better comparison between the measurement and the theoretical model. Also shown are the respective curves for γ in case for an assumed electron temperature of $T_e = 2000$ K.





ionization cross section would have to be lower by a factor of about ~ 100 (according to the model in equation (4.2.3)) for the curves γ to coincide with the measured data. In [Kor78] the cross section values for this case are around 10^{-18} m² to 10^{-23} m². With such a discrepancy, it is unlikely that the cesium atoms ionize mainly through collisions with ground state atoms, so the discussion now only is considering the case with electrons.

In case for the ionization through collisions with electrons a few more things have to be considered, that are the electron density and temperature and the scaling of the ionization cross section. Considering that in subsection 4.1.3 the data and Holtsmark model show a mismatch between the determined ion densities without the aid of the field parameter $E_{\rm ext}$ and the predicted value of $N = 10^{16} {\rm m}^{-3}$, the cross sections would have to be higher by a factor of 100, which otherwise would mean that the idea of electron impact ionization would have to be omitted. Another thing to mention is the assumed temperature of the electrons within the ionized gas. In [LW85][KKB⁺99] it is reported that in a 600 K cesium vapor a plasma with temperature $T_{\rm e} = 2000 {\rm K}$ is created through photo ionization. By plotting γ for such a relatively high temperature, one then can see (fig. 4.2.7) the mismatch between the theoretical model and experiment even. In figure 4.2.7, the curves for γ were calculated with the same cross sections previously estimated in the discussion at a temperature of $T_{\rm e} = 100 \,^{\circ}{\rm C}$ since a change in $\overline{\sigma}$ did not make the result any better.

At last, the behavior with the principal quantum number n of the cross section (and therefore the behavior of γ) is examined. Since it is unknown which scaling law must be applied to the system for a proper description, cases where the cross section scales with n^1 , n^2 , n^3 and n^4 (n as the principal quantum number) are tested. n^1 is applied, since in the work of Deutsch et. al. [DMB+06] (already mentioned in the previous paragraph) the cross sections show linear scaling behavior with the principal quantum number, whereas the n^4 is tested since the geometrical cross section exhibits this property. The quadratic and cubic scaling are also tested just in case the other scaling laws are not suited.

The result is depicted in figure 4.2.8. It shows that each tested scaling seems to fit quite well. However, for each growth of the exponent applied to the principal quantum number the mean cross section previously obtained has to be divided by a factor of 20. For linear scaling behavior the cross sections are still in a range, in which electron impact ionization can be assumed, whereas for higher exponents this assumption is not suited and another model would have to be used. Additionally, the best fit with experimental data is still achieved without scaling since for higher values of the exponent in the scaling behavior there is a growing misfit that especially can be seen for cubic n^4 scaling as depicted in figure 4.2.8.

5. Summary and outlook

Within the scope of this thesis an experimental setup for two photon excitation of cesium vapor in a thermal vapor cell is realized in order to examine bistable behavior. Particularly interesting in this thesis is the observed change in fluorescence of the cesium vapor along the laser beams. In this thesis it is exploited that the fluorescent effect is dependent on the laser power and detuning to control the position of transition between dark and bright part of the vapor.

In order to get a better understanding of this effect, the fluorescence emerging from the bright part is examined. The experiments can be categorized into to parts: The first part requires additional microwave excitation parallel to a non locked coupling laser, whereas the other part is done without the microwave and a locked coupling laser. A conclusion which can be drawn from the microwave experiment is that charges must be present within the cell as a Stark shift of about $\sim 1\,\text{GHz}$ to $\sim 3\,\text{GHz}$ of the resonance of the $42D_{5/2} \rightarrow 40F_{5/2}$ transition driven by the microwave is observed. From the measured shift an electric field in the range between $\sim 1\,\text{V/cm}$ and $\sim 2\,\text{V/cm}$ can be calculated. Parallel to that a theoretical model consisting of a convolution of the Holtsmark distribution and a Stark shifted Gaussian line profile is compared with the obtained data.

The other part of the experiments deal with the occurrence of charges within the cesium vapor. First a model for Penning Ionization is examined, which transpired that this model is not suitable since the expected shift of fluorescence peaks of the decays (especially the $28F_{5/2}$) is not observed when changing the Rydberg state the coupling laser is addressing. Next collisional processes, such as electron impact ionization and ionization through collisions with ground state atoms, were discussed. It appears that the model for electron impact ionization proves to suit as a better explanation for the occurrence of ions since the obtained values of the ionization cross section ($\sim 10^{-14} \text{ cm}^2$) had better agreement with literature values for this case compared to the one which assumes ground state collisional ionization.

Despite that, some discrepancies occurred. In case for the microwave experiments, the Holtsmark model demands an additional external field parameter $E_{\rm ext}$ since the electric field E_0 created by the charges alone predicts line broadening. Without the external field parameter, the calculated ion density did not match the predicted value[WUR⁺16] anymore. By incorporating $E_{\rm ext}$, a plasma parameter of about $N_{\rm d} = 100$ can be calculated. Moreover, the Debye length ranges roughly between 10 µm and 20 µm and the plasma frequency is in the GHz regime. In the discussion, whether the examined system is a plasma or not, this would mean that the system is in the region of a non-ideal plasma (see fig. 2.4.2) due to the fact of a relatively low plasma parameter and the low temperature $T_{\rm e}$ assumed (it is assumed that $T_{\rm e}$ thermalizes with the cell temperature) for the electrons. However, it is unclear whether it is justified to make this assumption for the temperature or not. Thus, to gain more insight on this topic, further studies and theoretical models should focus on this parallel to the discrepancy the Holtsmark model raises, which could mean that another model may be better

suited to describe the observed shifts.

In the other experiments, that dealt with the questions on how ions emerge within the cell, electron impact ionization seems to be the most plausible explanation, since the determined mean cross sections ranged in the same order as in [DMB⁺06]. However, applied scalings with the principal quantum number (power law from first to fourth order), also show relatively good agreement with the measured data with the difference, that for each time the power of n is raised the mean cross section has to be divided by a factor of 2. For linear scaling, the values of the cross section still would be explainable with electron impact ionization. Additionally, in order for this model to provide these cross sectional values the predicted value for the ion density of $n = 10^{16} \text{m}^{-3}$ is used, meaning the emphasis of further studies should focus on this topic.

A. Appendix

A.1. Characterization of the used spectrometer (SR-303i-B-9FT) and camera (iDus401A)

In order to know how accurate the SR-303i-B-9FT and the iDus camera are measuring the wavelengths, spectra are recorded with background light and the lasers, driving the $6S_{1/2} \rightarrow 7P_{3/2}$ (455.66 nm) and $6S_{1/2} \rightarrow 6P_{3/2}$ (852.347 31 nm) transitions, as light source. The background light came from the ceiling light illuminating the laboratory and consisted of Xe, Hg, Ar and Cr lines. Accurate values for the observed lines as a reference for the fit routine, the spectra are analyzed with, can be found on the NIST atomic spectra database[KYRa15]. As for the characterization



Figure A.1.1.: Calibration 'map' of the Shamrock grating spectrometer and the iDus camera from Andor showing the shift with its two degrees of freedom that are the horizontal pixel position on the CCD chip of the camera and the wavelength of incoming photons.



Table A.1.1.: Histogram of the calibration 'map' in figure A.1.1.

itself, spectra in the range from 300 nm to 1100 nm are recorded in 1 nm steps¹ and a 1 s exposure time five times at each wavelength. This enables the investigation on how accurate each pixel along the 1024 pixel axis (since the measurements are done in full vertical binning mode) on the CCD chip of the iDus camera is depending on the wavelength of arriving photons. For the following procedure, the mean spectrum, in which the before mentioned peaks are fitted with Gaussian functions, is then calculated for each spectra taken at specified center wavelength. From the Gaussian fits, the shift of peaks in respect to their actual position according to the literature values, can be calculated. The obtained shift is now dependent on the pixel position of the CCD chip of the iDus camera and the wavelength of incoming photons. For better visualization these shifts can be plotted in a 2D map, however for a full continuous visualization these values have to be interpolated first, of which the final result of such an analysis can be seen in figure A.1.1. Figure A.1.1 in return shows a histogram of the data from figure A.1.1. By fitting a normal distribution on to the histogram data, the mean shift and its error can be calculated, that in this case is $\overline{\Delta}_{\rm m} = 0.0395 \, {\rm nm} \pm 0.0692 \, {\rm nm}$, making it comparable with the $\pm 0.2 \, {\rm nm}$ wavelength accuracy from the specification sheet.

¹Depending on the grating, a different spectral range is available. The only way to measure a certain spectral area is to define the center wavelength the spectrum which is then recorded with in accordance to other specified parameters.

A.2. CW-Microwave experiments

Since the results in figure 4.1.3 shown in chapter 4.1.3 are only depicting the case at one distinct Rabi frequency for the probe laser, the same plots at the other Rabi frequencies are shown in the following.



Figure A.2.1.: Measurement series at $\Omega_{R,p} = 2\pi \times 2.11 \text{ MHz}.$



Figure A.2.2.: Measurement series at $\Omega_{\rm R,p} = 2\pi \times 2.58 \, {\rm MHz}$.



Figure A.2.3.: Measurement series at $\Omega_{\rm R,p}=2\pi\times 2.98\,\rm MHz.$



Figure A.2.4.: Measurement series at $\Omega_{\rm R,p}=2\pi\times 3.33\,\rm MHz.$



Figure A.2.5.: Measurement series at $\Omega_{\rm R,p}=2\pi\times 3.65\,\rm MHz.$



Figure A.2.6.: Measurement series at $\Omega_{\rm R,p}=2\pi\times$ 4.21 MHz.



Figure A.2.7.: Measurement series at $\Omega_{\rm R,p}=2\pi\times$ 4.47 MHz.



Figure A.2.8.: Measurement series at $\Omega_{\rm R,p}=2\pi\times$ 4.71 MHz.



Figure A.2.9.: Measurement series at $\Omega_{\rm R,p}=2\pi\times$ 4.94 MHz.



Figure A.2.10.: Measurement series at $\Omega_{\rm R,p}=2\pi\times5.16\,\rm MHz.$



Figure A.2.11.: Measurement series at $\Omega_{\rm R,p}=2\pi\times5.37\,\rm MHz.$



Figure A.2.12.: Measurement series at $\Omega_{\rm R,p}=2\pi\times5.57\,\rm MHz.$



Figure A.2.13.: Measurement series at $\Omega_{\rm R,p}=2\pi\times5.95\,\rm MHz.$



Figure A.2.14.: Measurement series at $\Omega_{\rm R,p}=2\pi\times$ 6.66 MHz.



Figure A.2.15.: Measurement series at $\Omega_{\rm R,p}=2\pi\times7.29\,\rm MHz.$



Figure A.2.16.: Measurement series at $\Omega_{\rm R,p}=2\pi\times 8.42\,\rm MHz.$

A.3. Collisional processes within the cesium vapor

Figures 4.2.5 and 4.2.6 from chapter 4.2.3 show only the case for excitation into the $50D_{5/2}$ state with the coupling laser at an reservoir temperature of $T_{\rm res} = 83 \,^{\circ}\text{C}$. However, measurements are done while exciting into the $30D_{5/2}$ Rydberg state and at an reservoir temperature of $T_{\rm res} = 92 \,^{\circ}\text{C}$. In the following the plots for these cases are presented.

A.3.1. Measurements at $T_{res} = 83 \,^{\circ}\text{C}$

The ground state density at this temperature has a value of $N = 2.9 \cdot 10^{18} \text{m}^{-3}$.

Table A.3.1.: Mean ionization cross sections σ of the $nD_{3/2}$, $nS_{1/2}$, $nF_{5/2}$ and $nP_{3/2}$ states through electron collisions while exciting the vapor to the $30D_{5/2}$ Rydberg state.

mean ionization cross sections $\overline{\sigma}$ (10 ⁻¹⁴ m ²) (30D _{5/2} , $T_{\sf res}$ = 83 °C)				
$\gamma(nD_{3/2}\to 6P_{1/2})$	0.23	$\gamma(nF_{5/2}\to 5D_{5/2})$	0.05	
$\gamma(nD_{3/2}\to 6P_{3/2})$	0.11	$\gamma(nF_{5/2} \to 5D_{3/2})$	0.20	
$\gamma(nS_{1/2}\to 6P_{3/2})$	5.00	$\gamma(nP_{3/2}\to 5D_{5/2})$	5.00	
$\gamma(nS_{1/2}\to 6P_{1/2})$	8.00	$\gamma(nP_{3/2}\to 5D_{3/2})$	5.00	



Figure A.3.1.: Excitation to the $30D_{5/2}$ Rydberg state at an reservoir temperature of $T_{res} = 83 \,^{\circ}\text{C}$. Shown are the decays of the $nD_{3/2}$ and $nS_{1/2}$ states together with the respective fluorescence spectrum.



Figure A.3.2.: Excitation to the 30D_{5/2} Rydberg state at an reservoir temperature of $T_{\rm res} = 83 \,^{\circ}$ C. Shown are the decays of the $nF_{5/2}$ and $nP_{3/2}$ states together with the respective fluorescence spectrum.

A.3.2. Measurements at $T_{res} = 92 \,^{\circ}C$

The ground state density at this temperature has a value of $N = 5.4 \cdot 10^{18} \text{m}^{-3}$.

Table A.3.2.: Mean ionization cross sections σ of the $nD_{3/2}$, $nS_{1/2}$, $nF_{5/2}$ and $nP_{3/2}$ states through electron collisions while exciting the vapor to the $30D_{5/2}$ Rydberg state.

mean ionization cross sections $\overline{\sigma}$ (10 ⁻¹⁴ m ²) (30D _{5/2} , $T_{\rm res} = 92^{\circ}$ C)			
$\gamma(nD_{3/2}\to 6P_{1/2})$	0.30	$\gamma(nF_{5/2}\to 5D_{5/2})$	0.11
$\gamma(nD_{3/2}\to 6P_{3/2})$	0.20	$\gamma(nF_{5/2}\to 5D_{3/2})$	0.14
$\gamma(nS_{1/2}\to 6P_{3/2})$	5.00	$\gamma(nP_{3/2}\to 5D_{5/2})$	5.00
$\gamma(nS_{1/2}\to 6P_{1/2})$	8.00	$\gamma(nP_{3/2}\to 5D_{3/2})$	5.00

Table A.3.3.: Mean ionization cross sections σ of the $nD_{3/2}$, $nS_{1/2}$, $nF_{5/2}$ and $nP_{3/2}$ states through electron collisions while exciting the vapor to the $50D_{5/2}$ Rydberg state.

mean ionization cross sections $\overline{\sigma}$ (10 ⁻¹⁴ m ²) (50D _{5/2} , $T_{\rm res}=$ 92 °C)				
$\gamma(nD_{3/2}\to 6P_{1/2})$	0.40	$\gamma(nF_{5/2}\to 5D_{5/2})$	0.25	
$\gamma(nD_{3/2}\to 6P_{3/2})$	0.30	$\gamma(nF_{5/2}\to 5D_{3/2})$	0.30	
$\gamma(nS_{1/2}\to 6P_{3/2})$	5.00	$\gamma(nP_{3/2} \to 5D_{5/2})$	5.00	
$\gamma(nS_{1/2}\to 6P_{1/2})$	8.00	$\gamma(nP_{3/2}\to 5D_{3/2})$	5.00	



Figure A.3.3.: Excitation to the $30D_{5/2}$ Rydberg state at an reservoir temperature of $T_{res} = 92 \degree C$. Shown are the decays of the $nD_{3/2}$ and $nS_{1/2}$ states together with the respective fluorescence spectrum.



Figure A.3.4.: Excitation to the $30D_{5/2}$ Rydberg state at an reservoir temperature of $T_{res} = 92 \degree C$. Shown are the decays of the $nF_{5/2}$ and $nP_{3/2}$ states together with the respective fluorescence spectrum.



Figure A.3.5.: Excitation to the 50D_{5/2} Rydberg state at an reservoir temperature of $T_{\rm res} = 92 \,^{\circ}$ C. Shown are the decays of the $nD_{3/2}$ and $nS_{1/2}$ states together with the respective fluorescence spectrum.



Figure A.3.6.: Excitation to the 50D_{5/2} Rydberg state at an reservoir temperature of $T_{\rm res} = 92 \,^{\circ}$ C. Shown are the decays of the $nF_{5/2}$ and $nP_{3/2}$ states together with the respective fluorescence spectrum.

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